COMMISSION OF THE EUROPEAN COMMUNITIES

PROCEEDINGS □ ACTES □ TAGUNGSBERICHTE

Seminar on

THE RADIOLOGICAL BURDEN OF MAN FROM NATURAL RADIOACTIVITY IN THE COUNTRIES OF THE EUROPEAN COMMUNITIES

Séminaire sur

LA CHARGE RADIOLOGIQUE DE L'HOMME LIEE A LA RADIOACTIVITE NATURELLE DANS LES PAYS DE LA COMMUNAUTE EUROPEENNE

Seminar über

DIE RADIOLOGISCHE BELASTUNG DES MENSCHEN DURCH NATÜRLICHE RADIOAKTIVITÄT IN DEN LÄNDERN DER EUROPÄISCHEN GEMEINSCHAFT

Le Vésinet □ PARIS □ 4-6 December 1979

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by the Directorate-General Employment and Social Affairs
Health and Safety Directorate

and the

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in collaboration with the

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P R O G R A M M E C O M M I T T E E

In drawing up the detailed Seminar Programme, the Commission has been able to call on the learned assistance of:

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F O R E W O R D

The Commission of the European Communities is coming to attach greater and greater importance to knowledge of the exposure of man to natural radiation. Experience of discharges of radioactive waste in the nuclear industry has in fact shown - for power stations in particular - that, generally speaking, the levels of exposure adhered to are at least two orders of magnitude lower than the maximum permissible levels laid down in the various legislations governing radiation protection. Moreover, the new recommendations of the International Commission on Radiological Protection and the directives proposed by the European Commission stress that the doses received by man must be as low as reasonably achievable, taking into account, of course, economic and social implications (optimization of radioprotection). Natural background radiation is used with increasing frequency as a criterion for evaluating the impact of a nuclear power station on the environment. Public awareness of all nuclear matters must not be neglected when making a detailed study of natural background radiation and its local and temporal variations. Levels may in fact vary considerably with altitude, ecological conditions (soil quality, food chains etc.), accomodation conditions (reduced ventilation, energy savings etc.) and living habits. On the other hand, some technologies may increase exposure to natural radiation (the use of certain building materials, production of phosphate fertilizers, coal-fired power stations, uranium extraction etc.). It must be remembered that the application of the concepts of group doses and collective doses to the population presupposes an in-depth knowledge of natural background radiation. For these reasons the Commission believes that the exposure of man to natural radiation should be studied and analysed as a matter of some urgency in the Member States of the Community in accordance with a programme drawn up on a joint basis.

The aim of the seminar organized jointly by the Directorate for Health and Safety (DG V) and the Biology, Radiation Protection and Medical Research Programme (DG XII) of the Commission is to compile existing

information, at the European level, to encourage those concerned to take all appropriate steps to quantify the various components of human exposure to natural radiation and to identify priority areas for study and research.

The many measures in hand or planned would make it possible in particular to examine the merit of undertaking epidemiological surveys which would involve a detailed statistical analysis of natural background radiation having regard to demographic factors.

With this in mind, the organizers are making these proceedings available to all interested parties.

F. VAN HOECK

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P R E F A C E

La Commission des Communautés Européennes accorde une importance croissante à la connaissance de l'exposition de l'homme à la radioactivité naturelle. En effet, la pratique des rejets, dans le nucléaire, a montré - notamment pour les centrales - que l'on respecte en général des niveaux d'exposition d'au moins deux ordres de grandeurs en deçà des valeurs maximales admissibles fixées dans les diverses législations sur la radioprotection. De plus, les nouvelles recommandations de la Commission Internationale de Protection radiologique et les propositions de directives de la Commission européenne mettent l'accent sur le fait que les doses reçues par l'homme doivent être aussi faibles que raisonnablement possible, tenant compte bien entendu des implications économiques et sociales (optimisation de la radioprotection). Aussi, se réfère-t-on de plus en plus au fond naturel de rayonnement comme critère de comparaison pour évaluer l'impact d'une centrale nucléaire sur l'environnement. Une prise de conscience du public en matière nucléaire ne peut être dissociée d'une étude précise du fond naturel de rayonnement et de ses variations locales et temporelles. En effet, les valeurs peuvent varier fortement selon l'altitude, les conditions écologiques (qualité du sol, chaîne alimentaire , etc.), les conditions d'habitat (réduction de la ventilation, économie d'énergie, etc.) et les conditions de vie. D'autre part, certaines technologies peuvent accroître l'exposition au rayonnement naturel (emploi de certains matériaux de construction, production d'engrais phosphatés, centrales thermiques à charbon, extraction de l'uranium, etc). Il est important de rappeler que l'application du concept de dose de groupe ou de dose collective à la population implique une connaissance approfondie du fond naturel de rayonnement. Aussi la Commission considère-t-elle que l'exposition naturelle de l'homme doit être d'une manière relativement urgente étudiée et analysée dans les Etats membres de la Communauté selon un programme défini en commun.

Le but du séminaire, organisé conjointement par la Direction Santé et Sécurité (DG V) et le Programme Biologie, Radioprotection et Recherche Médicale (DG XII) de la Commission est de parvenir à une synthèse au

niveau européen des informations existantes, d'inciter les milieux intéressés à effectuer des mesures là où cela semble souhaitable, afin de préciser les diverses composantes de l'exposition humaine à la radioactivité naturelle et de dégager des voies prioritaires d'études et de recherches.

Les nombreuses mesures en cours et envisagées permettraient notamment d'examiner l'utilité d'entreprendre des enquêtes épidémiologiques qui fassent appel à une analyse statistique précise du fond naturel de rayonnement eu égard à la situation démographique.

Dans cette perspective, les organisateurs mettent ces actes à la disposition de tous les milieux intéressés.

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V O R W O R T

Die Kommission der Europäischen Gemeinschaften misst der Kenntnis der Belastung des Menschen durch natürliche Radioaktivität eine zunehmende Bedeutung bei. Bei nuklearen Ableitungen, insbesondere aus Kernkraftwerken, werden - wie die Praxis zeigt - Expositionswerte eingehalten, die mindestens zwei Grössenordnungen unter den in den verschiedenen Strahlenschutzvorschriften festgelegten höchstzulässigen Werten liegen. Darüber hinaus wird in den neuen Empfehlungen der Internationalen Strahlenschutzkommission sowie in den Richtlinienvorschlägen der Europäischen Kommission der Schwerpunkt daraufgelegt, dass die von Menschen aufgenommenen Dosen unter Berücksichtigung wirtschaftlicher und sozialer Gesichtspunkte so niedrig wie möglich zu halten sind (Optimierung des Strahlenschutzes). Aus diesem Grunde geht man zunehmend dazu über, bei der Beurteilung der Auswirkungen eines Kernkraftwerkes auf die Umwelt die natürliche Grundstrahlung als Vergleichsbasis heranzuziehen. Ein Bewusstsein der Offentlichkeit in Sachen Kernenergie lässt sich nicht von einer vertieften Kenntnis der natürlichen Grundstrahlung und ihrer örtlichen und zeitlichen Schwankungen trennen. Die Pegelwerte können je nach Höhenlage, ökologischen Bedingungen (Bodenqualität, Nahrungskette usw.), Wohnverhältnissen (geringe Belüftung, Energiesparmassnahmen usw.) und Lebensbedingungen starken Schwankungen unterliegen. Zudem kann die Anwendung bestimmter Techniken (Verwendung gewisser Baustoffe, Herstellung phosphathaltiger Düngemittel, Nutzung kohlebefeuerter Kraftwerke, Urangewinnung usw.) die natürliche Strahlenbelastung der Bevölkerung erhöhen. Es ist wichtig, darauf hinzuweisen, dass das Konzept der Gruppen- bzw. Kollektivdosis eine genaue Kenntnis der natürlichen Grundstrahlung voraussetzt. Aus diesem Grunde ist die Kommission der Meinung, dass die natürliche Exposition des Menschen auf der Grundlage eines gemeinsam festgelegten Programms in den Mitgliedstaaten der Gemeinschaft mit einem gewissen Vorrang untersucht und analysiert werden sollte.

Ziel des gemeinsam von der Direktion Gesundheit und Sicherheit (GD V) und dem Programm Biologie, Strahlenschutz und medizinische Forschung (GD XII) der Kommission organisierten Seminars ist es, auf europäischer

Ebene einen Überblick über die vorhandenen Informationen zu gewinnen und die zuständigen Stellen zu veranlassen, an Orten, wo dies wünschenswert erscheint, Messungen vorzunehmen; auf diese Weise würden die verschiedenen Komponenten der natürlichen Radioaktivität, die zur Belastung des Menschen beitragen, bekannt und vorangig zu bearbeitende Untersuchungs- und Forschungsvorhaben aufgezeigt werden.

Die zahlreichen zur Zeit laufenden oder geplanten Messungen dürften es u.a. ermöglichen, den Nutzen epidemiologischer Erhebungen zu prüfen, für die bekanntlich eine genaue statistische Analyse der natürlichen Grundstrahlung unter Berücksichtigung der demographischen Gegebenheiten unerlässlich wäre.

Diese Erwägungen nicht zuletzt haben die Organisatoren veranlasst, diese Tagungsberichte allen interessierten Kreisen zur Verfügung zu stellen.

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C O N T E N T S

*) Data introduced from the floor

THE DETECTION OF BIOLOGICAL EFFECTS OF NATURAL RADIATION

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SUMMARY. Studies of human exposure to natural radiation are important for several reasons. Firstly for the indication that they may give about the possible biological hazard of such exposures, particularly when natural levels are high. Secondly, for suggestions of areas where epidemiological surveys might give direct evidence on the risks of radiation exposure at low dose rate, by comparing the frequency of cancer or genetic abnormality in areas of high and low natural radiation.

Positive results from any such survey would be of considerable importance in relation to radiation protection criteria for members of working or general populations exposed at comparable dose rates. Such results have hitherto been unobtainable owing to the lack of appropriate control populations, of adequate medical records, or of sufficiently large numbers exposed at the higher natural radiation rates.

The frequency, and even the occurrence, of harmful biological effects of natural radiation are therefore still largely a matter of inference, from frequencies observed at higher dose rates. Further work is needed on the best medical indices of harmful effect, the best ages for their detection, and the size of populations required for reliable detection of the effects of a given increase in natural radiation exposure.

RESUME. LA DETECTION DES EFFETS BIOLOGIQUES DES RAYONNEMENTS NATURELS. Les études sur l'exposition de l'homme aux rayonnements naturels sont importantes pour plusiers raisons: d'abord pour indiquer le danger biologique éventuel de ces expositions, en particulier si les niveaux naturels sont élevés, ensuite pour suggérer des zones où des enquêtes épidémiologiques pourraient mettre directement en évidence les risques d'une exposition aux rayonnements à faible débit de dose par la comparaison de la fréquence des cas de cancers ou d'anomalies génétiques dans les zones à rayonnement naturel élevé et faible. Les résultats positifs tirés de ces enquêtes revêtiraient une importance considérable pour définir des critères de protection contre les rayonnements ionisants, applicables aux travailleurs ou aux populations exposés à des débits de dose comparables.

Jusqu'ici on n'a pu obtenir de tels résultats, faute de populations témoins appropriées, de dossiers médicaux adéquats ou d'un nombre suffisant de personnes exposées aux plus hauts niveaux de rayonnement naturel.

Aussi, déduit-on encore largement la fréquence, voire l'occurence, d'effets biologiques nocifs dûs aux rayonnements naturels des fréquences observées à de plus hauts débits de dose. Il faudra d'autres travaux sur les meilleurs indices médicaux des effets nocifs, sur l'âge idéal pour leur détection et sur l'importance des populations à contrôler pour établir l'effet corrélatif à un accroissement de l'exposition aux rayonnements naturels.

KURZFASSUNG. NACHWEIS DER BIOLOGISCHEN AUSWIRKUNGEN DER NATURLICHEN STRAHLUNG. Untersuchungen über die Exposition des Menschen durch natürliche Strahlung sind aus mehreren Gründen von Bedeutung. Erstens, weil sie insbesondere bei hohen natürlichen Pegeln einen Hinweis auf das mögliche mit diesen Expositionen verbundene biologische Risiko geben können. Zweitens, weil sich auf diese Weise Gebiete eingrenzen lassen, auf denen epidemiologische Studien durch einen Vergleich der Häufigkeit von Krebserkrankungen oder genetischen Anomalien in Gebieten mit hoher und niedriger natürlicher Strahlung direkte Rückschlüsse auf die Gefahren der Strahlenexposition bei niedriger Dosisleistung zulassen können.
Positive Ergebnisse aus Studien dieser Art wären im Hinblick auf die Strahlenschutzkriterien für vergleichbaren Dosisleistungen ausgesetzte Arbeitnehmer und Mitglieder der übrigen Bevölkerung von grosser Bedeutung. Solche Ergebnisse liegen bisher nicht vor, weil geeignete Vergleichsgruppen der Bevölkerung und einschlägige ärztliche Aufzeichnungen fehlten oder weil die Anzahl der höheren natürlichen Strahlenpegeln ausgesetzten Personen nicht hinreichend gross war.
Die beobachtete Häufigkeit von schädlichen biologischen Auswirkungen der natürlichen Strahlenbelastung, und schon die Feststellung des Vorkommens selbst, basiert daher noch weitgehend auf Rückschlüssen aus den bei höheren Dosisleistungen gewonnenen Einsichten. Weitere Arbeiten über die zuverlässigsten medizinischen Anhaltspunkte für schädliche Auswirkungen, das vorteilhafteste Alter für ihre Feststellung un den Umfang der für einen zuverlässigen Nachweis der Auswirkungen einer gewissen Erhöhung der natürlichen Strahlenbelastung notwendigen Bevölkerungsgruppe sind erforderlich.

In this seminar we shall be discussing the largest source of radiation exposure of mankind worldwide: the source which, in most countries delivers at least two-thirds of the average genetically significant dose, diagnostic radiology being responsible for much the greatest part of the remainder[1]. We shall be reviewing the amount of this exposure under normal circumstances, and under circumstances in which some or all of these natural sources cause unusually high doses. And we will be examining the way in which such higher doses arise.

A survey of this kind is valuable for a number of reasons. Obviously it is important to know the amount and types of radiation to which mankind has always been exposed, and the extent of the natural variations from place to place in this amount, if only to afford some perspective on the frequency with which communities have for many generations been exposed to greater than typical amounts of radiation. Obviously also it could be of value to identify circumstances in which communities are at substantially greater exposure, and thus at potentially greater risk, than necessary - for example by choice of water supply or building material - where alternatives are easily available which involve a considerably lower exposure. In addition, however, a review of natural radiation exposure, and of conditions in which it is increased, could be of considerable value if it identified large communities which differed only in the level of radiation to which they were exposed, and in which the frequency of diseases or abnormalities could be compared and could give a direct estimate of whatever harmful effect radiation may have at low dose rate.

This would be particularly important, since the attempted estimation of low dose rate effects from small populations is so very vulnerable to the inevitable statistical uncertainities resulting from the use of small samples or from the presence of minor sources of bias. Even for working populations with typical average exposure rates of 5 mSv per year,[1] studies over very large numbers of man years are needed merely to detect, let alone estimate, increases of a few per cent in the normal cancer mortality (Table 1). And of course the amount of importance that we may attach to the size or the variation of natural background radiation rates, or to factors causing local increase in these rates, itself depends upon the amount of harm that exposure of populations at these rates is estimated to cause.

It is necessary therefore to consider the possibilities of estimating the frequency of any harmful radiation effects in man at normal or raised background exposure rates.

Before discussing the rather large size of populations that would be needed for this purpose, we must I think recognise certain other problems. It is of course important that the frequency of the effect studied, whether a disease or group of diseases or other abnormality, should be compared with that observed in a control population of the same cultural and dietary habits and with the same type of medical surveillance, but not equally exposed to radiation. It is desirable also that the comparison should be made over the same time period or allowing for trends with time. These requirements tend to exclude any useful studies of people living at high altitudes and exposed to raised cosmic radiation levels. Similarly an apparent correlation in fertility statistics with radiation levels in different parts of the monazite sand areas in Southern India proved to be due only to differences in cultural practices between the Christian, Hindu and Muslim communities which lived in the different parts of this coastal strip of land. It seems likely that difficulties in finding dietetically similar control groups might in the same way complicate any studies on populations with high levels, presumably of bone irradiation, resulting from raised concentration of bone seeking alpha emitters in the soil and hence in the diet; but this point needs review.

The efficiency with which harmful effects are ascertained in the exposed population must not only be equal to that in the control group; it must also be high. Where the comparisons are to be made by means of special surveys this may be achievable; and in the particular case of chromosome aberrations it can be and has been achieved with the accuracy required for demonstration of effects, for example in Finland[2], Brazil[3], and Bad Gastein[4]. The frequency of particular types of aberrations per unit radiation exposure has not, however, as much importance as the frequency of disease induced at low dose - at least until there is better understanding of the association between induced aberrations and induced disease. We must not forget the invaluable use of chromosome studies as a form of biological dosimetry, but I am not here primarily discussing the dosimetry of raised background areas.

The medical ascertainment of actual disease could, of course, usually be made as good as necessary by the expenditure of sufficient medical and technical effort. For some conditions which are readily identified, and which are persistent once they have developed, the comparison of frequencies could perhaps be made without unduly extensive surveys, if the surveys were acceptable to the communities involved. Studies of cataract or thyroid nodules could probably be undertaken in this way, although the distinction between malignant thyroid disease and benign and essentially harmless thyroid hyperplastic nodules - which are common locally in many areas, including the low background areas of Kerala, cannot in most cases be made by simple clinical examination. Down's syndrome might offer a similar opportunity, although an unequally effective ascertainment was clearly indicated [5, 6] as one of a number of defects in a study which at first suggested [7] an increase in this condition in the high background coastal areas of Kerala.

The low and unreliable ascertainment of many conditions, and particularly of all forms of cancer, has however constituted an overwhelming difficulty in the otherwise apparently ideal epidemiological situation in Kerala[8]. A relatively stable population of about 10,000 lives on a stretch of coast exposed to an average of 4 or 5 times the normal background radiation from the monazite sands on which the dwellings are built. Populations of generally similar habit and culture live in a similar way on other coastal areas which have normal radioactivity. And both areas are separated from the rest of the state inland by waterways which tend to limit movement and mixing of communities.

For both populations efficient medical and pathological services exist, at Trivandrum and Alleppey. A large proportion of people living in the coastal areas, however, are unlikely to have received medical advice which would, for example, identify the site or necessarily even the existence of malignancies. Also there is no required procedure for recording causes of death, so that any figures for disease incidence or mortality could at best be expected to refer only to disease or deaths occurring during the period of the survey - with a very large reduction in numbers for study as compared with the situations in which reliable records are available over a period of many years.

This problem of medical ascertainment, with diagnoses recorded in comparable pathological terminology and with preservation of records, is of course likely to affect many areas in which background radiation may be naturally or artificially increased, including perhaps many at high altitude or remote from conventional medical centres, or in countries without procedures for certification of the cause of death. The emphasis of this seminar, principally upon background variations within Europe, does at least focus upon situations in which medical records are likely to be maintained, and if so, maintained in consistent terminology according to the widely used international classification of disease. It is however a point which should perhaps be considered by the seminar and by the community - the extent to which the causes of death as recorded on death certificates are appropriately accessible to valid epidemiological research, or would in any countries be withheld on grounds of confidentiality.

There are thus a number of particular problems in basing quantitative studies of harmful effects of radiation upon variations in the exposure from natural background - problems of finding proper control populations, of recording comprehensively the frequency of effects and of avoiding bias in the efficiency of this recording in the exposed group. And there are problems also in estimating the doses actually received by the population in their daily life - (in Kerala dosemeters were incorporated in small ornaments of religious character which were given away free and worn on necklaces). Nor is the frequency of deaths from a given disease in a city's records necessarily to be associated with the background radiation rate in that city. For a short time it seemed significant that the death rate from leukaemia in Aberdeen - a city built of granite and built on granite - was higher than that in Edinburgh, in which the rocks are largely carboniferous and the background is largely normal. And indeed the difference in rates was significant, but the significance appeared to be due to the presence of a distinguished haematologist in Aberdeen to whose clinic patients with leukaemia came for treatment, but were ultimately recorded as having died of the disease in Aberdeen.

While therefore there are numerous difficulties in establishing proper conditions for a valid comparison of disease rates in different areas, by far the largest problem in practice is likely to be that of finding large enough populations exposed to high enough background levels to

show significant evidence of,for example,increased cancer induction. No way has yet been found of distinguishing a radiation induced cancer of any particular cell type, from a naturally occurring cancer of the same cell type. The evidence for radiation induction of cancers necessarily depends therefore on a numerical increase in frequency of such cancers, and so, critically, on statistical criteria for the significance of any increase observed. The detection of an increase may be very easy in the particular case of some sources of internal radiation exposure, if the sites of localisation or retention of a radionuclide give rise to conspicuous numbers of a normally rare type of tumour; and the bile duct tumours of the liver induced by thorotrast may perhaps be an extreme example of this sort. For external radiation however, causing more or less uniform whole body exposure, the problem would ordinarily be that of detecting a small number of radiation induced cancers, in the presence of much larger numbers of microscopically and clinically indistinguishable tumours that would occur normally in the exposed population.

Some idea of the size of populations required to obtain valid risk estimates from natural background exposure can be obtained by looking again at the purely statistical limitations (Table 2). For any given size of study, we need to compare the possible number of, for example, induced cancers with the number of cancers which would occur naturally during the study - since detectibility will depend upon this "signal to noise" comparison. The "noise" level - the total cancer incidence or mortality - will depend on the region considered, the age structure of the population and other factors, but will be proportional to the size of study - the size of population multiplied by the number of years over which it is studied. The size of "signal", the number of induced cancers, will be similarly proportional to the size of study - the number of person-years included. It will also be proportional to the excess radiation dose rate in the group studied, and to the rate with which tumours are induced per unit of this excess exposure.

For detectibility of the increased numbers in the presence of the very much larger numbers expected in normally exposed populations, the simple estimate by Poisson statistics again gives an adequate approximation. It is here assumed(Table 2):-

(a) that the "control" rate of occurrence is known without error, ie. from even larger studies on populations which are exposed only to normal background levels but are otherwise comparable. This will not necessarily be possible, so these data give minimum estimates of survey size.

(b) that induced cancers form a small percentage of all cancers observed, so that the term $I + KR$ is adequately approximated by I.

(c) that, with cancers constituting of the order of 20% of all causes of death, the Poisson estimate based on deaths recorded will approximate adequately to a more exact binomial estimate of error based on the proportion of such deaths.

The total cancer mortality for England and Wales was 2600 $10^{-6}y^{-1}$ in 1975 [9]. The fatal cancer induction rate (including leukaemia) is taken by ICRP [10] as about 12 $10^{-6}mSv^{-1}$. If this induction rate applies at raised background levels, its detection would thus require some millions of person years of study, even assuming full ascertainment of cancer deaths and a large and comparable control population. For example, for I = 2600 $10^{-6}y^{-1}$ and K = 12 $10^{-6}mSv^{-1}$, the excess will not be detectible (Table 2) unless the size of survey - the product PT - exceeds $72/R^2$. For regions with twice the normal background - ie. with an excess exposure of R = 1 $mSv.y^{-1}$, a survey of 72 million person years is thus required. For an average of say five times normal as in Kerala with R = 4, 4.5 million person years would be needed for detection of an increased cancer mortality.

So how can our efficiency of study be improved? Referring again to the approximate criterion for detectibility, that the size of study (PT) should exceed $4I/K^2R^2$, it appears that for any region of raised background (R), we should study the frequency of any event for which the value of I/K^2 was as low as possible. When the event studied is the occurrence of fatal cancers, of all types and at all ages, the values of I/K^2 are high.

Are we helped by examining one particular type of cancer, in which a higher than average induction rate might give an advantage in detection, which more than offset the reduction in numbers and hence the poorer statistical discrimination of signal to noise? The values for induction of individual fatal cancers are necessarily uncertain, but those derived from UNSCEAR[1] and ICRP[10], strongly suggest (Table 3) that detectibility would not be substantially improved by study of any one type of cancer mortality only. (It may be noted that the values of I/K^2 correspond to the person years required for detection in an area exposed at three times normal background levels.)

For total cancer incidence, the necessary person years of study seem likely to be smaller, although the practical difficulties of registering incidence are likely to be much greater in many regions than those for recording mortality, particularly for retrospective surveys. For many individual forms of cancer, the incidence is too nearly equal to the mortality to offer any advantage in surveying. The induction rate for all cancers (Table 4) is heavily influenced however by the high induction rates assumed for breast, with a mortality taken as 50%, and for thyroid, with a likely mortality of between 5 and 10% for radiation induced cancers. The prospects for detection by limited surveys appear to be optimal, on the data considered here, for thyroid cancers. Here again, however, as for incidence studies in general, the "Indeterminancy Principle" appears to come relentlessly into its own. The incidence of thyroid nodules depends critically upon how closely and expertly a population is examined; the malignancy rate in papable nodules cannot be reliably estimated without operation; and the normal incidence of both benign and malignant nodules commonly varies within wide limits from area to area; while such trivial factors as the fat content of the skin of the neck in different regions could substantially change the detectibility of small tumours - and small tumours of malignant histology occur frequently in some communities at post mortem examination, after causing no symptoms during life. This whole situation requires close numerical analysis, however, if surveys in high background areas are being considered.

The final discriminant of potential value is the age range over which it may be optimal to study any change in the normal occurrence of disease. Cancer incidence and mortality both increase steeply with age.

There might well therefore be advantages in confining the study to cancers occurring at younger ages, when the signal to noise ratio might be much higher than it is when all ages are taken together. It is not yet possible to answer this question reliably, however, for lack of necessary information on three points:-

a) the variation of induction rate with age at exposure, which is known for a few cancers only.

b) the latency, and the distribution of latencies, between exposure and detection of the cancer, which can be estimated for leukaemia only.

c) the variation of such latencies with age, where it is known that mean latencies are likely to be smaller, at least after exposure in utero than in adult life.

In the absence of such information, only very tentative estimates can be made of the advantage that might be gained. As an example, however, values of I in Fig. 1 give natural cancer incidence and mortality rates at different ages (as averages for males and females). The values of K estimate the frequency with which induced cancers would develop at different ages in a population exposed at constant dose rate, assuming an induction rate which was constant with age at exposure. The assumption is also made that leukaemia is induced, at all ages, with the distribution of latencies observed in Hiroshima and Nagasaki; and that, for all other cancers, latencies have twice these values.

On these obviously very oversimplified bases, the relationship I/K^2 between the rates of occurrence of cancer (per million at each age) is least at an age between 20 and 30. On these assumptions, an analysis based on the cancer mortality occurring in the age range 15 to 40 would allow detection of an increased mortality in about one-third of the time that would be required if the study was based on the cancer mortality in the whole population at all ages.

The question of whether there is an optimum age range in which to detect the number of induced tumours in excess of the naturally occurring ones, therefore appears to be worth examination [11]. This is of potential importance since, without some such method, the size of surveys that need to be made appears to be so large; and studies of populations exposed to raised background levels from various causes in Japan [12], the United States [13], Brittany [14], China [15] and Sweden [16] have involved difficulties for these and allied reasons.

It is important, however, that the optimal criteria for detection of any excess cancer, or other, incidence rates, be considered in the planning of any such studies. This is partly to guide the planning, or even to indicate the value of a study. It is important also however that if a wide range of data are collected at all ages, rigorous statistical criteria be applied to such comparisons as may appear to be significant, since, if the incidence or mortality of a variety of tumours is studied at a variety of ages in both sexes, the possibilities of fortuitous excess values in retrospectively chosen comparisons are obviously increased. Epidemiological studies in areas of high background will be of sufficient complexity without such additional complications.

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Table 1

Detectibility of fatal cancer induction rates in a male working population exposed at 5 mSv y^{-1}

Size of study (man years)	10^7	10^6	10^5	10^4	10^3
Numbers (N) of fatal cancers from "natural causes"	36,000	3600	360	36	3.6
Excess required for detectibility ($2\sqrt{N}$)	380	120	38	12	3.8
Induction rate required for detectibility (10^{-4} Sv^{-1})	76	240	760	2400	7600
Induction rate indicated [10] by epidemiological data (10^{-4} Sv^{-1})	125				

Note

Natural cancer mortality is that for males [9] of ages as for the UK working population. Detectible excesses are given approximately by Poisson statistics. Detectible induction rates are based on assumed equilibrium with average exposure [1] of 5 $mSv.y^{-1}$ giving a collective dose, eg. in 10^7 man years of 5.10^4 man sieverts.

Table 2

In a population of P million, recorded for T years

Expected number (eg. cancers) from "natural causes" = IPT
if natural rate is $I.10^{-6}.y^{-1}$

Excess due to increased radiation exposure = KRPT
if induction rate is $K.10^{-6}$ mSv^{-1}
and excess exposure is $R.mSv.y^{-1}$ (whole body)

So excess detectible if $KRPT > 2\sqrt{(I + KR)PT}$
or $PT > 4I/K^2R^2$ approx.

Table 3

Cancer mortality (all ages)		I	K	I/K^2
All forms	(M & F)	2600	12	18
Leukaemia	(M & F, excl. CLL)	51	2	13
Lung	(M & F)	680	2	170
Thyroid	(F)	12	0.7	25
Breast	(F)	470	5	19

Note

For leukaemia, chronic lymphatic leukaemia is excluded since it has not been shown to be radiation induced. For the thyroid, the induction rate in females is taken to be 2.3 times that in males [1].

Table 4

Cancer incidence (all ages)[17]	I	K	I/K^2
All forms (M & F, excl. skin)	3020	30	3.4
Thyroid (F)	20	10	0.2
Salivary (M & F)	14	2	3.5
Breast (F)	770	10	7.7

Note Skin cancers are excluded owing to the considerable difficulties in obtaining reliable incidence figures. For the thyroid, the (rounded) value of K correspondends to a 7% mortality of radiation induced thyroid cancers in females. The corresponding value for the breast assumes a 50% mortality. For the salivary glands, the value of I includes mixed salivary tumours, which are not distinguished in UK registry data, and the value of K is derived from UNSCEAR[1].

Fig. 1.

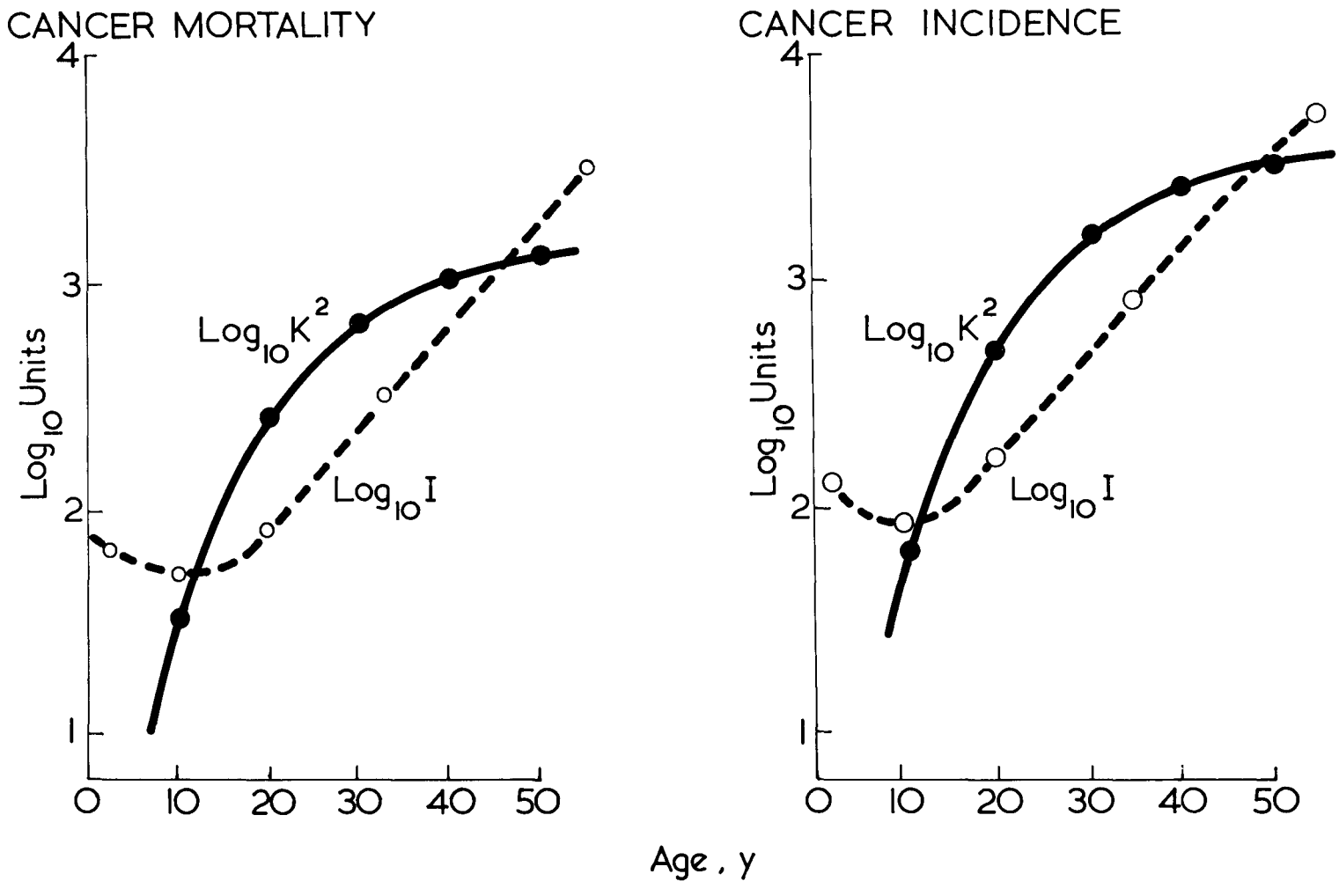
Values of I ($10^{-6}y^{-1}$) are average rates for males and females

(a) for cancer mortality, in England and Wales, 1975[9]

(b) for cancer incidence, for an English tumour registry in a population of 9 million (S. London Metropolitan region) for 1967/71[17]

Values of K are based upon induction rates [1, 10] for a population assumed to be exposed at 3 times normal background (3 mSv y^{-1}).

Minimum values of ($\log I - \log K^2$) occur at about age 25.


CANCER MORTALITY
CANCER INCIDENCE
Log10 Units
Log10 K2
Log10 I
4
3
2
1
0
10
20
30
40
50
Age, y

EXPOSITION AUX RAYONNEMENTS IONISANTS D'ORIGINE NATURELLE. TECHNIQUE DE MESURE - REVUE DES DONNEES EUROPEENNES - MESURES AUX FRONTIERES

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Paris - FRANCE

RESUME. L'auteur expose tout d'abord les difficultés qui apparaissent lorsqu'il s'agit de relier des mesures ponctuelles, aussi précises soient-elles, à l'exposition réelle des populations. Il faut en effet, tenir compte des importantes variations du champ de rayonnement naturel dans l'espace et dans le temps.

Après avoir passé en revue les données disponibles dans les différents pays de la Communauté et étudié les techniques de mesure utilisées, on expose en détail les méthodes adoptées dans le cadre de l'étude de l'irradiation naturelle en France.

Ces méthodes sont les suivantes:

- mesures en intégration pendant six mois avec des dosimètres thermoluminescents,
- mesures de débit avec un radiamètre à scintillateur plastique sélectionné après comparaison avec les radiamètres du même type existant actuellement.

Enfin, le rapport décrit une campagne de mesures réalisée à des fins d'intercomparaison dans les différents pays limitrophes de la République Fédérale Allemande du 11 au 22 juin 1979.

KURZFASSUNG. NATÜRLICHE BESTRAHLUNG VON AUSSEN - BENUTZTE MESSTECHNIK ÜBERBLICK ÜBER DIE EUROPÄISCHEN DATEN UND MESSUNGEN IN GRENZGEBIETEN.

Zunächst werden die Schwierigkeiten abgehandelt, die auftreten, wenn Messungen an einem Aufpunkt - so genau sie auch sein mögen - Aufschluss über die tatsächliche Strahlenbelastung der Bevölkerung geben sollen.

In der Tat müssen hierzu starke räumliche und zeitliche Schwankungen des natürlichen Strahlungsfeldes berücksichtigt werden.

Nach einem Überblick über die in den einzelnen Ländern der Gemeinschaft gewonnenen Daten und über die verwendete Messtechnik werden im einzelnen die bei der Untersuchung über natürliche Strahlung in Frankreich herangezogenen Methoden erläutert.

Es handelt sich dabei um:

- Integrationsmessungen, die über sechs Monate mit Thermolumineszenzdosimetern durchgeführt werden;
- Messungen der Dosisleistung mit einem Strahlungsmessgerät mit Kunststoffszintillator, das nach einem Vergleich mit den zur Zeit vorhandenen Strahlungsmessgeräten gleicher Bauart ausgewählt wurde.

Abschliessend wird eine Messkampagne, die zu Vergleichszwecken in den einzelnen an die Bundesrepublik Deutschland angrenzenden Ländern vom 11. bis 22. Juni 1979 durchgeführt wurde, beschrieben.

SUMMARY. EXPOSURE TO NATURALLY-OCCURRING IONISING RADIATION, MEASUREMENT TECHNIQUES, A REVIEW OF EUROPEAN DATA AND MEASUREMENTS AT FRONTIERS. The author first presents the difficulties which arise when one tries to derive the true population exposure from measurements at individual points, no matter how precise. It is in fact necessary to take into account the significant variations which occur in the radiation field in space and time. After a review of the available data in the different Community countries and a study of the techniques used, the author presents in detail the methods adopted within the framework of the study of external irradiation in France.

These methods are as follows:

- integrated measurements over six months using thermoluminescent dosemeters,
- dose-rate measurements with a plastic scintillator selected after comparison with other similar instruments available.

The author then describes the results of a field study carried out from 11th-22nd June 1979 for intercomparison purposes in the frontier areas of countries neighbouring the German Federal Republic.

Dans le cadre d'un contrat avec la Commission des Communautés Européennes le Service Central de Protection contre les Rayonnements Ionisants (SCPRI) a été chargé de faire le bilan de toutes les données européennes concernant les niveaux de l'exposition des populations aux rayonnements ionisants d'origine naturelle et d'harmoniser ces données afin d'obtenir une carte européenne cohérente.

Il importe de considérer avec réalisme le but qui peut être atteint; en premier lieu nous poserons donc la question de la signification que l'on peut accorder à une carte de l'exposition.

Ensuite nous décrirons les techniques de mesure que nous avons sélectionnées.

Nous donnerons une brève revue des données européennes disponibles qu'il nous a été demandé d'harmoniser.

Enfin nous présenterons les résultats des mesures qui ont été effectuées aux frontières pour homogénéiser les résultats obtenus dans les différents pays.

SIGNIFICATION D'UNE CARTE DE L'EXPOSITION

En matière d'exposition d'origine externe aux rayonnements ionisants naturels, un certain nombre de faits sont actuellement établis :

- en moyenne l'exposition annuelle est de l'ordre de 100 millirads par an,
- les variations autour de cette valeur moyenne revêtent une amplitude notable (couramment, de 80 à 250 millirads par an),
- les variations à grande échelle sont essentiellement fonction d'une part de la nature géologique des sols, qui conditionne l'exposition au rayonnement tellurique, d'autre part de la latitude et de l'altitude pour ce qui concerne l'exposition au rayonnement cosmique.

La recherche du détail, dans ce domaine, conduit rapidement à des difficultés de mesure et d'interprétation; compte tenu de l'intérêt réduit que l'on pourrait en attendre, il n'apparaît pas raisonnable de poursuivre ces investigations au-delà de la confirmation de ces faits.

En dehors, en effet, de très rares régions du globe où l'exposition effective de populations statistiquement significative dépasse un rad par an, on ne peut guère espérer tirer des conclusions d'éventuelles enquêtes épidémiologiques de quelque nature qu'elles soient; la variabilité des doses individuelles, le taux important d'apparition spontanée des affections en cause, la faible incidence de l'exposition aux rayonnements ionisants à des doses de l'ordre de 100 millirads par an, la difficulté de séparer clairement les paramètres et de disposer de groupes témoins, rendent fort aléatoires et discutables les tentatives dans ce sens.

Il convient par conséquent de considérer avec la plus grande prudence toute carte présentant des résultats d'exposition aux rayonnements d'origine externe naturelle. En effet, aux variations régionales liées à la nature géologique du sol, se superposent, surtout en milieu habité (et c'est celui qui nous intéresse), des variations locales dues aux revêtements du sol, aux cultures, etc.

L'exposition dans une agglomération est incontestablement différente de l'exposition en rase campagne, mais au sein même de l'agglomération, il existe aussi d'importantes variations d'un quartier à l'autre et, pour un même quartier, de non moins importantes variations d'une rue, ou d'un immeuble, à l'autre.

Si l'on considère de telles variations qui revêtent des amplitudes analogues à l'échelle continentale, régionale et locale, elles évoquent immédiatement les courbes aux propriétés apparemment paradoxales que les mathématiciens nomment fractals : de telles courbes correspondent, dans la nature, par exemple au profil des côtes ou des chaînes de montagnes, dont on sait cependant que la longueur précise n'est pratiquement pas mesurable.

Il convient donc de savoir ne pas considérer les cartes de l'exposition naturelle au-delà de ce qu'elles doivent modestement rester : la représentation approximative d'une réalité très complexe. Y chercher le détail serait donc illusoire, et d'ailleurs, de toutes manières, inutile.

TECHNIQUE DE MESURE

Le choix d'une technique de mesure de l'exposition naturelle externe doit tenir compte de deux difficultés :

- l'une d'ordre technique : le détecteur doit avoir une réponse suffisamment indépendante de l'énergie. Sa sensibilité doit être suffisante pour apprécier, avec une bonne précision, une fraction de microrad par heure ou, en intégration, un millirad pour un temps d'exposition de six mois. Il doit pouvoir être mis en oeuvre sur place et pas seulement dans les conditions très particulières du laboratoire,
- l'autre concernant la représentativité des mesures : le choix du quadrillage est, à cet égard, capital.

Nous avons retenu deux modalités de mesure, l'une du débit, l'autre de la dose intégrale, qui s'avèrent complémentaires.

Dosimètres intégrateurs

Le dosimètre intégrateur utilisé par le SCPRI pour la mesure de la dose due au rayonnement naturel comprend :

- un dosimètre photographique classique, avec son étui porte-filtres, analogue au dosimètre utilisé pour la surveillance individuelle des travailleurs,
- deux dosimètres thermoluminescents logés dans une gaine de vinyle thermosoudée et placée au dos du film photographique.

L'ensemble, dosimètre photographique et dosimètres thermoluminescents, est placé dans une pochette en vinyle soudée hermétiquement de façon à éviter toute altération par l'humidité.

Le dosimètre photographique ne sert pas directement à la mesure du rayonnement ambiant, mais ce film permet de signaler éventuellement des conditions anormales d'utilisation telle qu'une irradiation intempestive par un rayonnement de basse énergie ou une contamination radioactive.

Les dosimètres thermoluminescents associés comprennent un fritté de fluorure de lithium (LiF) de 4,5 mm de diamètre et un cristal de fluorure de calcium (CaF2) activé au dysprosium de 6 mm x 6 mm. Sur une période de six mois, la dose intégrée est significative (entre 30 et 150 mrad) avec les deux types de dosimètres.

Les résultats concernant 865 points de mesure à l'extérieur des habitations et 946 à l'intérieur sont présentés dans un autre exposé (1).

Mesures de débits de dose avec un radiamètre à scintillateur plastique

Les mesures effectuées avec des dosimètres intégrateurs peuvent être utilement complétées par des mesures de débit de dose. Le radiamètre permet en effet une exploration plus complète d'un site en écartant les valeurs non représentatives alors que le dosimètre intégrateur peut avoir été placé par hasard en un point non significatif, dans un champ de rayonnement anormalement bas ou élevé en raison d'une hétérogénéité quelconque de ce champ.

Parmi les différents modèles existant actuellement nous avons retenu le radiamètre à scintillateur plastique utilisé dans le cadre d'une vaste opération réalisée en RFA pour le compte du Ministère Fédéral de l'Intérieur(2).

Cet appareil est particulièrement adapté aux mesures de très faibles niveaux d'irradiation et son scintillateur possède une composition atomique assez proche de celle des tissus biologiques.

Il mesure de façon très précise la composante de l'irradiation naturelle d'origine tellurique; par contre la composante cosmique ne produit qu'un bruit de fond assez difficile à évaluer. Nous avons effectué à cet égard des mesures sur un lac de la région parisienne qui nous ont conduits à retenir une contribution du rayonnement cosmique équivalente à 1 µR/h. C'est cette valeur qui a été également adoptée par PFISTER, PHILIPS et PAULY utilisant le même type d'appareil (3).

Comparaison des résultats obtenus avec les dosimètres thermoluminescents et le radiamètre à scintillateur plastique

Afin de vérifier l'équivalence des résultats obtenus avec les deux techniques de mesure nous avons organisé une mission dans le département de l'Yonne dans lequel des thermoluminescents (TLD) avaient été placés en 25 points chez des particuliers. Dans chaque cas une mesure a été effectuée au même endroit avec le radiamètre.

La figure 1 montre la distribution des rapports entre les valeurs données par les deux systèmes. L'accord est très satisfaisant malgré des valeurs extrêmes de 0,55 et 1,45 qui ne sont pas aberrantes si l'on tient compte du fait que les deux mesures n'ont pas été effectuées dans des conditions absolument équivalentes en raison des variations du champ de rayonnement dans le temps et dans l'espace.

REVUE DES DONNEES EUROPEENNES

Le Tableau I est un résumé de toutes les données disponibles dans les pays de la Communauté. Il apparaît que tous les pays ne sont pas intéressés de la même façon à l'étude de l'exposition des populations aux rayonnements d'origine naturelle.

On note également que les méthodes utilisées sont très diverses, ce qui ne va pas sans provoquer des difficultés de compatibilité entre les résultats des différents pays.

C'est dans le but d'harmoniser les résultats des pays de la Communauté que le SCPRI a été chargé, dans le cadre d'un contrat CCE, de sélectionner la ou les méthodes de mesure les plus appropriées.

MESURES DE L'EXPOSITION NATURELLE AUX FRONTIERES

Le SCPRI a organisé à deux reprises en juin 1978 et juin 1979 une campagne de mesures aux frontières des différents pays limitrophes de la RFA, ce dernier pays pouvant servir de référence compte tenu du travail important qu'il a réalisé dans le domaine de l'irradiation naturelle.

Ces campagnes ont été bien entendu réalisées en coopération étroite avec les autorités des différents pays et les laboratoires concernés, que nous tenons tout spécialement à remercier, car c'est à leur aide, à leur compréhension, à leur efficacité, et à une indéniable conscience européenne partout ressentie, qu'en est dû le succès.

Organisation et déroulement des campagnes de mesures

Une première mission a été organisée en juin 1978, après accord préalable avec les pays concernés. Deux agents du SCPRI se sont rendus en RFA, Belgique et Luxembourg du 5 au 9 juin 1978 pour effectuer un certain nombre de mesures et surtout faire l'étude des difficultés éventuelles pouvant apparaître dans une telle mission, que ce soit sur le plan technique ou sur le plan administratif.

Le dispositif expérimental comprenait deux radiamètres dont l'un était couplé à un enregistreur et fonctionnait en permanence à bord du véhicule. L'autre radiamètre était utilisé pour des mesures plus significatives sur le terrain. Nous avons défini une unité de lieu que l'on peut dénommer "site". En chaque site, cinq mesures au moins ont été réalisées dans un rayon de 20 mètres environ; les valeurs relevées étaient portées sur une fiche spéciale (fig.2) sur laquelle étaient indiquées par ailleurs toutes les observations possibles sur les conditions météorologiques et la nature du terrain. Les mesures étaient effectuées en dirigeant le cristal vers le sol à 1 mètre de hauteur. Les sites étaient choisis de préférence loin des agglomérations pour éviter la contribution des matériaux de construction qui est extrêmement variable et dont l'étude ne constituait pas l'objet de notre travail.

Plus de 60 mesures dans 11 sites ont été réalisées au Luxembourg et plus de 100 mesures dans les 20 sites répartis sur les frontières franco-belge et franco-allemande.

Cette première mission ayant donné des résultats encourageants, une seconde plus importante a été organisée dans les mêmes conditions d'accords préalables du 11 au 22 juin 1979 aux frontières de la RFA avec la Belgique, les Pays-Bas et le Danemark.

Le dispositif technique et le mode opératoire étaient identiques à ceux de la première mission. Plus de 500 mesures ont été effectuées dans une centaine de sites répartis sur un itinéraire de 3500 km.

Résultats

Les résultats des deux campagnes de mesures sont réunis dans les Tableaux II à VI. Chaque site est identifié par le nom de la localité la plus proche, les valeurs minimale, maximale et moyenne sont exprimées en microrad/heure et les renseignements complémentaires concernant la nature du terrain sont notés en observation. Les valeurs moyennes sont également portées sur la figure 3 qui donne une vue d'ensemble de tous les résultats enregistrés lors des deux missions. La figure 4 montre plus en détail les valeurs relevées au Luxembourg.

Dans ces conditions, des mesures effectuées à l'air libre et le plus souvent loin des agglomérations donnent des résultats qui sont fonction essentiellement de la nature géologique des terrains.

On note une grande zone comprise entre Hambourg et Dortmund constituée par des terrains quaternaires de dépôts glaciaires et dans laquelle les doses enregistrées sont les plus basses (moins de 3 µrad/h). Au nord de Hambourg et jusqu'à la frontière danoise les doses sont légèrement plus élevées sauf dans une région de marais située près de la frontière côté allemand. De Düsseldorf à la frontière française les débits de dose augmentent jusqu'à 8 et 9 µrad/h sur des terrains primaire et triasique.

Toutes les valeurs observées sont en bon accord avec celles données dans le rapport du Ministère Fédéral de l'Intérieur allemand (2). Si dans quelques cas il n'y a pas identité parfaite, cela provient de la très grande hétérogénéité locale du champ de rayonnement.

Enfin la figure 5 montre un extrait de l'enregistrement continu effectué sur le véhicule. On vérifie le fait bien connu (13) que lors des traversées d'agglomérations le débit de dose croît toujours de façon très importante. De même à l'intérieur d'une même localité l'irradiation externe varie considérablement selon les matériaux des habitations ou le revêtement de la chaussée.

DISCUSSION ET CONCLUSION

Les campagnes de mesure effectuées aux frontières ont montré un accord satisfaisant avec les mesures réalisées par la RFA. Il devient donc possible, sur cette base, d'envisager d'harmoniser les valeurs relevées dans les pays frontaliers.

Le SCPRI se propose donc, dans le courant de l'année 1980, de reprendre la totalité des valeurs disponibles dans les différents pays à la lumière de ces résultats, ainsi que les mesures qu'ils ont effectuées entre temps. Le but est l'obtention d'une carte européenne de l'exposition externe naturelle.

Cependant, comme nous l'avons précisé dans notre rapport "Synthèse des informations existantes sur l'irradiation externe liée à la radioactivité naturelle en Europe" (4), et rappelé en introduction de la présente publication, nous insistons une fois encore sur le fait que cette carte ne saurait traduire plus qu'un aperçu général et très synthétique des niveaux d'exposition moyens.

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Tableau I : Bilan des principales études réalisées dans les pays de la Communauté Européenne.

Pays	Auteurs	Champ d'application	Méthode utilisée
BELGIQUE (5)	Institut d'Hygiène et d'Epidémiologie	- Recherche de gisements uranifères	- Radiamètre à scintillateur NaI
DANEMARK (6)	Centre de Recherche de RISØ de la Commission de l'Energie Atomique	- Evolution de l'irradiation externe dans 10 fermes modèles	- Spectrométrie NaI
		- Mesures sur tout le territoire à l'extérieur des habitations	- Chambre d'ionisation à haute pression
FRANCE (1), (7)	Service Central de Protection contre les Rayonnements Ionisants (SCPRI)	- Extérieur et intérieur des habitations dans tout le pays chez des particuliers	- Dosimètres thermoluminescents
		- Mesures localisées de débits de dose à l'extérieur des habitations	- Radiamètre à scintillateur plastique
	Commissariat à l'Energie Atomique	- Mesures à l'intérieur des habitations chez des particuliers dans 6 départements	- Dosimètres thermoluminescents
IRLANDE (8)	McAULAY I.R. Trinity College Dublin	- Mesure du rayonnement tellurique à l'extérieur des habitations en 300 points du territoire	- Chambre d'ionisation à haute pression
ITALIE (9), (10)	Comitato Nazionale per l'Energie Nucleare (CNEN)	- Mesure du rayonnement tellurique à l'extérieur des habitations dans tout le territoire et à l'intérieur des habitations dans ROME	- Chambre d'ionisation à haute pression
		- Mesure du rayonnement cosmique sur des lacs	- Spectrométrie γ
LUXEMBOURG	SCPRI	- Mesures à l'extérieur des habitations	- Radiamètre à scintillateur plastique
PAYS - BAS (11)	Laboratoire de Recherches sur les Rayonnements - Institut National de la Santé Publique	- Mesure de l'irradiation à l'extérieur des habitations	- Scintillateur plastique
REPUBLIQUE FEDERALE ALLEMANDE (2)	Ministère fédéral de l'Intérieur	- Mesures en 25000 points à l'extérieur des habitations et 30000 points à l'intérieur des habitations	- Radiamètre à scintillateur plastique
ROYAUME UNI (12)	National Radiological Protection Board (NRPB)	- Mesure de l'irradiation à l'intérieur des habitations	

Tableau II : Irradiation d'origine tellurique (µrad/h)

LUXEMBOURG

Site	Minimum	Maximum	Moyenne	Observations
REMICH	7,7	8,2	8,0	pelouse de l'église
MOUTFORT	3,0	4,5	3,6	bois de hêtres
ROLLINGEN	4,8	5,1	4,9	prairie
Croisement route CLERVAUX-HOSCHEID	7,8	8,5	8,3	champ sur terrain schisteux
CLERVAUX(Abbaye)	7,0	9,4	7,6	
DRAUFELT	7,5	7,7	7,6	champ
WILTZ	6,5	8,2	7,4	parkings sortie sud (schistes)
ESCHE/SURE	8,0	11,0	9,0	prairie sur fond de vallée à proximité roches affleurantes
SCHWEICH	2,7	3,0	2,8	bois sur sol grèseux
STEINFORT	4,1	6,0	4,8	jardin sur sol sablonneux

Tableau III: Irradiation d'origine tellurique (prad/h)

DANEMARK

Site	Minimum	Maximum	Moyenne	Observations
ST JYNDEVAD	1,9	2,1	2,0	sable gris glaciaire
GALLEHUS	2,8	3,0	2,9	prairie artificielle
RUSBOLD/HØJER	3,5	4,2	3,9	" "
KLIPLEV	2,0	2,6	2,3	" "
ULLERUP	4,2	4,6	4,5	" "
SØNDERBØRG	3,7	4,1	3,9	" "
ABENRA	3,6	4,2	3,9	dépôts glaciaires grossiers
EMMERSKEDE	2,3	3,1	2,8	prairie

Tableau IV : Irradiation d'origine tellurique (µrad/h)

PAYS - BAS

Site	Minimum	Maximum	Moyenne	Observations
NIEUW-BEERTA	4,7	4,9	4,8	prairie artificielle terre lourde
WEDDE	1,8	2,1	1,9	champ
JIPSINGHUIZEN	1,5	2,0	1,7	jardin sablonneux
NIEUW-WEERDINGE	1,4	1,6	1,4	champ sablonneux
ERM	1,7	1,9	1,8	prairie
COEVORDEN	1,5	1,6	1,5	prairie artificielle sablonneuse
MARIENBERG	1,5	1,8	1,6	prairie sur sol sablonneux
ALMELO	1,5	2,3	1,8	prairie
BECKUM/HAAKSBERGEN	1,7	2,0	1,9	champ sablonneux
GROENLO	1,8	2,5	2,2	prairie artificielle
HARREVELD	2,4	2,6	2,5	prairie
ZEDDAM	3,8	5,7	4,6	prairie argileuse
VELP	3,0	3,1	3,1	champ de seigle sur sol argileux
NIMEJE	2,6	3,0	2,8	prairie
AFFERDEN	1,7	2,2	1,9	champ sablonneux
DE HAMERT	1,3	1,8	1,6	champ et bosquet sablonneux
BELPELD	2,1	2,5	2,3	prairie sur sol sablonneux
ST JOOST	2,5	2,8	2,7	champ
BEEK	4,2	5,7	5,2	prairie
BREUST	5,2	5,8	5,5	prairie artificielle

Tableau V : Irradiation d'origine tellurique (µrad/h)

BELGIQUE

Site	Minimum	Maximum	Moyenne	Observations
ARLON	3,6	4,0	3,8	champ cultivé
GEROUVILLE	4,0	5,2	4,5	prairie naturelle
LAMBERTEAU	4,2	4,9	4,7	" "
5 km Est BOUILLON	5,5	6,6	6,1	bois sapins sur séricito-schistes
BOUILLON	7,6	8,0	7,5	place du château
MENUCHENET	4,7	5,0	4,8	bois
BIEVRE	5,1	6,6	5,8	prairie
SCOTTON	4,0	4,8	4,4	bois de hêtres
Nord de WELLIN	5,4	6,0	5,8	bois de chênes sur marno-calcaire
CELLES	3,0	4,5	3,5	jardin sur calcaire dinantien
HASTIERES	3,4	5,4	4,5	jardin bord de Meuse
LIEGE	7,0	10,0	8,5	place de la gare Guillemin
GOMZE	6,3	7,1	6,7	prairie argileuse sur schistes
TIEGE/SART	4,6	4,8	4,7	prairie
STAVELOT	5,4	5,6	5,5	"
SALMCHATEAU	5,0	5,5	5,2	"
HOUFFALIZE	5,3	6,0	5,6	"
BASTOGNE	4,3	4,9	4,5	champ
NEUFCHATEAU	5,4	6,0	5,8	prairie (terrain schisteux)
FLORENVILLE	3,7	4,3	4,0	prairie
BOUILLON	5,3	6,0	5,6	micaschistes

Tableau VI : Irradiation d'origine tellurique (µrad/h)

ALLEMAGNE

Site	Minimum	Maximum	Moyenne	Observations
PERL	7,1	7,2	7,2	prairie artificielle
NITTEL	6,1	7,8	6,7	argile de bas de pent grès marno-calcaire
N 51 (20 km N Trier)	5,4	6,7	6,3	champ de blé
SEFFERN	5,1	7,8	6,5	jardin public
HEMESPAND-DENSFELD	4,9	5,4	5,0	bois de sapins sur schistes grèseux
STADTSKILL	5,4	7,2	6,0	champ de blé sur grès schisteux bariolé
N 285 carrefour de SCHMITHEIM	3,7	4,5	4,0	bois de sapins
NIETERSFELD	6,2	6,8	6,6	champ de blé
HEIMSBACH	5,2	6,5	6,3	bois de sapins sur schistes
NIDDEGEN	4,5	5,9	5,0	champ cultivé
DÜREN	3,6	4,9	4,7	jardin public
JÜLICH	3,9	5,9	5,5	champ sablonneux
ELFGEN	3,9	5,5	5,0	champ riche en humus
NORF	4,5	5,2	4,8	champ sablonneux
GRIMLINGHAUSERN	4,9	5,0	4,9	jardin sur alluvions du Rhin
DÜSSELDORF (Gurlitt Strasse)	4,0	4,3	4,1	sur gazon
OSTERATH	4,5	5,5	4,9	jardin public
VINNSBRUCK	3,0	5,7	3,9	prairie et bosquet
N 58 carrefour ISSUM/WESEL	4,5	4,8	4,7	champ cultivé
BUDERICH	4,0	5,4	4,8	prairie artificielle
RAESFELD	2,5	3,3	2,7	champ cultivé sablonneux
WESEKE	2,0	2,7	2,3	champ
NIENBORG	2,6	3,0	2,7	champ de seigle
BENTHEIM	2,1	2,2	2,1	prairie humide
NORDHORN	1,4	2,0	1,7	champ
LINGEN/EMS	1,8	2,3	2,2	gazon sur sol sablonneux
MEPPEN	2,3	2,6	2,4	forêt de sapins sur sol sablonneux

Tableau VI (suite 1)

Site	Minimum	Maximum	Moyenne	Observations
KLUSE	2,2	2,5	2,3	landes sur sol sablonneux (Heide)
STEENFELDE	2,0	2,5	2,3	prairie artificielle
SCHWERISDORF	2,2	2,4	2,3	landes sur terrains sablonneux
LINSWEGE	1,6	1,8	1,7	champ cultivé sur marais à tourbière
JADE	1,6	3,8	2,6	prairie artificielle sur marais à tourbière
SANDSTEBE	1,9	2,5	2,1	sol dunaire
STUBBEN	2,6	3,0	2,8	champ humifère et sablonneux
GLINDE	1,4	1,8	1,6	prairie artificielle sur marais à tourbière et sablonneux
OSTEN	3,3	4,1	3,8	prairie artificielle
BELTING	3,7	4,2	4,0	bocage
TERKELSKOFT	3,9	4,0	4,0	champ cultivé
GLUCKSTADT	2,6	3,8	2,9	prairie bord de mer
WIMMERSBULL	2,9	3,2	3,0	prairie artificielle
RODENAS	3,7	3,9	3,8	" "
NIEBULL	1,5	1,9	1,8	prairie humide sur marais sablonneux
LADELUND	1,6	1,9	1,8	sable gris glaciaire
OSTERBY	1,9	2,2	2,0	prairie sur sol sablonneux
SANKELMARK	3,2	4,2	3,7	prairie
GETTORF	3,4	3,7	3,6	forêt de trembles
PRETZ	3,1	3,8	3,4	forêt sur sol sablonneux
RATZBEK	3,7	5,1	5,0	champ de blé
HAMBURG-MAMMSTORF	3,2	3,7	3,4	sol forestier sablonneux
SCHESSEL	1,2	1,6	1,4	forêt de pins et chênes sur sol sablonneux
SAARBRUCK	9,0	11,0	10,0	jardin en ville
ALTENKESSEL	4,9	7,4	6,1	forêt
PÜTTLINGEN	5,6	8,0	6,7	prairie
BOUS	7,6	9,5	8,5	grès bigarré

Tableau VI (suite 2)

Site	Minimum	Maximum	Moyenne	Observations
SWALLBACH	4,6	7,4	6,2	prairie
LISTDORF	5,8	6,2	6,0	prairie dans vallée Saar
MECHERN	6,1	9,0	7,4	" "
SCHEMLING	4,1	5,5	4,8	champ cultivé
Côte 408 route de REMICH	8,0	8,5	8,2	" "

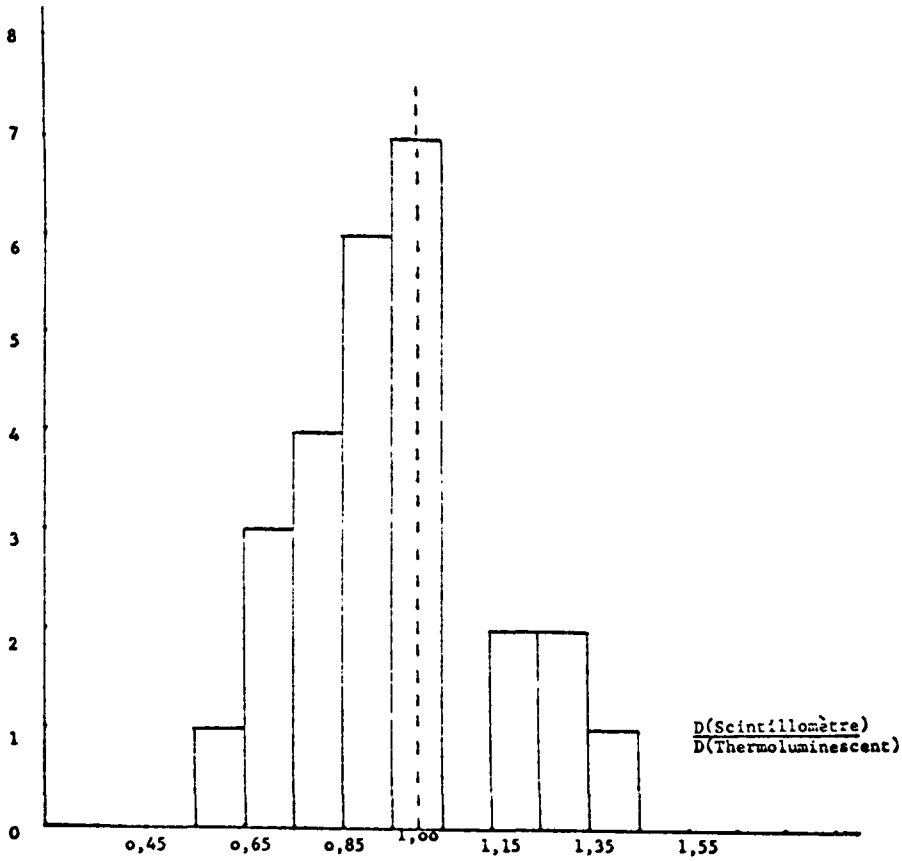


Fig. 1 -Distribution des rapports entre les valeurs données par le Scintillomètre et celles données par les dosimetres thermoluminescents

Figure 2 : Questionnaire relatif aux mesures avec scintillomètre

MESURE DE L'IRRADIATION AMBIANTE

Lieu :

Date : Heure :

Conditions météorologiques :

Localisation des points de mesure (schéma) :

Environnement :

Mesures :

	1	2	3	4	5	Moyenne
Appareil n°						
Appareil n°						
Nature du sol						

Observations :

Fig. 3 : Mesure de l'irradiation externe d'origine tellurique aux frontières de la République Fédérale Allemande (µrad/h).

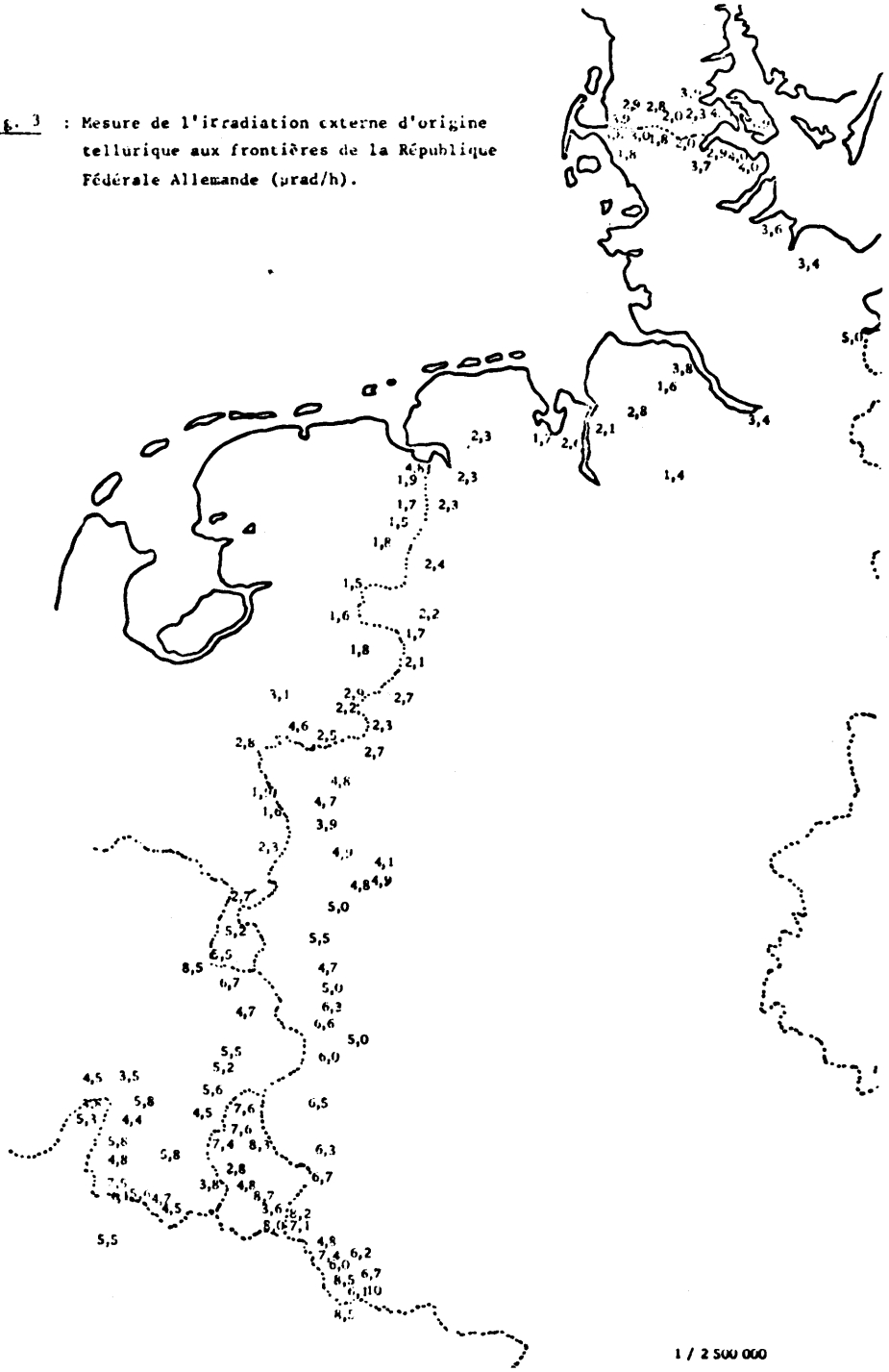


Fig. 4 : Mesure de l'irradiation externe d'origine tellurique au Luxembourg (µrad/h).

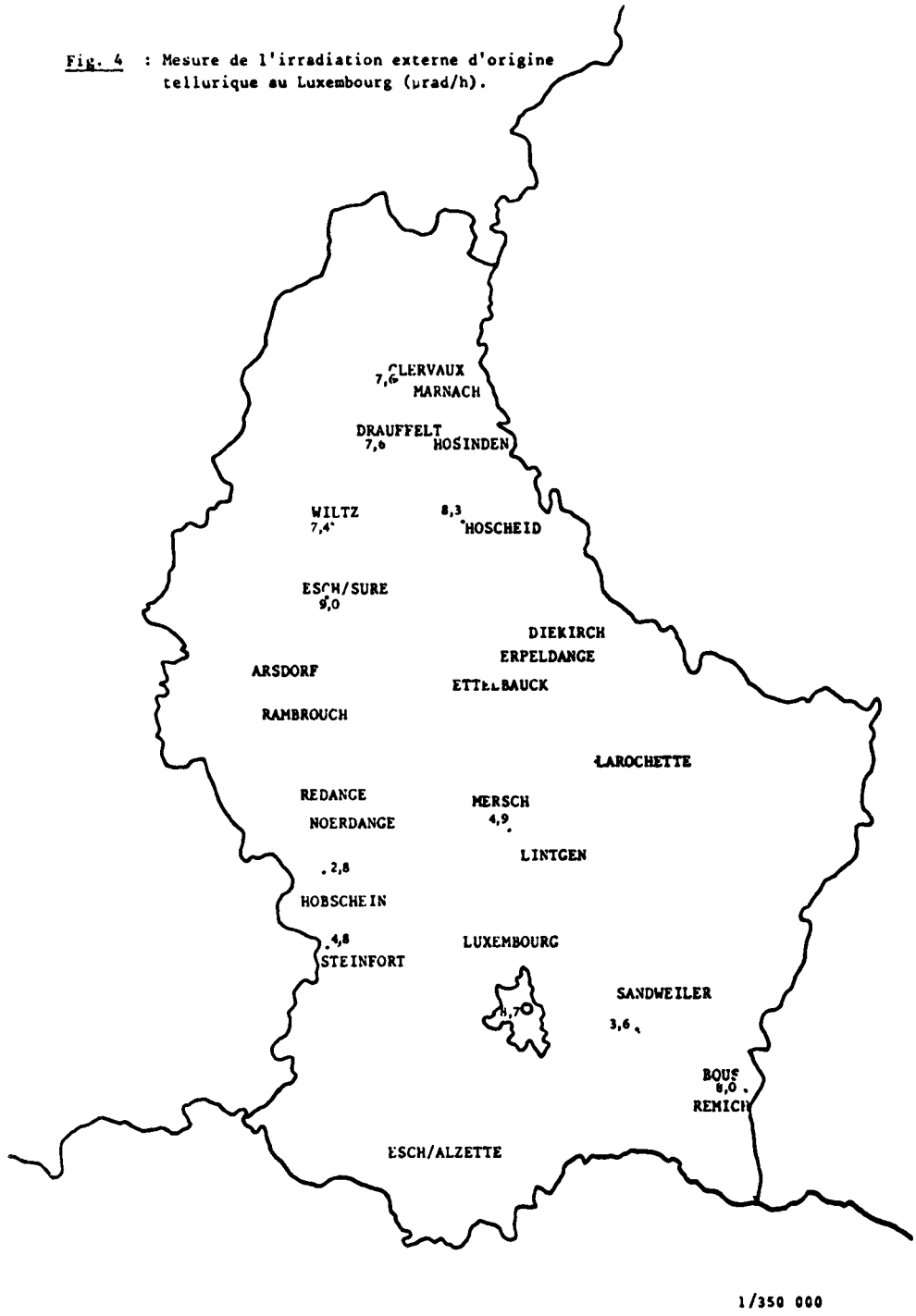
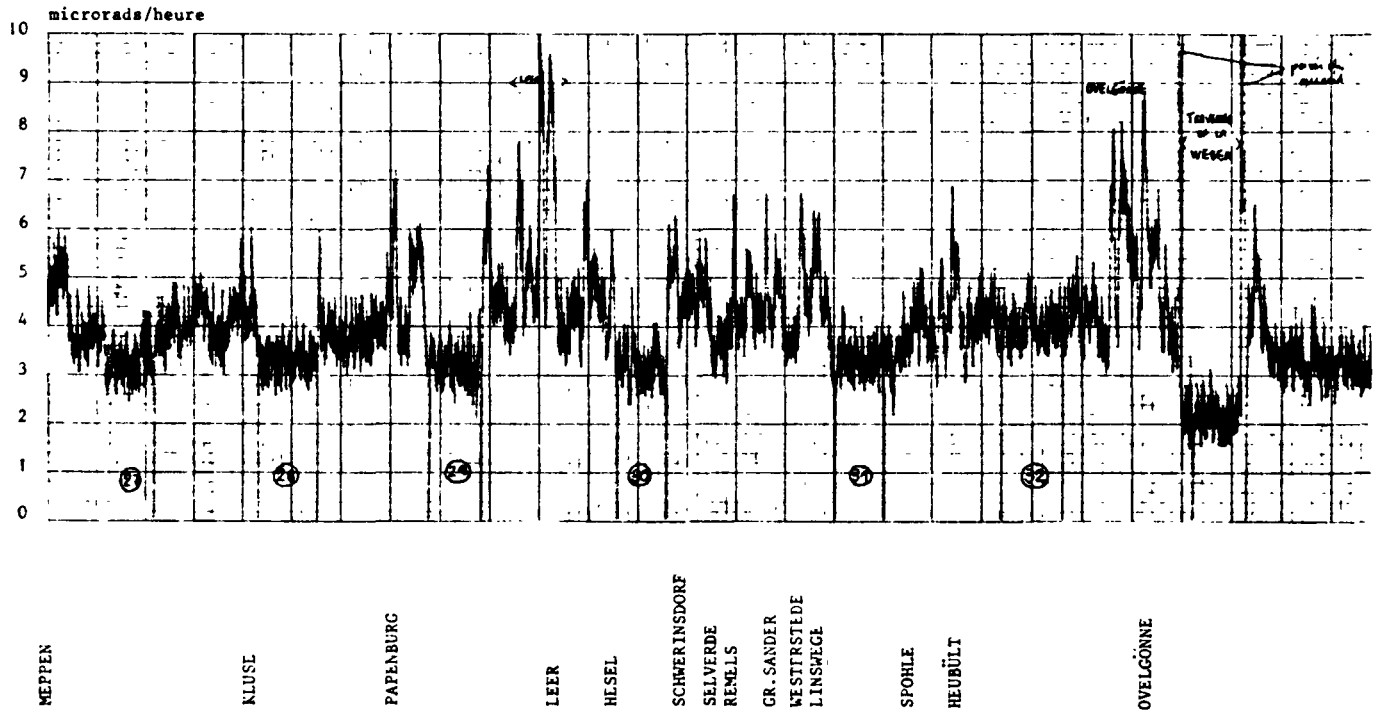


Figure 5 : Extrait de l'enregistrement de l'irradiation externe d'origine tellurique à bord d'un véhicule



DIE TERRESTRISCHE UND KOSMISCHE STRAHLUNG IN DER BUNDESREPUBLIK DEUTSCHLAND

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KURZFASSUNG. Die jüngste systematische und detaillierte Analyse der Verteilung der durch die terrestrische Strahlung bewirkten Ortsdosisleistungen in der Bundesrepublik Deutschland wurde auf Anregung und unter der Schirmherrschaft des Bundesministers des Innern im Zeitraum 1973/74 von verschiedenen nationalen Instituten durchgeführt. Ergebnisse dieser Analyse, die sich auf etwa 25 000 Messungen der Ortsdosisleistungen im Freien und auf etwa 30 000 Messungen der Ortsdosisleistung in Räumen stützen, werden dargelegt, interpretiert und diskutiert.
Die Verteilung der in der Bundesrepublik Deutschland durch die kosmische Strahlung bewirkten Ortsdosisleistungen wurde berechnet. Die Ergebnisse der Berechnungen sowie die grundlegenden Daten und Annahmen, die den Berechnungen zugrunde gelegt wurden, werden dargelegt und diskutiert.

SUMMARY. LEVELS OF TERRESTRIAL AND COSMIC RADIATION IN THE FEDERAL REPUBLIC OF GERMANY. The most recent systematic and detailed analysis of the distribution of exposure rates due to terrestrial radiation in the Federal Republic of Germany was performed on behalf of and under the auspices of the Federal Ministry of Interior by various national institutes in 1973/74. Results of the analysis, based on about 25,000 outdoor- and about 30,000 indoor exposure rate measurements are presented, interpreted and discussed.
The distribution of exposure rates due to cosmic radiation in the Federal Republic of Germany has been calculated. The results of the calculations, the basic data used and the assumptions made in these calculations are presented and discussed.

RESUME. LE RAYONNEMENT TERRESTRE ET COSMIQUE EN REPUBLIQUE FEDERALE D'ALLEMAGNE. La dernière analyse systématique et détaillée de la distribution des débits de dose locaux dus au rayonnement terrestre en République fédérale d'Allemagne a été réalisée en 1973/74 par divers instituts nationaux, à l'instigation et sous le patronage du ministre de l'intérieur. Les résultats de cette analyse, qui se fondent sur quelques 25.000 mesures du débit de dose local en plein air et sur quelques 30.000 heures à l'intérieur des locaux, sont exposés, interprétés et discutés.

On a calculé la distribution des débits de dose locaux dus au rayonnement cosmique en République fédérale d'Allemagne. Les résultats des calculs ainsi que les données et hypothèses de base qui ont été utilisés pour les calculs, sont exposés et discutés.

Die jüngste systematische und detaillierte Analyse der Verteilung der durch die terrestrische Strahlung bewirkten Ortsdosisleistungen in der Bundesrepublik Deutschland wurde auf Anregung und unter der Schirmherrschaft des Bundesministers des Innern von verschiedenen nationalen Instituten 1973/74 durchgeführt.
Die Ergebnisse dieser Analyse stützen sich auf 25.000 Ortsdosisleistungsmessungen im Freien und 30.000 Ortsdosisleistungsmessungen in Wohnhäusern, die mit kalibrierten Standardinstrumenten durchgeführt wurden.

Unter der Annahme des sehr unwahrscheinlichen Daueraufenthaltes im Freien variierten die aus den Ortsdosisleistungen berechneten Jahres-Keimdrüsendosen zwischen 3 und 250 mrem und lieferten einen bevölkerungsgewichteten Mittelwert von etwa 38 mrem.

Unter der Annahme des Daueraufenthaltes in Wohngebäuden variierten die Jahres-Keimdrüsendosen zwischen 9 und 210 mrem und lieferten einen bevölkerungsgewichteten Mittelwert von etwa 50 mrem.

Mit einem Aufenthaltsfaktor von 0,8 für den Aufenthalt in Räumen und einen Aufenthaltsfaktor von 0,2 für den Aufenthalt im Freien ergibt sich als Folge der Exposition durch die terrestrische Strahlung in Räumen und im Freien für die Jahres-Keimdrüsendosis ein bevölkerungsgewichteter Mittelwert von etwa 48 mrem. Dieser Wert zeigt, daß für den Vorteil,der mit dem Aufenthalt in Räumen verbunden ist, eine Erhöhung der mittleren Jahres-Keimdrüsendosis durch die terrestrische Strahlung um etwa 10 mrem in Kauf genommen wird.

Bevölkerungsgewichtete Mittelwerte wurden durch Abschätzung der mittleren Jahresdosen für jeden der 560 Stadt- und Landkreise gewonnen, für die die Bevölkerungsdaten verfügbar sind.

Mittlere Jahresdosen als Folge der Exposition durch die kosmische Strahlung wurden für jeden dieser Kreise aus der Höhe ü.M. der Hauptsiedlungsgebiete errechnet. Nach diesen Berechnungen ergibt sich ein bevölkerungsgewichteter Beitrag der kosmischen Strahlung zur mittleren Jahres-Keimdrüsenäquivalentdosis unter Annahme des Daueraufenthaltes im Freien von etwa 33 mrem und unter Annahme des Daueraufenthaltes in Räumen von etwa 28 mrem. Unter Verwendung der o.a. Aufenthaltsfaktoren ergibt sich der tatsächliche Wert zu etwa 29 mrem.

Daraus ergibt sich, daß bei Berücksichtigung der terrestrischen und kosmischen Strahlung für den Vorteil, der mit dem Aufenthalt in Räumen verbunden ist, insgesamt nur eine Erhöhung der mittleren Jahres-Keimdrüsendosis um etwa 5 mrem in Kauf genommen wird.

Verfahren und Ergebnisse der Analysen über die durch die terrestrische Strahlung bewirkten Ortsdosisleistungen in der Bundesrepublik Deutschland sind in einem Bericht des Bundesministers des Innern [17] ausführlich dargestellt worden. Deshalb können die folgenden Ausführungen auf einige ergänzende in dem Bericht nicht enthaltene Auswertungen beschränkt werden.

Abb. 1 veranschaulicht die Verteilung der für die Stadt- und Landkreise festgestellten Mittelwerte der Ortsdosisleistung durch terrestrische Strahlung im Freien, die in mR pro Jahr angegeben sind. Als Gebiete mit überdurchschnittlich hohen Ortsdosisleistungen lassen sich unschwer der Bayerische Wald und das Fichtelgebirge, bestimmte Bereiche des Schwarzwaldes, des Saarlandes und der Vulkaneifel identifizieren. Dieses Ergebnis entspricht der geologischen Struktur dieser Gebiete, in denen oberflächennah uranhaltige Mineralien vorkommen.

Für die Differenz (D_i-D_e) der mittleren Ortsdosisleistungen in Gebäuden (D_i) und im Freien (D_e) ergibt sich ein komplexes Bild, weil der Wert von D_i nicht nur von der Dosisleistung (D_b), die durch den Gehalt der Baumaterialien an natürlich

radioaktiven Stoffen bestimmt wird, sondern auch in bestimmtem Maße von D_e abhängt. Diese Abhängigkeit kann grob durch die in Abb. 2 angegebenen Relationen dargestellt werden.

Abb. 3 veranschaulicht die Verteilung der für die Stadt und Landkreise festgestellten Differenz der Mittelwerte aus der Ortsdosisleistung in Gebäuden und der Ortsdosisleistung im Freien. Die Werte sind ebenfalls in mR pro Jahr angegeben. Es wird deutlich, daß in Gebieten, in denen die Ortsdosisleistung im Freien 100 mR pro Jahr überschreitet, die Differenz sogar negative Werte annehmen kann. In diesen Fällen ist $(1-\frac{1}{n})\ D_e > D_b$ (vgl. Abb. 2). Ist die Ortsdosisleistung im Freien geringer, so wird die Ortsdosisleistung in Gebäuden vorwiegend durch die in den Baumaterialien enthaltenen natürlichen radioaktiven Stoffe bestimmt. Im Norden der Bundesrepublik werden danach vorwiegend Baumaterialien mit geringem Gehalt an natürlich radioaktiven Stoffen verwendet. Dagegen haben die erhöhten Differenzwerte in Teilen des Saarlandes auf verbreitete Verwendung von Schlackensteinen und in Teilen des Rheinlandes und der Rheinpfalz auf die verbreitete Verwendung von Bims aus der Vulkaneifel schließen lassen.

Für die Berechnung der durch die kosmische Strahlung bedingten Mittelwerte der Ortsdosisleistung in den Stadt- und Landkreisen wurde angenommen, daß der langjährige Mittelwert der Jahresäquivalentdosis $\overline{H}$ in der in Abb. 4 dargestellten Weise von der Höhe des Bezugsortes über dem Meeresspiegel abhängt. Dabei ist $\overline{H} = \overline{H}_i + \overline{H}_n$, wobei H_i den Beitrag der ionisierenden Komponente und $\overline{H}_n$ den langjährigen Mittelwert des Beitrages der Neutronenkomponente bezeichnet.
H variiert zwischen H max (bei solarem Minimum) und H min (bei solarem Maximum). Die Daten wurden von UNSCEAR /27/ übernommen und beziehen sich auf den Aufenthalt im Freien.
Als Qualitätsfaktoren wurden angesetzt

- für die ionisierende Komponente QF = 1
- für die Neutronenkomponente QF = 6,5.

Die Fehler der Berechnungen hängen vor allem von der Unsicherheit ab, mit der die Qualitätsfaktoren behaftet sind.

Bezüglich des Aufenthaltes in Gebäuden ist auch deren Abschirmung gegen die kosmische Strahlung in Rechnung zu stellen. UNSCEAR [2] stützt sich auf einen NRPB-Bericht [3], in dem angenommen wird, daß eine Betonschicht mit einer Dicke von etwa 20 cm (Massenbedeckung von 50g/cm^2) eine Gesamtschwächung der kosmischen Strahlung um etwa 30% bewirkt. Lauterbach und Kolb [4] haben in einer jüngeren Arbeit jedoch gezeigt, daß erst eine Massenbedeckung von 100g/cm^2 zu einer Schwächung der kosmischen Strahlung um 30% führt. Wird angenommen, daß im Mittel eine Massenabdeckung von 40g/cm^2 vorliegt, ist danach beim Aufenthalt in Gebäuden die Ortsdosisleistung um 15% geringer als im Freien und liegt somit bei etwa 28 mrem pro Jahr. Unter Anwendung der o.a. Aufenthaltsfaktoren ergibt sich damit eine Jahres-Keimdrüsenäquivalentdosis durch die kosmische Strahlung von etwa 29 mrem.

Die Ergebnisse der hier behandelten Erhebungsmessungen und Berechnungen haben erheblich zum Verständnis der Parameter beigetragen, die auf die Strahlenexposition durch die terrestrische und kosmische Strahlung in der Bundesrepublik Deutschland von wesentlichem Einfluß sind, sie haben im übrigen die Grundlage für ergänzende Erhebungsmessungen gebildet,über die gesondert berichtet wird.

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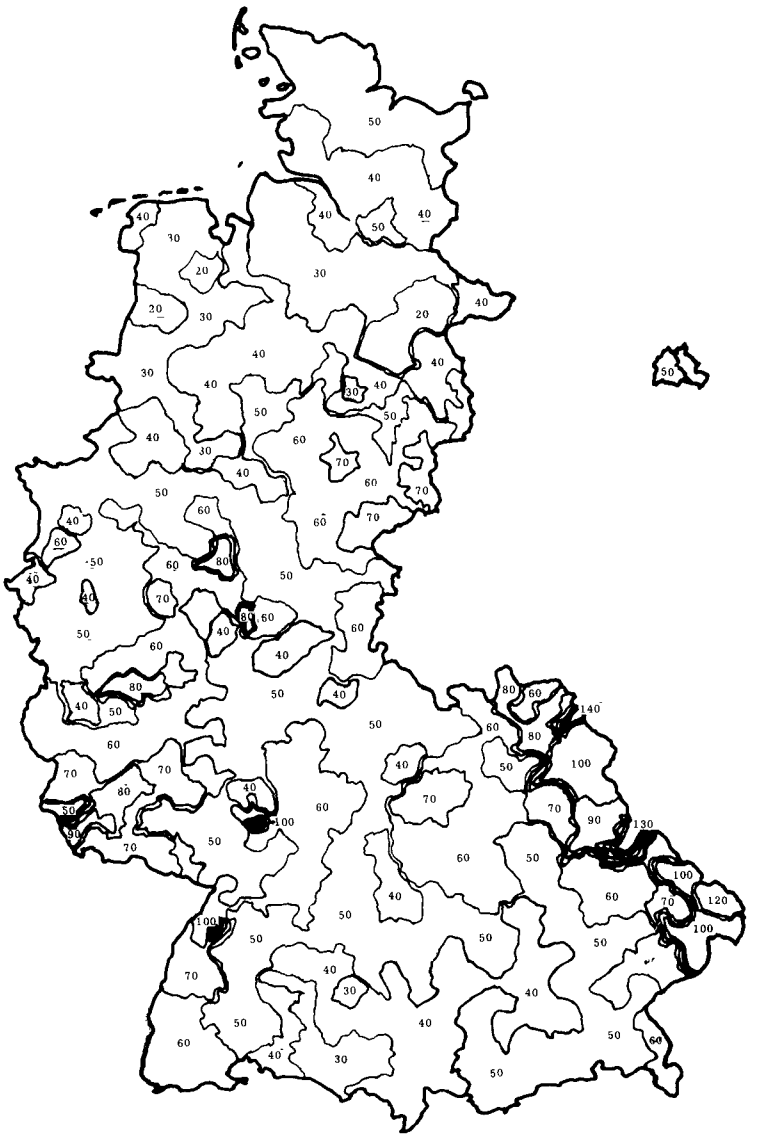
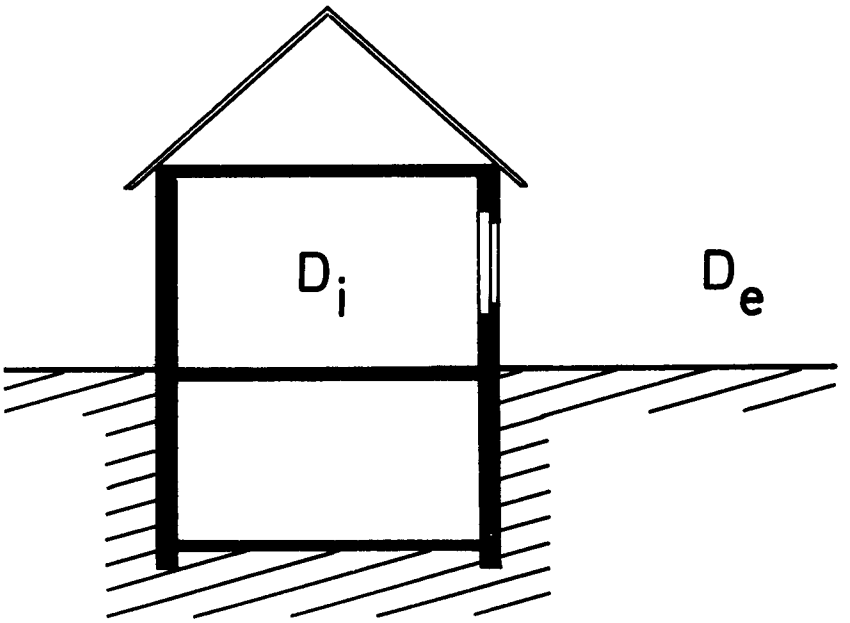


Abb. 1:
Verteilung der für die Stadt- und Landkreise festgestellten Mittelwerte der Ortsdosisleistung durch die terrestrische Strahlung im Freien (D_e in mrem pro Jahr)



$$D_i = D_b + \frac{1}{n} D_e$$

$$D_b = D_i - \frac{1}{n} D_e$$

$$D_i - D_e = D_b - D_e\left(1 - \frac{1}{n}\right)$$

Abb. 2·

Einfaches Modell zur Erläuterung der Relationen zwischen

D_i = Dosisleistung im Gebäude

D_e = Dosisleistung im Freien

D_b = Dosisleistung durch Baumaterialien bedingt

n = Abschirmfaktor des Gebäudes

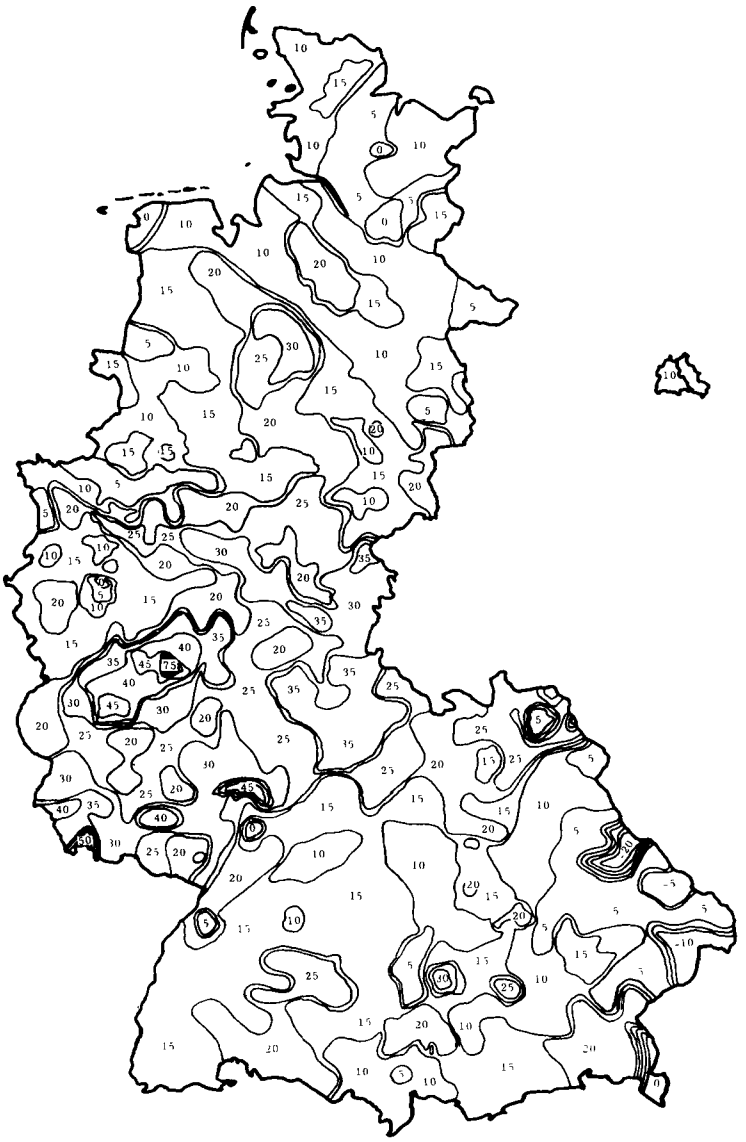


Abb. 3:

Verteilung der für die Stadt- und Landkreise festgestellten Differenzen der Mittelwerte zwischen der Ortsdosisleistung in Gebäuden (D_i) und der Ortsdosisleistung im Freien (D_e)

(D_i - D_e in mrem pro Jahr)

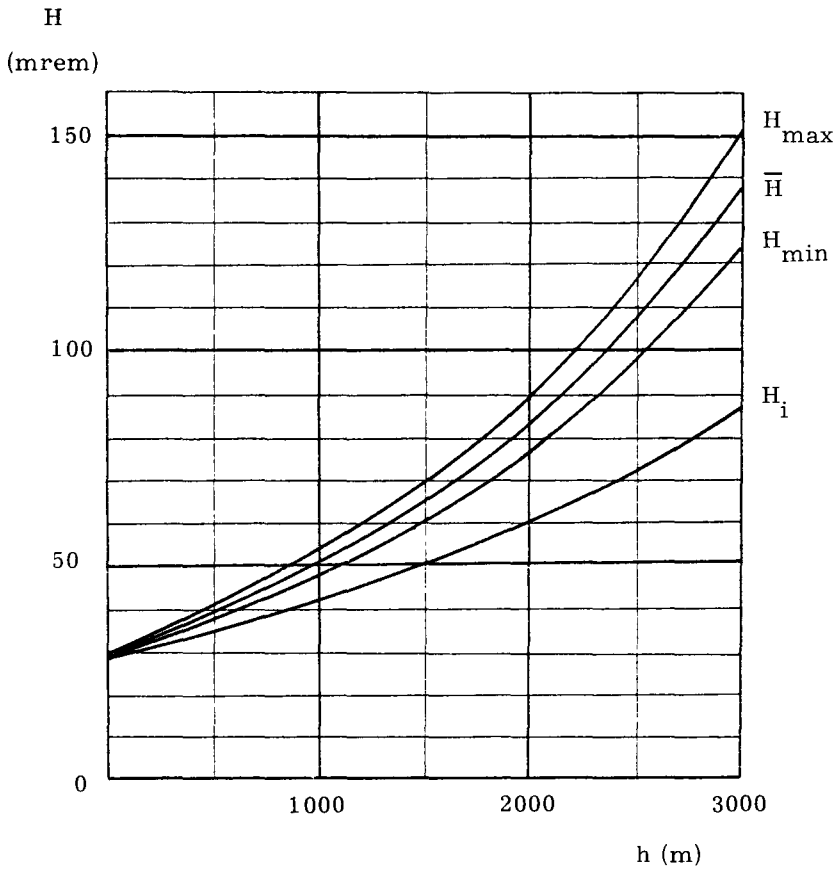


Abb. 4:

Jahresäquivalentdosen H(mrem) durch die kosmische Strahlung in Abhängigkeit von der Höhe ü. M. h(m)

ERHÖHTE TERRESTRISCHE STRAHLUNG DURCH SCHWERMINERALANREICHERUNGEN AN DER KÜSTE DER INSELN NORDDEUTSCHLANDS

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ZUSAMMENFASSUNG. An der Küste der Ost- und Nordfriesischen Inseln findet durch Wasser- und Windeinwirkung an einigen Stellen eine Anreicherung schwerer Sandkörner mit hohen Schwermineralgehalten statt. Sie haben hohe U-238- und TH-232-Gehalte. Hierdurch ergeben sich lokal wesentlich höhere Ortsdosisleistungen der terrestrischen Gammastrahlung, als üblicherweise in Norddeutschland herrscht. Die starken Erhöhungen verändern sich durch Einwirkung der See und des Windes von Jahr zu Jahr. Die bisher höchste gemessene Ortsdosisleistung beträgt 500 mrem/a. Wären die Gebiete mit den höchsten Schwermineralanreicherungen grösser, so könnten Ortsdosisleistungen bis etwa 1000 mrem/a auftreten. Sie sind nach unserem heutigen Kenntnisstand die höchsten natürlichen Ursprungs in der Bundesrepublik Deutschland. An der Küste der Inseln in Norddeutschland liegt die gleiche Ursache für die erhöhte natürliche Ortsdosisleistung wie an der Küste in Kerala in Indien vor. Dort liegen die Ortsdosisleistungen jedoch zwischen 40 und 7000 mrem/a. Im Seegebiet vor den Ost- und Nordfriesischen Inseln lagern in einem Volumen von etwa $5,6 \cdot 10^6$ m^3 etwa 40 t Uran und 80 t Thorium.

SUMMARY. ENHANCED TERRESTRIAL RADIATION DUE TO ENRICHED HEAVY MINERALS ON THE SHORES OF ISLANDS OFF NORTHERN GERMANY. On the shores of the East and North Frisian Islands an enrichment of heavy sand grains with high heavy mineral concentrations results from the influence of wind and water. These sand grains have high U-238 and Th-232 concentrations and hence enhanced local terrestrial gamma radiation dose rates occur compared with the norm for Nothern Germany. The high enrichment effects change from year to year due to the influence of sea and wind. The highest local dose rate measured to date is 500 mrem/a. If the regions with the highest heavy mineral enrichments were larger, local dose rates of up to approximately 1000 mrem/a might occur. These rates are thought to be the highest of natural origin in the Federal Republic of Germany. The origin of the enhanced local dose rates on the shores of the islands off Northern Germany is the same as for the Kerala coast in India. There, however, local dose rates

vary from 40 to 7000 mrem/a. Approximately 40 t Uranium and 80 t Thorium in a volume of approx. 5.6 x 10^6 m^3 are present in the seabed around the East and North Frisian Islands.

RESUME. EXPOSITION TERRESTRE ACCRUE AUX RAYONNEMENTS EN RAISON DE L'ENRICHISSEMENT EN MINERAUX LOURDS LE LONG DES COTES DES ILES D'ALLEMAGNE DU NORD. Sur la côte des îles de Frise orientale et septentrionale, l'eau et le vent produisent localement un enrichissement en grains de sable lourd à fortes teneurs en minéraux lourds. Ces grains ont de fortes teneurs en U-238 et Th-232. Il en résulte localement des débits de dose terrestre gamma plus élevés que la normale en Allemagne du Nord. Les forts enrichissements se modifient d'année en année sous l'effet de la mer et du vent. On a mesuré des débits de dose locaux atteignant 500 mrem/a. Si les régions à plus forte teneur en minéraux lourds étaient plus étendues, les débits de dose locaux pourraient atteindre environ 1000 mrem/a. Ces débits sont pensés être les plus élevés d'orgine naturelle en République Fédérale d'Allemagne. Sur la côte des îles d'Allemagne du Nord, l'accroissement du débit de dose local a la même origine que sur la côte du Kerala en Inde. Là, cependant, les débits de dose varient entre 40 et 7000 mrem/a. Dans la région maritime située le long des îles de la Frise orientale et septentrionale, approximativement 40 t d'uranium et 80 t de thorium se trouvent dans un volume d'environ 5,6 . 10^6 m^3.

1. Einleitung

In der Bundesrepublik Deutschland ist die natürliche Ortsdosisleistung der terrestrischen Strahlung recht unterschiedlich. Sie ist mit etwa 2o bis 4o mrem/a in Norddeutschland - Sandboden - und in den Voralpen - Kalkboden - am niedrigsten /1/, /2/, /3/, /4/. Bisher war man der Meinung, daß die höchsten Werte von etwa 25o mrem/a ohne menschliche Einwirkung oberhalb Granitgestein im Bayerischen Wald auftreten. Weitere Gebiete mit hoher terrestrischer Ortsdosisleistung sind der Schwarzwald, der Odenwald, das Saarland und der Harz. Durch Veränderung der 2Π-Geometrie in Städten und Nutzung natürlichen Baumaterials wie Granitgestein, Rotliegendes usw. für Straßen, Bauten usw. wurden die Ortsdosisleistungen erheblich verändert. Eine weitere Veränderung erfolgte z.B. durch Verwendung von Schlackensteinen als Straßenbelag und von Gips in Gebäuden. Auf Straßen in Norddeutschland mit Kupferschlackenstein aus Mansfeld in der Deutschen Demokratischen Republik - Rotliegendes - treten Ortsdosisleistungen bis etwa 35o mrem/a auf. Interessant ist, daß es an der Küste Norddeutschlands daneben eine natürliche Erhöhung der terrestrischen Ortsdosisleistung durch Einwirkung der See und des Windes gibt. Die hier auftretenden terrestrischen Ortsdosisleistungen sind wahrscheinlich die höchsten natürlichen Ursprungs in der Bundesrepublik Deutschland.

2. Gebiete mit erhöhter natürlicher Ortsdosisleistung

Die Abb. 1 zeigt z.B. die Veränderung der Ortsdosisleistung durch terrestrische Strahlung an einem Küstenabschnitt der Insel Norderney am 21.1o.1979. Während die terrestrische Ortsdosisleistung über trockenem Sand etwa 15 mrem/a beträgt, erhöhte sich die Ortsdosisleistung bei der Messung bis auf etwa 2oo mrem/a. Der Szintillationszähler Sz 25/64BD des Laboratoriums Prof. Berthold mit Ratemeter-Scaler LB 1821 und Schreiber wurde in einem Rucksack auf dem Rücken in etwa 1 m Höhe getragen. Am Ort mit der höchsten Ortsdosisleistung, der ein

wenig seitlich von der eingezeichneten Route lag, betrug die Ortsdosisleistung, gemessen mit einem Szintillations-Dosimeter H 72o1/5/ der Physikalisch Technischen Bundesanstalt in Braunschweig in 1 m Höhe etwa 35o mrem/a und auf dem Boden 5oo mrem/a. Im Ort Norderney beträgt die Ortsdosisleitung etwa 5o mrem/a. Auf einem Straßenstück von etwa 2oo m Länge am Ortseingang, das mit Kupferschlackensteinen belegt ist, beträgt die Ortsdosisleistung in 1 m Höhe etwa 3oo mrem/a.

Die Orte mit erhöhter natürlicher Ortsdosisleistung fallen in der Regel durch ihre dunkle Färbung auf. Die Abb. 2 zeigt z.B. das Bodenprofil am Ort mit der höchsten Ortsdosisleistung in Abb. 1. Oben liegt eine helle Sandschicht. Danach folgen zwei etwa 5 cm dicke dunkle Sandschichten. In etwa 3o cm Tiefe ist wieder normaler weißer Sand.

Die dunklen Schichten sind auch in der Düne, die von der See teilweise abgetragen wurde, zu erkennen, siehe Abb. 3, 4 und 5.

Als ein weiteres Beispiel ist an der dunklen Färbung in den Abb. 6, 7, 8 und 9 ein Gebiet mit erhöhter natürlicher Ortsdosisleistung auf der Insel Sylt zu erkennen. Während die Ortsdosisleistung über dem weißen normalen Sand etwa 15 mrem/a beträgt, ist die maximale Ortsdosisleistung oberhalb der dunklen Sandschicht in 1 m Höhe 15o mrem/a und auf dem Boden 27o mrem/a. Die obere dunkle Sandschicht ist hier jedoch im Gegensatz zu der in Abb. 2 sehr dünn, siehe Abb. 1o. Die dunkle Sandschicht hatte sich hier nicht an einer Düne wie in den Abb. 2 bis 5, sondern an einer als Inselschutz künstlich im Jahre 1978 angeschwemmten Sandschicht, die nur teilweise abgetragen wurde, ausgebildet.

Sehr schöne Bilder entstehen dort, wo bei Ebbe ablaufendes Wasser von weißem Sand verdeckte dunkle Sandschichten freischwemmt. Die Abb. 11 wurde am 2.12.1978 am Strand von Hörnum auf der Insel Sylt aufgenommen. Andere Beispiele sind in /6/ dargestellt.

3. Grund für die erhöhte Ortsdosisleistung

Aus den Gamma-Spektren einer dunklen und hellen Sandprobe wird sofort deutlich, warum die Ortdosisleistung der terrestrischen Strahlung ansteigt. Die Abb. 12 und 13 zeigen die Gamma-Spektren einer Sandprobe, die am 2.1o.1978 ungefähr an der gleichen Stelle wie in Abb. 3 genommen wurde. Sowohl U 238 als auch Th 232 sind in den dunklen Sandproben um etwa einen Faktor 1oo bzw. etwa 75 angereichert. Die Zerfallsprodukte sind im Gleichgewicht im dunklen Sand. Es zeigen sich keine Linien des Atomwaffenfallouts, der Ableitungen aus kerntechnischen Anlagen wie der Wiederaufarbeitungsanlage Cap la Hague, Frankreich, und Windscale, Großbritannien /8/, oder von wissenschaftlichen Untersuchungen über die Sandbewegungen mit Cr 51 oder Sc 46 /9/, /1o/, /11/.

Die Tab. 1 zeigt als Beispiel die Aktivität von U 238 und Th 232 einiger Bodenproben. Die Probe Nr. 3 stammt von der ersten dunklen Schicht in Abb. 2 und die Probe Nr. 5 von der dunklen Schicht in Abb. 1o. Granit hat zum Vergleich pro Gramm etwa eine Aktivität von 1,5 pCi U238, 1,5 pCi Th 232 und 25 pCi K 4o /12/, /13/. In den an der Küste der Ost- und Nordfriesischen Inseln sich ausbildenden dunklen Sandschichten ist daher wahrscheinlich oberirdisch die höchste Aktivität natürlichen Ursprungs in der Bundesrepublik Deutschland.

Wären die Sandschichten mit der in Tab. 1 angegebenen Aktivität unendlich ausgedehnt, oder was praktisch das gleiche ist, etwa o,5 m dick und hätten sie einen Durchmesser von etwa 3oo m, so errechnen sich mit dem Umrechnungsfaktor in /14/ und /15/ die in Tab. 2 angegebenen Ortsdosisleistungen. Die gemessenen höchsten Werte an den verschiedenen Orten liegen wegen der geringeren Ausdehnung der dunklen Bodenschichten um mehr als einen Faktor 2 niedriger.

Fragt man nun nach dem physikalischen Grund für die erhöhte natürliche Strahlung an einigen Stellen an der Küste der Norddeutschen Inseln, so fällt sofort die hohe Dichte des dunklen

Sandes gegenüber dem hellen Sand auf. Während der getrocknete helle Sand von Probe 1 eine Dichte von 2,1 g/cm^3 hat, beträgt z.B. die Dichte des trockenen dunklen Sandes von Probe 2 3 g/cm^3 /6/.

Die Abb. 14 zeigt den dunkel aussehenden Sand unter dem Mikroskop. Im dunklen Sand gibt es Körner, die wasserhell, gelb, rosa, rot, grün, hellbraun, braun und schwarz aussehen. Die Häufung der schwarzen und dunkel gefärbten Körner geben dem dunklen Sand die auffällige Färbung. Im hellen Sand überwiegt nur die Zahl der hellen Sandkörner. Ein Teil der schwarzen Körner läßt sich mit einem Hufeisenmagneten aussondern. Die Dichte dieser Körner wurde mit einem Pyknometer zu 4,1 g/cm^3 bestimmt. Einge Sandkörner haben eine Dichte über 5 g/cm^3. Im Vergleich hierzu beträgt die Dichte des hellen Sandes, der praktisch nur aus Quarz besteht, 2,6 g/cm^3.

Aufschluß über die Zusammensetzung des dunklen und hellen Sandes gibt eine Röntgenfluoreszenzanalyse, die im Gemeinschaftslabor für Elektronenmikroskopie der Rheinisch-Westfälischen Technischen Hochschule Aachen angefertigt wurde, siehe Abb. 15 und 16. Während der helle Sand praktisch nur aus Silizium besteht, enthält der dunkle Sand vor allem Eisen und Titan. Daneben ist offenbar viel Zirkon im dunklen Sand. Der dunkle Sand enthält also einen hohen Anteil Schwermineralien. Die Zusammensetzung einzelner Körner ist in /6/ dargestellt.

Die Anreicherung von Schwermineralien in den sogenannten Erzseifen an der Küste Norddeutschlands ist seit Anfang des letzten Jahrhunderts bekannt. Von v. Engelhardt /16/ wird theoretisch beschrieben, wie eine Wasser- bzw. Windsortierung der Sande zustandekommt. Er unterscheidet zwischen Schweben, Rollen und Springen. Bei schwebender Partikelverfrachtung fallen bei Abnahme der Geschwindigkeit zunächst die schweren Partikel aus. Erfolgt dies wegen Strömungsumlenkung über einen langen Zeitraum, so erfolgt eine Trennung von leichten und schweren Partikeln. Auch bei einer reibenden Fortbewegung, die in /16/ mit Rollen bezeichnet wird, ist die Reibkraft eine Funktion vom Partikelgewicht. Es erfolgt eine Ordnung der Körner nach dem Reibwiderstand bei der Bewegung. Hierauf ist auch zurückzuführen,

daß in der Regel die dunklen Partikel kleiner sind als die hellen. Bei einem spezifischen Gewicht von 5 g/cm^3 für Magnetit und 2,5 g/cm^3 für Quarz ist das Radiusverhältnis $r_{Quarz}/r_{Magnetit} \approx 2$, siehe /16/. Meist beobachtet man eine springende Fortbewegung der Sandkörner, die als Überlagerung von Schweben und Rollen angesehen werden kann. Die Sortierung in leichtere und schwerere Fraktionen wurde auch von Scheidhauer /17/ an den Sanden des Elbsandsteingebirges festgestellt.

In Abb. 17 wurde eine homogene Mischung des trockenen Sandes der Probe 2 in Tab. 1 auf ein Papier geschüttet. Einige Hin- und Herbewegungen der Papierunterlage entmischten den Sand in Bereiche mit höherer und niedrigerer Dichte.

Die Entstehung von Schwermineralseifen an der Küste Norddeutschlands kann man sich danach wie folgt vorstellen: Im Wasser erfolgt durch die oben beschriebenen Vorgänge die erste Aufkonzentrierung. Eine weitere Konzentrierung erfolgt z.B. vor der Düne durch Windeinfluß, siehe Abb. 18. Aufgrund der unterschiedlichen Windgeschwindigkeiten und Windrichtungen bilden sich in der Düne Nester aus, in denen die schweren Partikel angereichert sind. Nach Wasmund /18/ erfolgt die Lagerstättenbildung durch Seegangseinwirkung, bei der die Vordüne abgetragen wird. Hierbei werden die Leichtmineralien weggeschwemmt, während die Schwermineralien liegen bleiben. Dies liegt in der Nähe des Schiffwracks auf der Insel Norderney (Abb. 1) vor, siehe Abb. 2 bis 5. Die Anreicherung von Schwermineralien ohne vorherige Bildung einer Düne zeigen die Abb. 6 bis 9.

Die erste größere Bestandsaufnahme der Schwermineralseifen erfolgte 1938, siehe /18/ und /19/. Eine zweite Erkundung im Bereich des Niedersächsischen Küstenraumes und der Deutschen Bucht erfolgte 1977 von Ludwig /2o/. Es wurden jedoch die Schwermineralseifen an den Küsten ausgeklammert, da man der Meinung ist, daß wegen des großen Freizeitwertes der Strände ein Abbau der Schwermineralien dort nicht möglich ist. Die Abb. 19 zeigt nach /2o/ die Gebiete vor den Ost- und Nordfriesischen Inseln, in denen eine Schwermineralanreicherung von größer als 2 % gemessen wurde.

Ausgangsmaterial des Küstensandes sind die Gesteine des Skandinavischen Kontinents, die auch auf dem Festland Norddeutschlands anzutreffen sind. Die Steingründe werden als Endmoränen des Warthestadiums und der Weichseleiszeit gedeutet /21/. Die Überflutung der südlichen Hälfte der Nordsee begann erst vor etwa 1o ooo Jahren, als mit dem Abschmelzen des Eises über der Ostsee sich das Gebiet der Nordsee senkte /21/, /22/. Der Sand ist durch Zerkleinerung der quarzhaltigen Eruptivgesteine entstanden. In dem Sand findet man die festen Mineralkörnchen, vor allem der Granite, wieder. Schwermineralanreicherungen findet man überall dort, wo die Fließgeschwindigkeit des Wassers sehr ausgeprägt ist, vor allem bei den Ostfriesischen Inseln von Westen nach Osten. Da hohe Schwermineralgehalte an den Stränden nicht möglich sind, wenn im davorliegenden Seegebiet nicht eine Vorkonzentrierung erfolgt ist, ist überall dort, wo im Seegebiet hohe Schwermineralkonzentrationen gemessen wurden, am Strand mit Erzseifen mit hohen Schwermineralkonzentrationen zu rechnen. Dieses hat sich weitgehend bestätigt.

Um das Uran und Thorium in den schweren Körnern weiter zu lokalisieren, wurde am Institut für Chemische Technologie der Kernforschungsanlage Jülich die Probe Nr. 2 in Tab. 1 mit Hilfe eines Magnetabscheiders getrennt /23/. In Tab. 3 ist der in vier Fraktionen weiter zerlegte paramagnetische Anteil, der diamagnetische und der ferromagnetische Anteil mit den gemessenen Aktivitäten /23/ aufgeführt. Es zeigt sich keine gleichmäßige Verteilung über die 6 Anteile. Sehr hoch sind die Aktivitäten im paramagnetischen Anteil C und D. Alle 6 Fraktionen haben unterschiedliche Färbung. Die Dichte des trockenen ferromagnetischen Anteils ist 4,4 g/cm^3, des paramagnetischen Anteils A 4,2 g/cm^3, des von B 3,5 g/cm^3, des von C 5,8 g/cm^3, des von D 3,5 g/cm^3 und die des diamagnetischen Anteils 2,3 g/cm^3.

Von allen 6 Fraktionen wurden Röntgenfluoreszenzanalysen gemacht /6/. Danach besteht der ferromagnetische Anteil aus Magnetit (Fe_3O_4), Ilmenit ($FeTiO_3$) und Chromit (Cr_2FeO_4) /19/, /2o/. Im paramagnetischen Anteil sind im wesentlichen enthalten Titanit ($CaTiO[SO_4]$), die Titanminerale (TiO_2) Rutil, Anatas, Brookit und Leukoxen, Zirkon ($ZrSiO_4$) mit Einschlüssen von Hafnium, Uran, Thorium sowie Yttrium /2o/. Im paramagnetischen Anteil D

wurden Körner aus Monazit ($Ce[PO_4]$), in dem Thorium in hoher Konzentration enthalten ist, gefunden /6/.

Es scheint auch Thorit ($Th[SiO_4]$) im Sand vorhanden zu sein. Die hohe natürliche Gamma-Ortsdosisleistung wird jedoch nur teilweise durch Monazit hervorgerufen. Der Hauptanteil dürfte darauf zurückzuführen sein, daß im Zirkon Uran und Thorium enthalten sind. In der Mineralogie ist allgemein bekannt, daß dort, wo Zirkon auftritt, auch Uran und Thorium vorhanden sind /24/, /25/, /26/. Die Uran- und Thoriumgehalte können in Zirkon größer als 1 ooo ppm sein.

Mit Hilfe der hier gemessenen Aktivitäten und der von Ludwig /2o/ bestimmten Zirkonmasse von $3{,}8 \cdot 1o^4$ t im Seegebiet vor den Ost- und Nordfriesischen Inseln läßt sich abschätzen, daß in einem Volumen von $5{,}6 \cdot 1o^6$ m^3 etwa 4o t Uran und 8o t Thorium enthalten sind.

Vergleicht man die Daten mit denen an der Küste von Kerala in Indien /27/, /28/, so zeigt sich, daß an der Küste Norddeutschlands die gleiche Ursache für die erhöhte Ortsdosisleistung vorliegt. Nach /27/ liegen die Ortsdosisleistungen in Kerala zwischen 4o und 7ooo mrem/a. Im Gegensatz zu Kerala befinden sich auf Bereichen mit wesentlich erhöhten Schwermineralanreicherungen nicht ständig Menschen. Über sie gehen vor allem Strandwanderer. Während der Badezeit ist es jedoch auch möglich, daß sich Personen über längere Zeiträume in Gebieten mit erhöhter Ortsdosisleistung aufhalten.

Auch an dieser Stelle sei Herrn Dr. H. Kautzky vom Deutschen Hydrologischen Institut und Herrn Dr. I. Gans vom Institut für Wasser-, Boden- und Lufthygiene des Bundesgesundheitsamtes für Vergleichsmessungen gedankt. Danken möchte ich auch Herrn Prof. E. Merz für die Analysen am Institut für Chemische Technologie der Kernforschungsanlage Jülich, Herrn Dr.-Ing. W.-G. Burchard für die Analysen im Gemeinschaftslaboratorium für Elektronenmikroskopie der RWTH Aachen, Herrn Dr. U. Lauterbach und Herrn Dr. W. Kolb von der Physikalisch-Technischen Bundesanstalt für die Überlassung eines Meßgerätes H 72o1 sowie Herrn K. Hunsänger vom Lehrstuhl für Reaktortechnik der RWTH Aachen

für Messungen.

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Proben-Nr.	Ort	Datum	Aktivität [pCi/g]		Bemerkung
			U 238	Th 232	
1	Norderney	2.1o.78	o,2	o,15	Weißer Sand vom Strand in Abb. 1 K 4o: 5 pCi/g
2	Norderney	2.1o.78	19	12	Dunkler Sand vom Strand in Abb. 1
3	Norderney	21.1o.79	2o	22	Dunkler Sand von der obersten Schicht in Abb. 2
4	Sylt	2.12.78	16	2o	Dunkler Sand vom Strand in Hornum
5	Sylt	24.9.79	1o	24	Dunkler Sand von dunkler Schicht in Abb. 7
6	Juist	2o.1o.79	8	5	Dunkler Sand von Westspitze in Höhe Westbake
7	Wangerooge	17.1o.79	8	6	Dunkler Sand vom Strand von Wangerooge

Tab. 1: Gemessene Aktivitäten in verschiedenen Sandproben

Tab. 1: Measured activities in different sand samples

Proben-Nr.	Ort	Theoretisch berechnete höchste Ortsdosisleistung [mrem/a]	Höchste gemessene Ortsdosisleistung [mrem/a]	
			Bodenhöhe	1m Höhe
1	Norderney	15	15	15
2	Norderney	7oo		25o
3	Norderney	1ooo	5oo	35o
4	Sylt	85o		6o
5	Sylt	9oo	27o	15o
6	Juist	3oo	12o	8o
7	Wangerooge	35o		9o

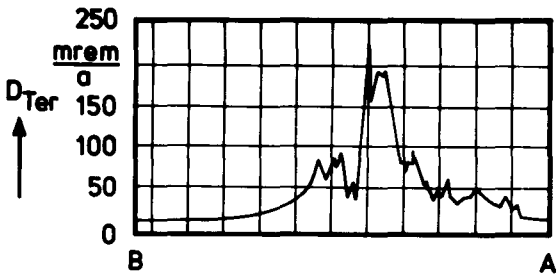
Tab. 2: Vergleich der nach der Aktivität des Sandes bei unendlicher Ausdehnung berechneten Ortsdosisleistung der terrestrischen Strahlung mit Meßwerten

Tab. 2: Comparison of the calculated local dose rate of the terrestric radiation basing on the activity of the sand for infinite expansion with measured data

Anteil 1		Stromstärke [A]	Gewichtsanteil am trockenen Sand	Aktivität bezogen auf den jeweiligen Anteil [pCi/$g_{tr,A}$]		Aktivität bezogen auf die gesamte Mischung [pCi/$g_{tr,ges}$]		Farbe
				U 238	Th 232	U 238	Th 232	
Ferromagnetisch			o,65	o	o	o	o	schwarz
Paramagnetisch	A	o,3	27,7	3	4	o,8	1,1	wasserhell, rosa, rot, schwarz
	B	o,6	38,o	12	18	4,6	6,8	grün, rosa, rot, braun, schwarz
	C	o,7	4,o	124	86	5	3,4	wasserhell, gelb, schwarz
	D	1,o	o,55	84	358	o,5	2	wasserhell, gelb, grün, rot, braun, schwarz
Diamagnetisch (Quarz)			29,1	41	18	12	5,2	wasserhell, hellbraun
Mittelwert			1oo			23	18,5	

Tab. 3: Aufteilung der trockenen dunklen Sandprobe Nr. 2 in Tab. 1 mit Hilfe eines Magnetabscheiders in 6 Fraktionen und Aktivitätsbestimmung der einzelnen Fraktionen /3/

Tab. 3: Separation of the dry sand sample no. 2 in tab. 1 using magnetic separation into 6 fractions and results of the activity measurements of the different fractions /3/



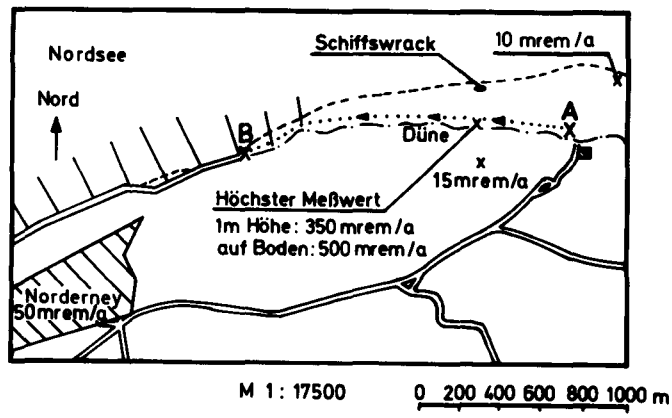


Abb. 1: Bereich mit erhöhter Ortsdosisleistung durch terrestrische Strahlung an der Küste von Norderney

Fig. 1: Area with enhanced local dose rate due to terrestric radiation at the coast of Norderney

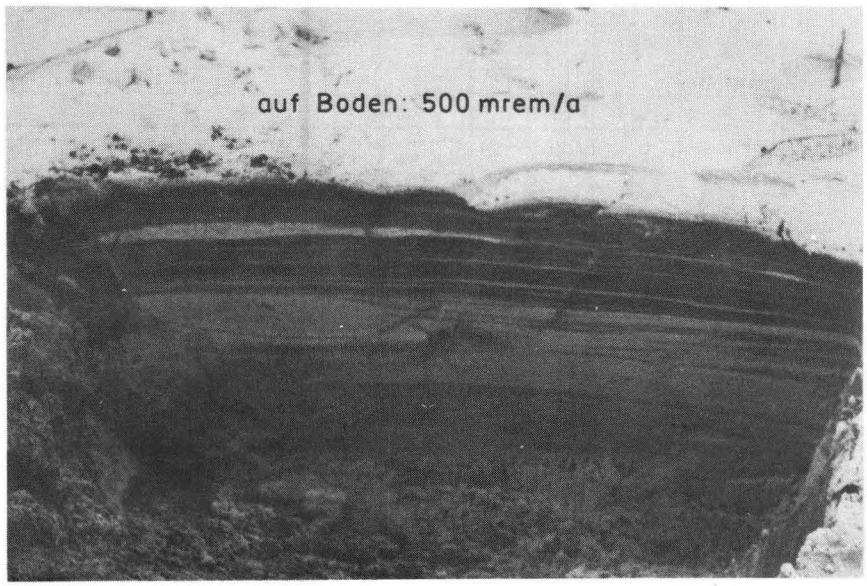


Abb. 2: Bodenprofil von der Stelle der höchsten Ortsdosisleistung in Abb. 1
Fig. 2: Ground side-view of the place with the highest local dose rate in fig. 1

Abb. 3: Dünenabtrag durch angreifende See in der Nähe der höchsten Ortsdosisleistung in Abb. 1
Fig. 3: Dune detrition due to attacking sea near to the highest local dose rate in fig. 1

Abb. 4: Dunkle Sandschichten in der Düne in der Nähe der höchsten Ortsdosisleistung
Fig. 4: Dark sand layers in the dune near to the highest local dose rate

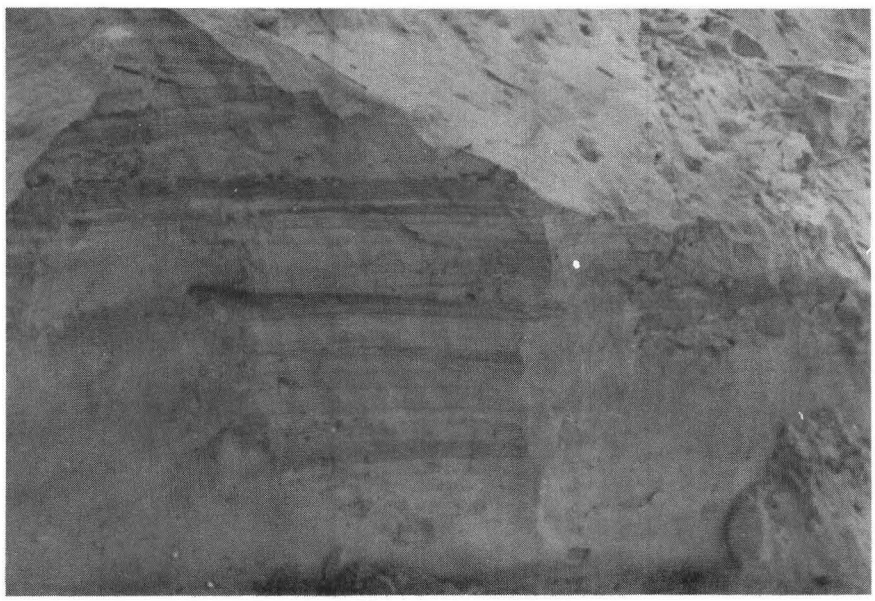


Abb. 5: Ausschnitt von Abb. 4
Fig. 5: Detail of fig. 4

Abb. 6: Dunkle Sandfläche mit erhöhter Ortsdosisleistung am Strand von Westerland auf Sylt
Fig. 6: Dark sand space with enhanced local dose rate at the beach of Westerland on the Isle of Sylt

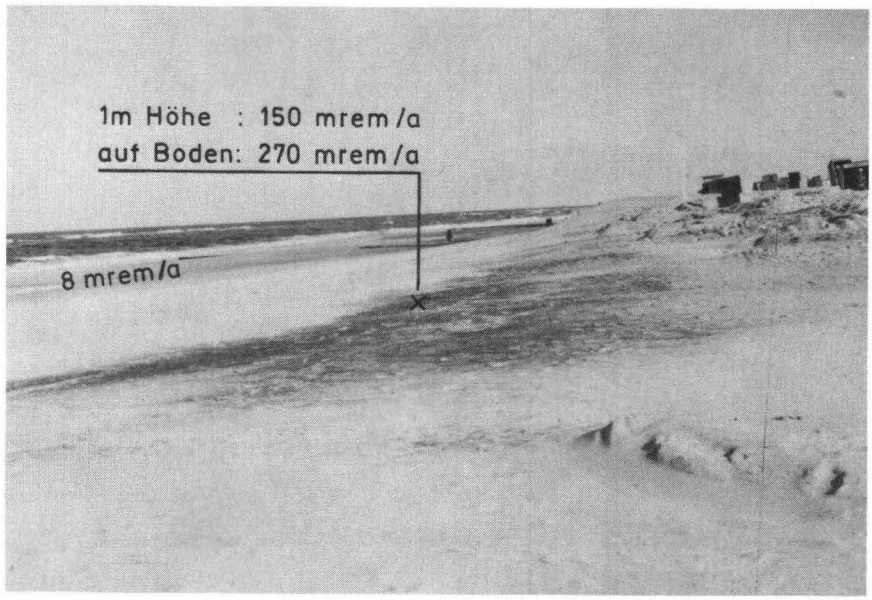


Abb. 7: Dunkle Sandfläche von Abb. 6 aus anderer Richtung photographiert
Fig. 7: Dark sand space in fig. 6 photographed from a different direction

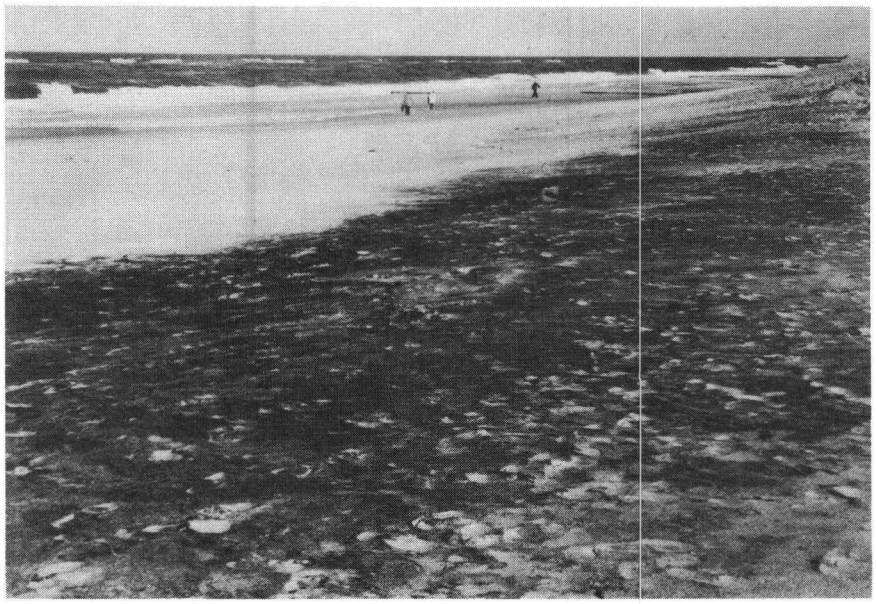


Abb. 8: Mitte der dunklen Sandfläche in Abb. 6
Fig. 8: Middle of the dark sand space in fig. 6

Abb. 9: Dunkle Sandschichten an Stellen, an denen Strandkörbe standen, hinter der dunklen Sandfläche in Abb. 6
Fig. 9: Dark sand space at places where beach chairs stood behind the dark sand space in fig. 6

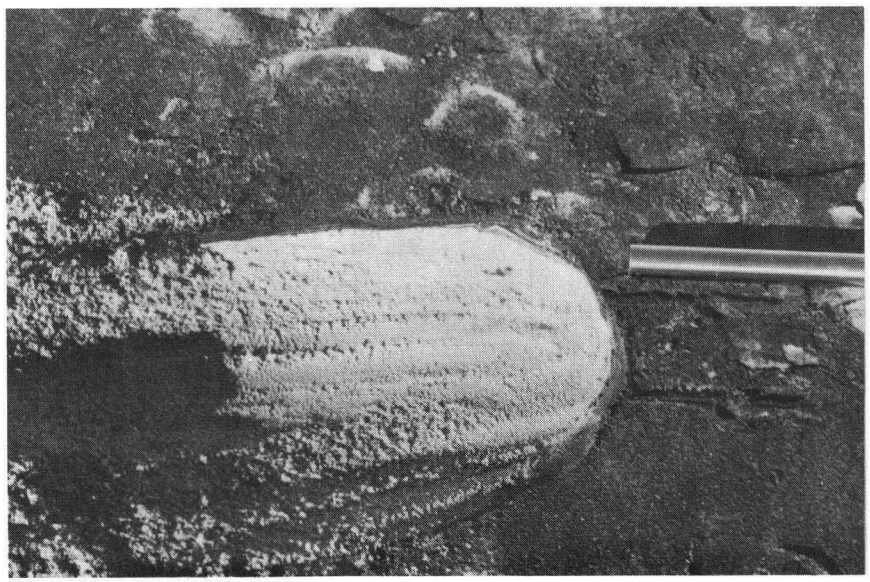


Abb. 1o: Bodenprofil von der dunklen Sandfläche in Abb. 6
Fig. 1o: Ground side-view of the sark sand space in fig. 6

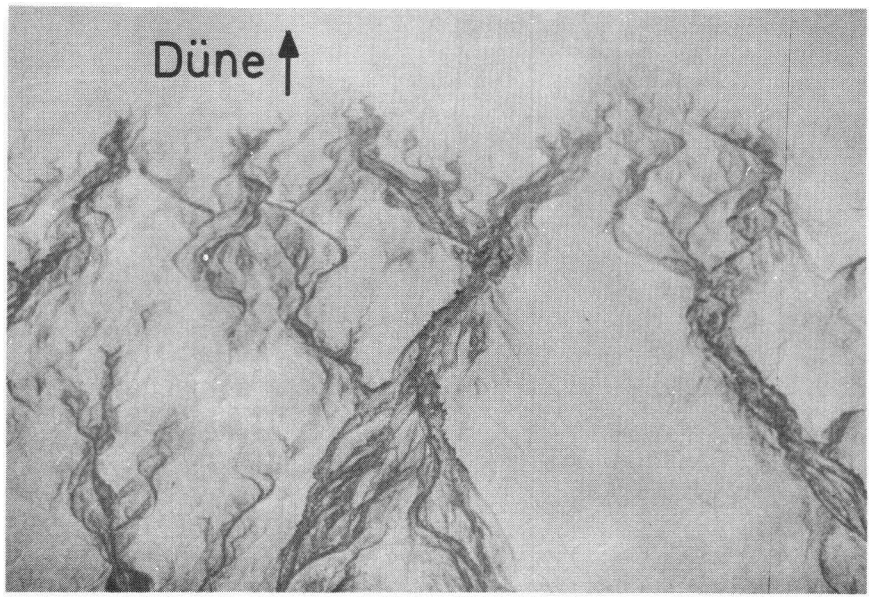


Abb. 11: Unter hellem Sand liegende dunkle Schicht, die durch bei Ebbe ablaufendes Wasser teilweise sichtbar wurde
Fig. 11: Dark layer lying under light sand that became partly visible due to ebbing water

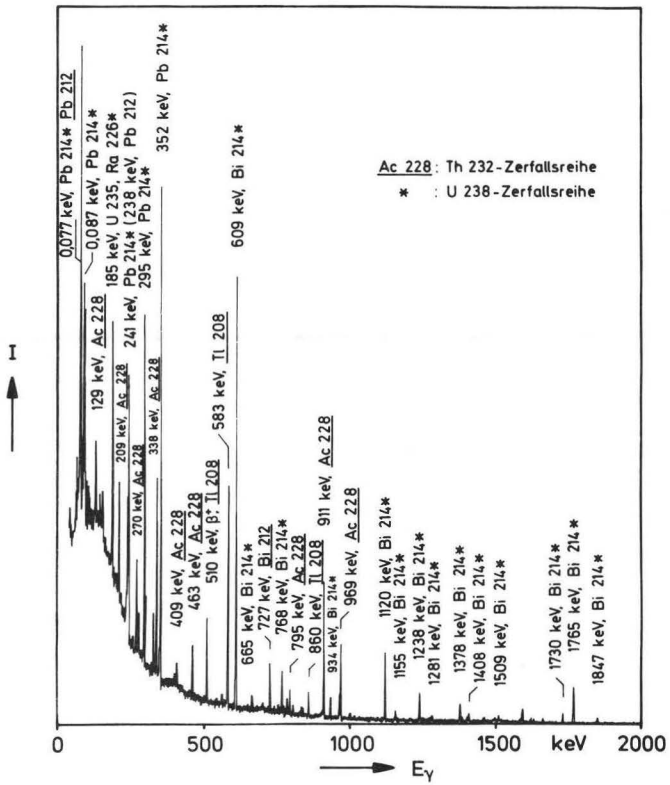


Abb.12: Gammaspektrum der dunklen Sandprobe Nr. 2 in Tab. 2 von Norderney [7]

(Probemasse: 1000 g feucht, Meßzeit: 10^4 s)

Fig.12: Gamma spectrum of the dark sand sample Nr. 2 in table 1 of Norderney [7]

(mass of sample: 1000 g wet, testing time 10^4 s)

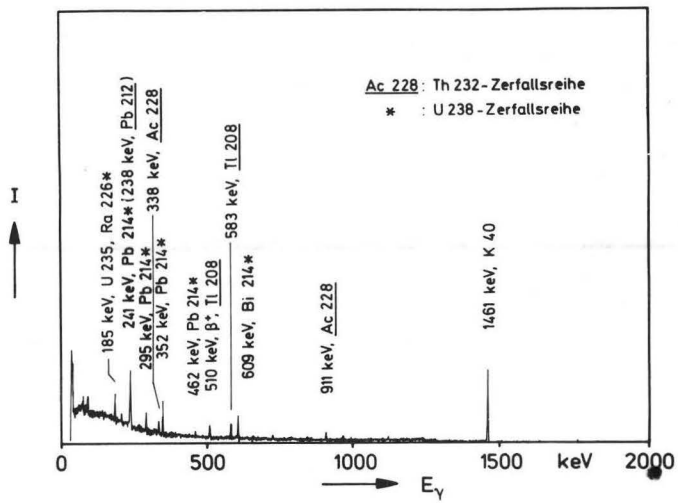


Abb.13: Gammaspektrum der hellen Sandprobe Nr. 1 in Tab. 1 von Norderney [7]

(Probemasse: 1000 g feucht, Meßzeit: $5 \cdot 10^4$ s)

Fig.13: Gamma spectrum of the light sand sample Nr. 1 in table 1 of Norderney [7]

(mass of sample: 1000 g wet, testing time $5 \cdot 10^4$ s)

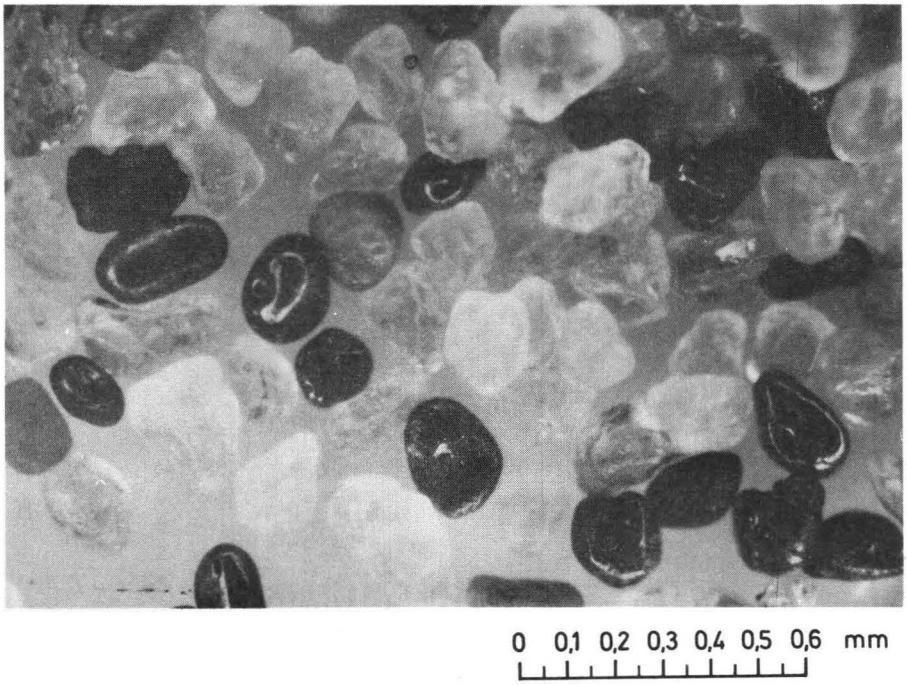


Abb. 14: Dunkler Sand von Probe 2 in Tab. 1 unter dem Mikroskop
Fig. 14: Dark sand of sample 2 in table 1 under the microscope

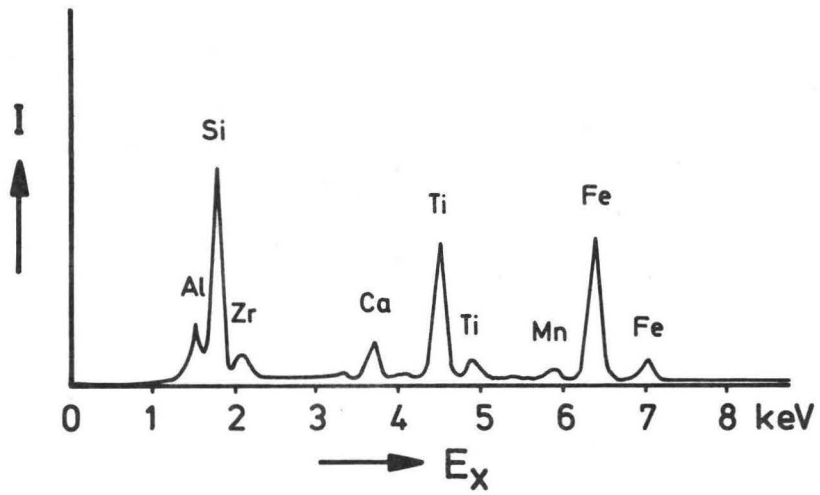


Abb. 15: Röntgenfluoreszenzanalyse von der dunklen Sandprobe Nr. 2 in Tab. 1 (Flächenanalyse)
Fig. 15: X-ray fluorescence of the dark sand sample no.2 in table 1 (total surface)

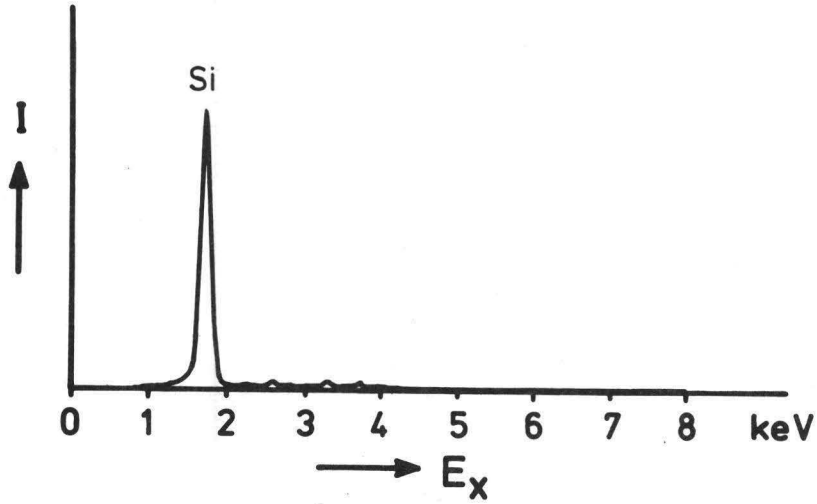


Abb. 16: Röntgenfluoreszenzanalyse von der hellen Sandprobe Nr. 1 in Tab. 1 (Flächenanalyse)

Fig. 16: X-ray fluorescence of the light sand samle no.1 in table 1 (total surface)

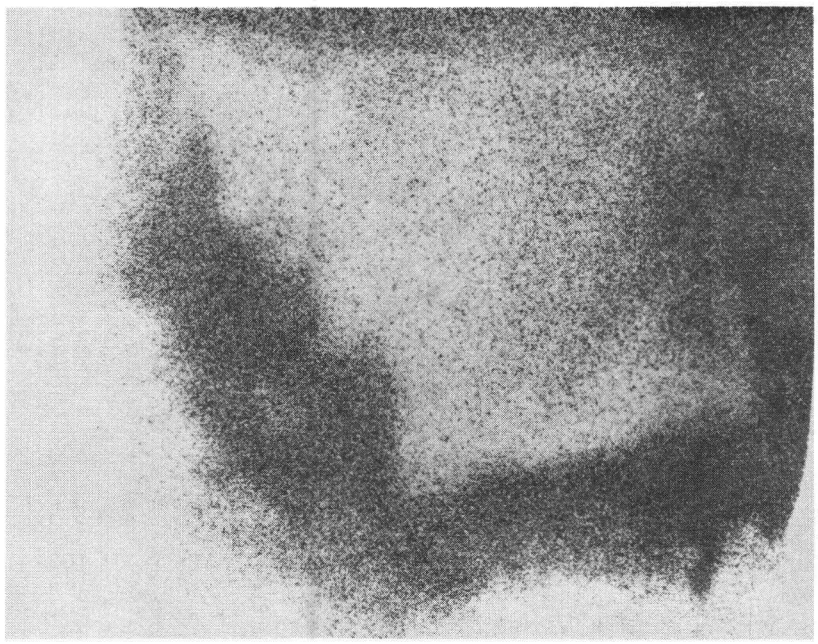
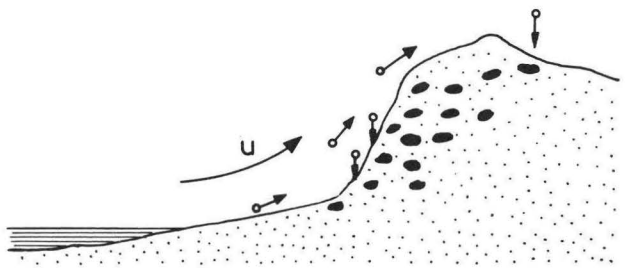
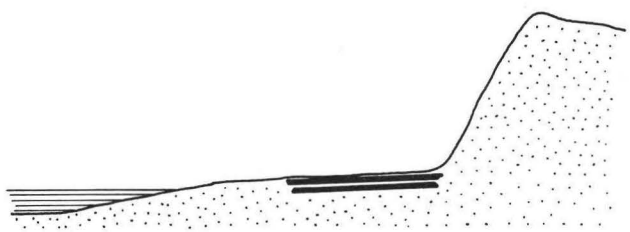


Abb. 17: Entmischung des dunklen Sandes durch Bewegen der Papierunterlage

Fig. 17: Separation of the dark sand by moving the paper support



Anreicherung von Schwermineralien in der Düne



Bildung einer Schwermineralseife nach einer Sturmflut

Abb. 18: Entstehung einer Schwermineralseife durch Wind und angreifende See

Fig. 18: Development of a heavy mineral placer due to wind and attacking sea

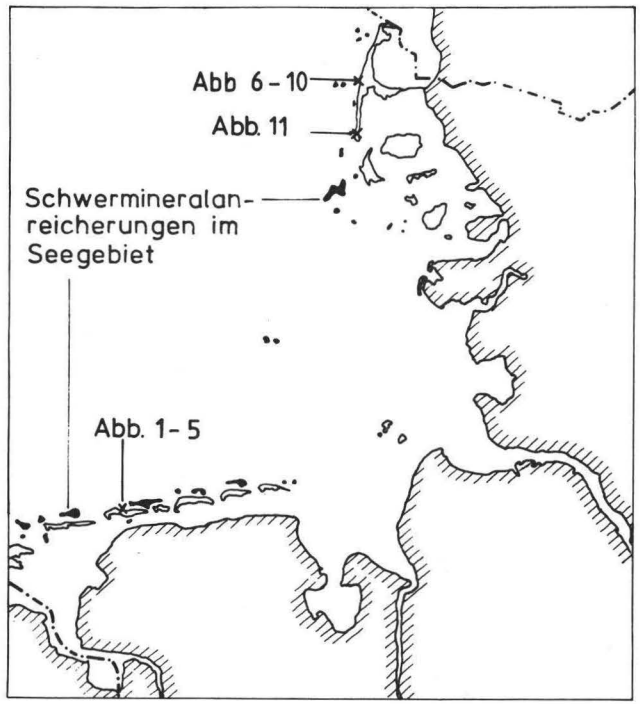


Abb. 19: Seegebiete mit Schwermineralgehalten von größer als 2 % nach Ludwig /2o/ An den Stränden nahe dieser Gebiete liegt eine erhöhte natürliche Gamma-Strahlung vor.

Fig. 19: Sea areas with heavy mineral concentrations of more than 2 %, see Ludwig /2o/. On the beaches near these areas an enhanced natural gamma radiation exists

NATURAL RADIATION BACKGROUND LEVELS IN THE NETHERLANDS AND MEASUREMENT DIFFICULTIES ENCOUNTERED

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SUMMARY. Results of measurements of the natural radiation background in the Netherlands by means of plastic scintillators are summarized. It appears that there is a good correlation between the type of soil, the activity concentration in the soil material and the radiation at 1 m above the ground in the open. Lowest levels were found on coarse sandy soils (1.7 μR/h, excluding cosmic radiation) and highest levels on sea clay (7.5 μR/h).
One of the difficulties encountered during the survey was a deviating response of the instrument for cosmic radiation. The causes of this effect are discussed. A few remarks will be made as to the possibilities of Compton recoil spectrometry for the measurement of the natural radiation background.

RESUME. NIVEAUX DE RAYONNEMENT NATUREL AUX PAYS-BAS ET DIFFICULTES DE MESURE EPROUVEES. Les résultats, rassemblés ici, des mesures du rayonnement naturel effectuées aux Pays-Bas à l'aide de scintillateurs plastiques montrent une bonne corrélation entre le type de sol, la concentration d'activité dans le matériau du sol, et le rayonnement à 1 m au-dessus du sol à l'air libre. Les plus bas niveaux proviennent de sables grossiers (1,7 μR/h cosmique exclu), et, les plus hauts, de l'argile marin (7,5 μR/h). Une des difficultés éprouvées a été une dérive de la réponse de l'appareil de mesure au rayonnement cosmique. Les causes en sont examinées. On mentionne les possibilités du spectromètre à recul Compton, pour mesurer le rayonnement naturel.

KURZFASSUNG. DER PEGEL DER NATÜRLICHEN UMGEBUNGSSTRAHLUNG IN DEN NIEDERLANDEN UND MESSTECHNISCHE PROBLEME SEINER ERMITTLUNG. Zusammenfassung von Ergebnissen der Messungen natürlicher Umgebungsstrahlung in den Niederlanden mit Kunststoffszintillatoren. Offensichtlich besteht eine gute Korrelation zwischen Bodentyp, Aktivitätskonzentration im Bodenmaterial und Strahlung im Freien in 1 m Höhe über Grund. Die niedrigsten Pegel wurden auf grobkörnigen Sandböden festgestellt (1,7 μR/h, ohne kosmische Strahlung), die höchsten auf Marschton (7,5 μR/h).

Zu den aufgetretenen Schwierigkeiten gehörte ein variierende Anzeige des Instruments für die Messung kosmischer Strahlung. Die Ursachen des Effekts werden erörtert. Kurz gestreift werden auch die Möglichkeiten der Compton-Rückstoss-Spektrometrie zur Messung der natürlichen Umgebungsstrahlung.

Measurements of the natural radiation background have been carried out in the Netherlands since 1966. The main objective of the survey was not so much to study the general very low radiation levels but to look for variations and to establish a relationship between these levels and the soil type.

When we started our work we used a cylindric all plastic scintillator (Nuclear Enterprice NE 105) of 5 inch height and 5 inch diameter coupled by means of lucite light collector to a low dark current photomultiplier tube (EMI 6097 S). The canning of the scintillator consisted of 0.5 mm Al and 2.5 mm MgO for increased reflectance. In order to reduce the dark current to an insignificant level the PM was cooled down to below $5^{\circ}C$.

The exposure rate can not only be estimated from the observed anode current after suitable calibration, but also from the pulse height spectra recorded at the output of the PM. As the interaction of gamma radiation in the scintillator is predominantly the Compton effect this has been termed Compton recoil spectrometry.

More recently we have restarted our investigation of natural radiation on a somewhat larger scale. In order to speed up the measurements the equipment described above was abandoned and replaced by scintillation monitors of the type developed in Western Germany. The detector consists of a 2" x 2" plastic scintillator together with the PM housed in a handheld probe. The associated electronics are mounted in a separate housing. As you are probably familiar with type of instrument I will not go into detail. The energy response, which is fairly good above about 100 keV was somewhat improved by coating the plastic with zinc sulphide. Calibration of the instrument was carried out with suitable radioactive sources of known intensity.

Let me now present some results.

Figure 1 shows a map of the area in the Netherlands where a survey has been

carried out. The regions were selected according to soil type and they cover the most important different soils. At all locations a number of readings were at a height of 1 meter above the ground. Due to statistical variations sufficient time was taken to allow for a good estimate. In spite of this procedure the accuracy of the measurements is estimated as of the order of 10%. The total of measurements, including travel time was about 20.
In order to avoid local disturbancies of the radiation field, care was taken to ensure that the distance to roads was at 50 meters and soil had at least superficially an undisturbed appearance.
Furthermore at all locations soil samples were taken at a depth of about 30 cm below the surface. These samples were dried in the laboratory subsequently overnight at a moderate temperature and weighted.
A known aliquot was transfered to a Sodiumiodide scintillation counter and the activity counted in a suitable energy interval usually between 100 - 500 keV or 100 - 2000 keV. No attempt was made to identify individual radionuclides such as potassium or isotopes from the natural uranium and thorium series. From the constant ratio of the selected intervals it was concluded that these were no important variations in nuclide composition independent of the origen of the sample.
In Figure 2 is shown a histogram of the results of exposure rate measurements.
It is evident that the variation in exposure rate is relative small.
The average value is 4.8 μR/h with a standard deviation of 1.8 μR/h.
The highest value was found on a clay type soil in Groningen 9.7 μR/h the lowest values were encountered on sandy soil in the centre of the country.
In Table 1 the results are summarized arranged according to soil type.

Soil types were taken from the official geological map and represent the estimates.

In Figure 3 the exposure rate is plotted versus the soil activity measured in the energy interval from 100 - 2000 keV.

In view of the uncertainties inherent in this type of work the agreement is fair. This meanes that a knowledge of the soil type enables one to estimate the exposure rate with reasonable accuracy.

It is interesting to note that the soil activity extrapolated to zero yield a value of 1.7 μR/h which is a direct measure of the cosmic-ray response.

Next to the field measurements some preliminary measurements of the exposure rate in houses were made. The aim of this survey is to estimate the actual radiation doses received by the population due to natural radiation in their environment. For that a selected group of persons were asked to wear during a period thermoluminescent dosimeters while at the same time the exposure rates in their homes and daily working surroundings were measured. In Figure 4 a histogram of the results is shown. Each point represents the average of at least 7 separate measurements in different parts of the houses.

It is clear that the values are rather low except in some cases where natural stone has been used. In general it was found that the radiation level in houses is about 30% higher than outside in agreement with experience obtained elsewhere. It is of some interest to notice that the correlation between the TLD results and the exposure measurements is rather low. This might be an indication that the radiation level inside houses is not a direct indicator for actual doses received by people. Further research is needed to confirm this conclusion.

Finally I should like to say something about the difficulties we encountered

during our work with scintillation monitors.

First of all due to the dark current of the photomultiplier the instrument shows a variable offset which introduces an error in the readings of the order of up to 5%. It is very difficult to compensate for this effect completely because of the large temperature variations encountered in field work. Combined with the statistical fluctuation the overall accuracy of the measurements in practice can be estimated as less than 10 to 20% which may be sufficient in most cases.

A more serious problem is that the respons for cosmic rays is definitely too low due to saturation effects in the PM. This can be demonstrated clearly in Figure 5 which shows the exposure-rate in μ R/h as a function of height over sea and over land.

It is evident that over sea the level found is about a factor 2 lower than the expected value of about 3.8 μR/h. The measured exposure rate at zero height is in agreement with the value found from soil measurements extrapolated to zero activity. Although a correction factor can be applied and cosmic radiation varies little, calibration procedures are much more difficult.

In order to overcome this problem we have recently built a new scintillation monitor where the anode current of the PM is measured as a series of pulses that can be discriminated against pulses too large (cosmic radiation) or too small (dark current). I will not go into technical details but show you only the block diagram and transfer caracteristic of the equipment in Figures 6 and 7 .

In this way we were able to reduce the contribution of cosmics rays to about 0.3 μR/h in the energy interval from 25 KeV to 2.7 MeV.

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Figure captions.

Fig. 1 Chart of the Netherlands with initial measurement areas.

Fig. 2 Histogram of 128 measurements of exposure-rate in the open field.

Fig. 3 Exposure-rate (μR/h) versus soil-activity (cpm/g 0-2 MeV).

Fig. 4 Histogram of 50 measurements of exposure-rate in dwellings.

Fig. 5 Exposure-rate (μR/h) versus altitude (feet) (over sea and over land).

Fig. 6 Block-diagram of single-channel exposure-rate monitor.

Fig. 7 Transfer-characteristic of single-channel exposure-rate monitor.

Deposit	Exposure rate (μR/h)	Soil activity (cpm/g)	
		0 - 0.5 MeV	0 - 2 MeV
Fine sands	2.4	0.27	0.36
Silty fine sands upon gravel	2.7	0.35	0.45
Peat	2.8	0.26	0.38
Coarse sands	3.0	0.32	0.41
Clay/Peat	4.8	1.11	1.38
Clay upon coarse sands	5.7	0.95	1.19
Clay upon gravel	6.3	0.79	0.99

Table 1 Average exposure-rate for 7 different soils, average soil-activity

THE NETHERLANDS NATURAL BACKGROUND RADIATION
INITIAL MEASUREMENT AREAS

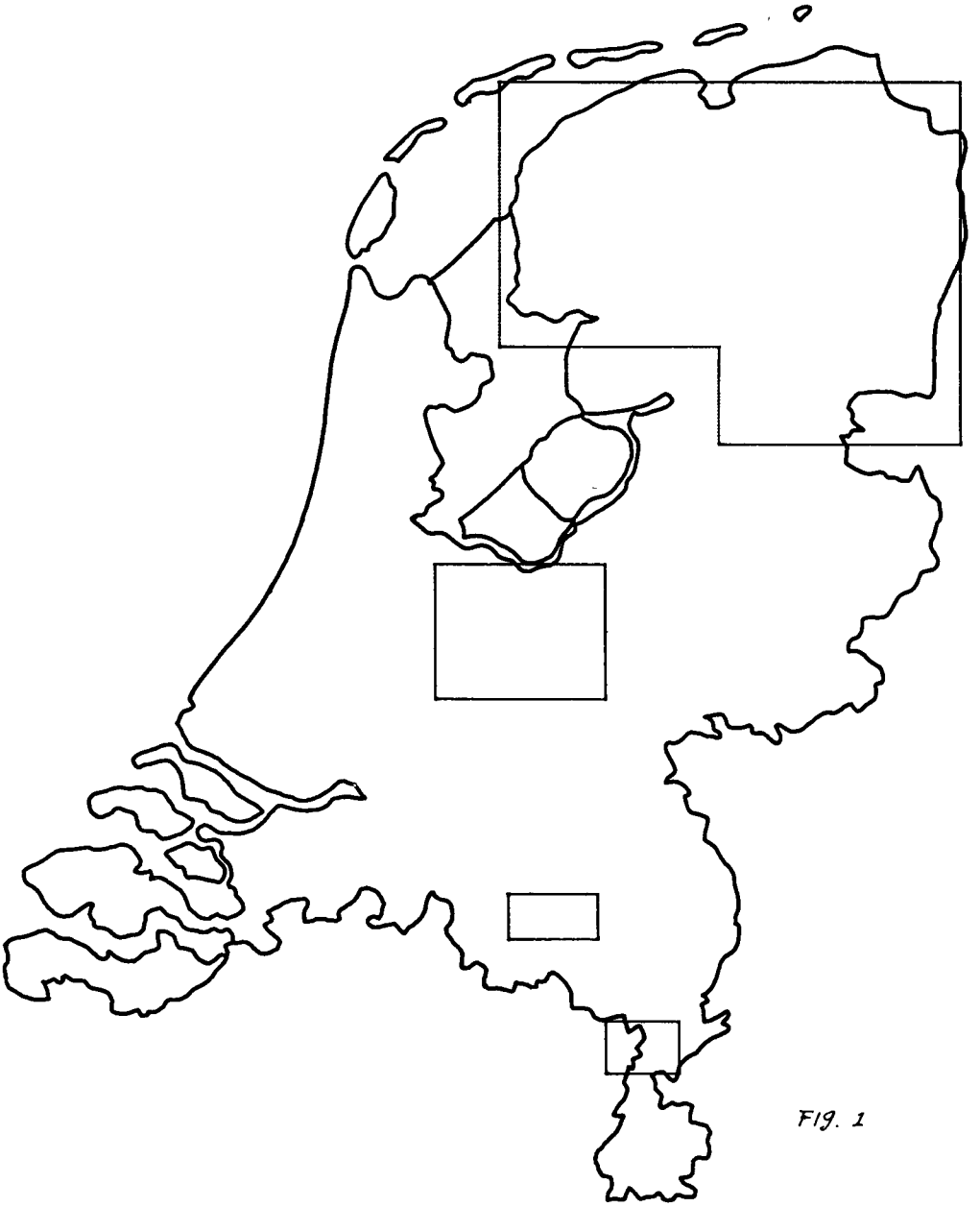

Fig. 1

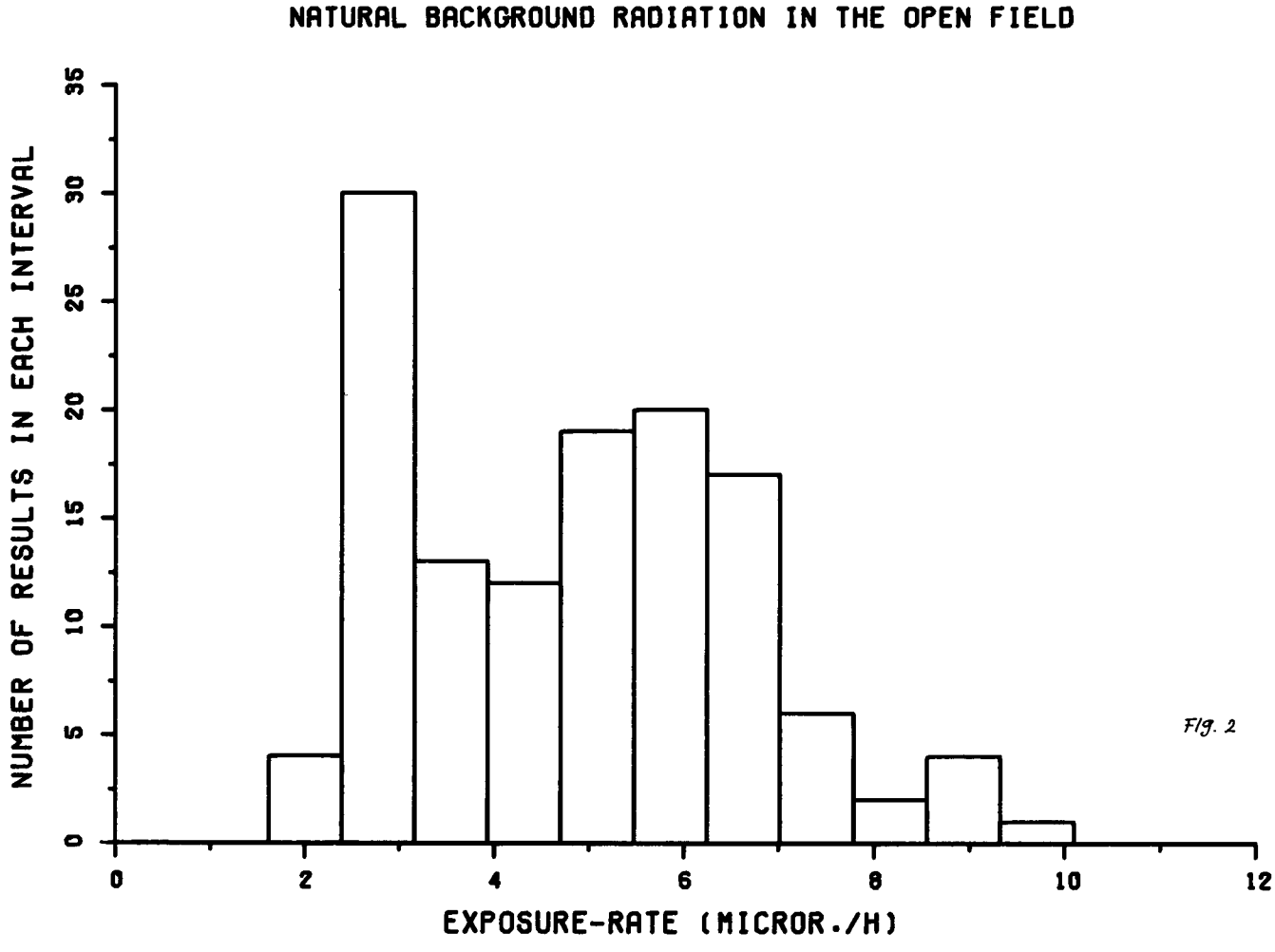
NATURAL BACKGROUND RADIATION IN THE OPEN FIELD
NUMBER OF RESULTS IN EACH INTERVAL
0
5
10
15
20
25
30
35
0
2
4
6
8
10
12
EXPOSURE-RATE (MICROR./H)

Fig. 2

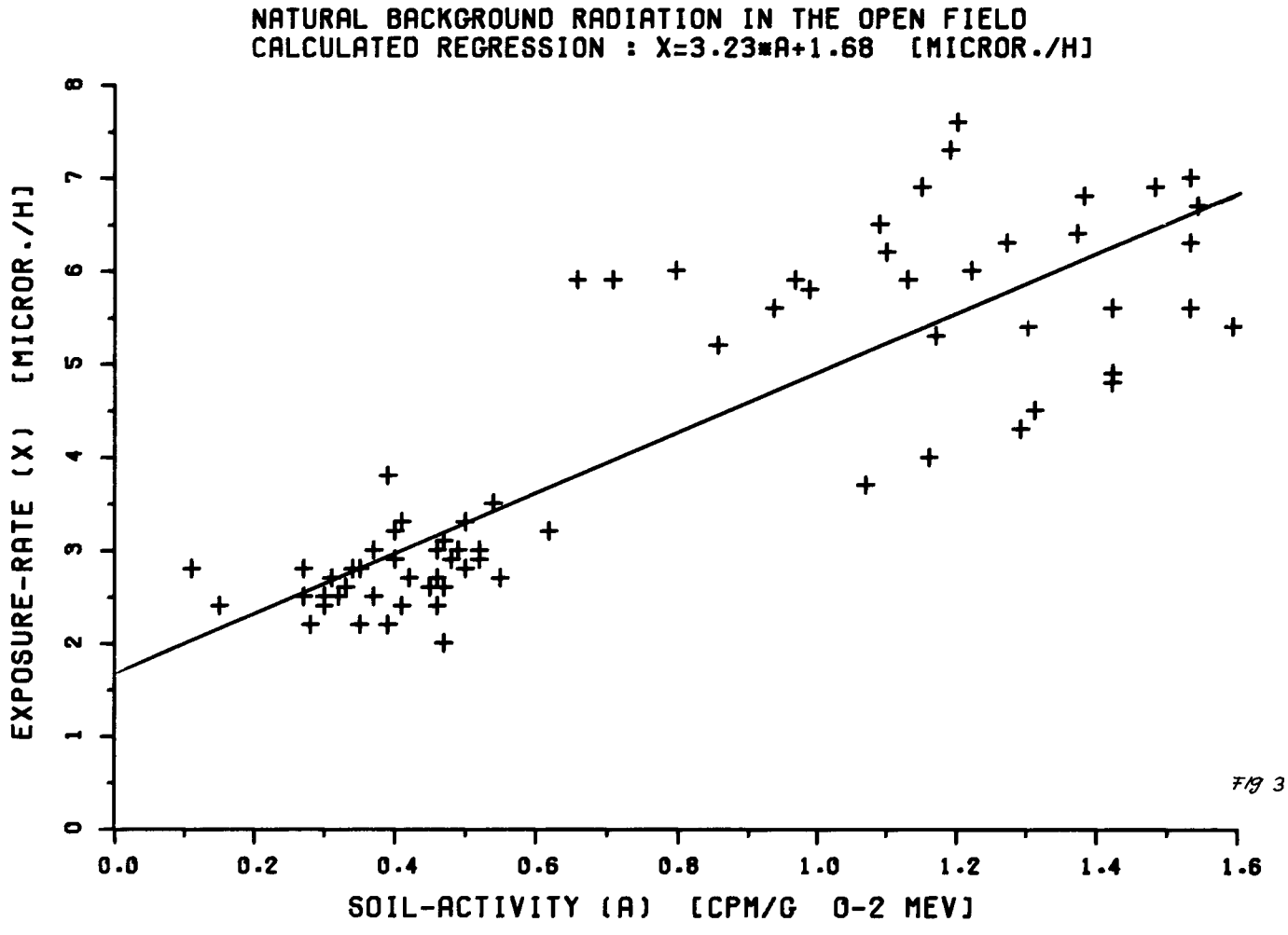
NATURAL BACKGROUND RADIATION IN THE OPEN FIELD
CALCULATED REGRESSION : X=3.23*A+1.68 [MICROR./H]
EXPOSURE-RATE (X) [MICROR./H]
0
1
2
3
4
5
6
7
8
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
SOIL-ACTIVITY (A) [CPM/G 0-2 MEV]

Fig 3

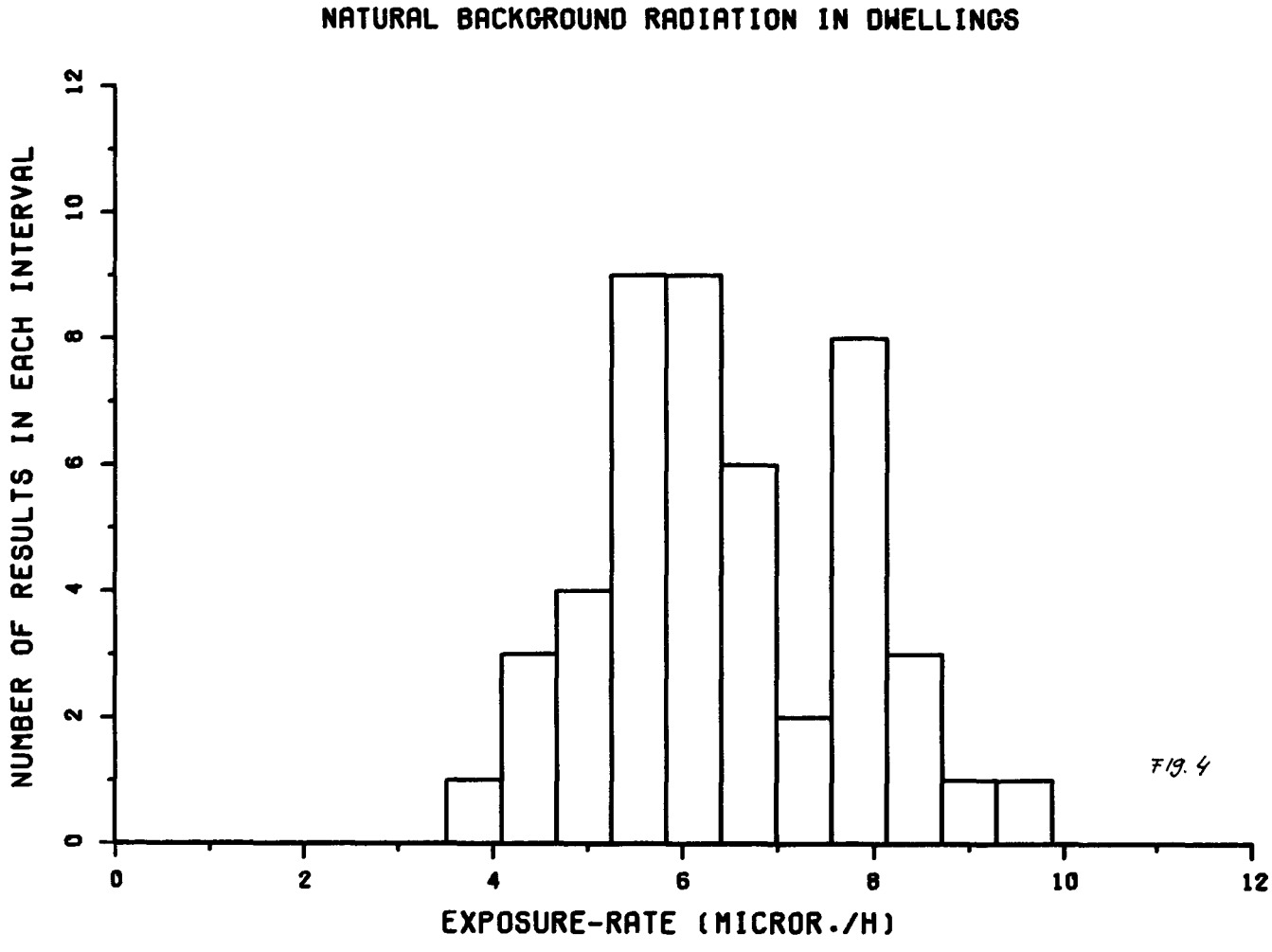
NATURAL BACKGROUND RADIATION IN DWELLINGS
NUMBER OF RESULTS IN EACH INTERVAL
0
2
4
6
8
10
12
EXPOSURE-RATE (MICROR./H)
0
2
4
6
8
10
12

Fig. 4

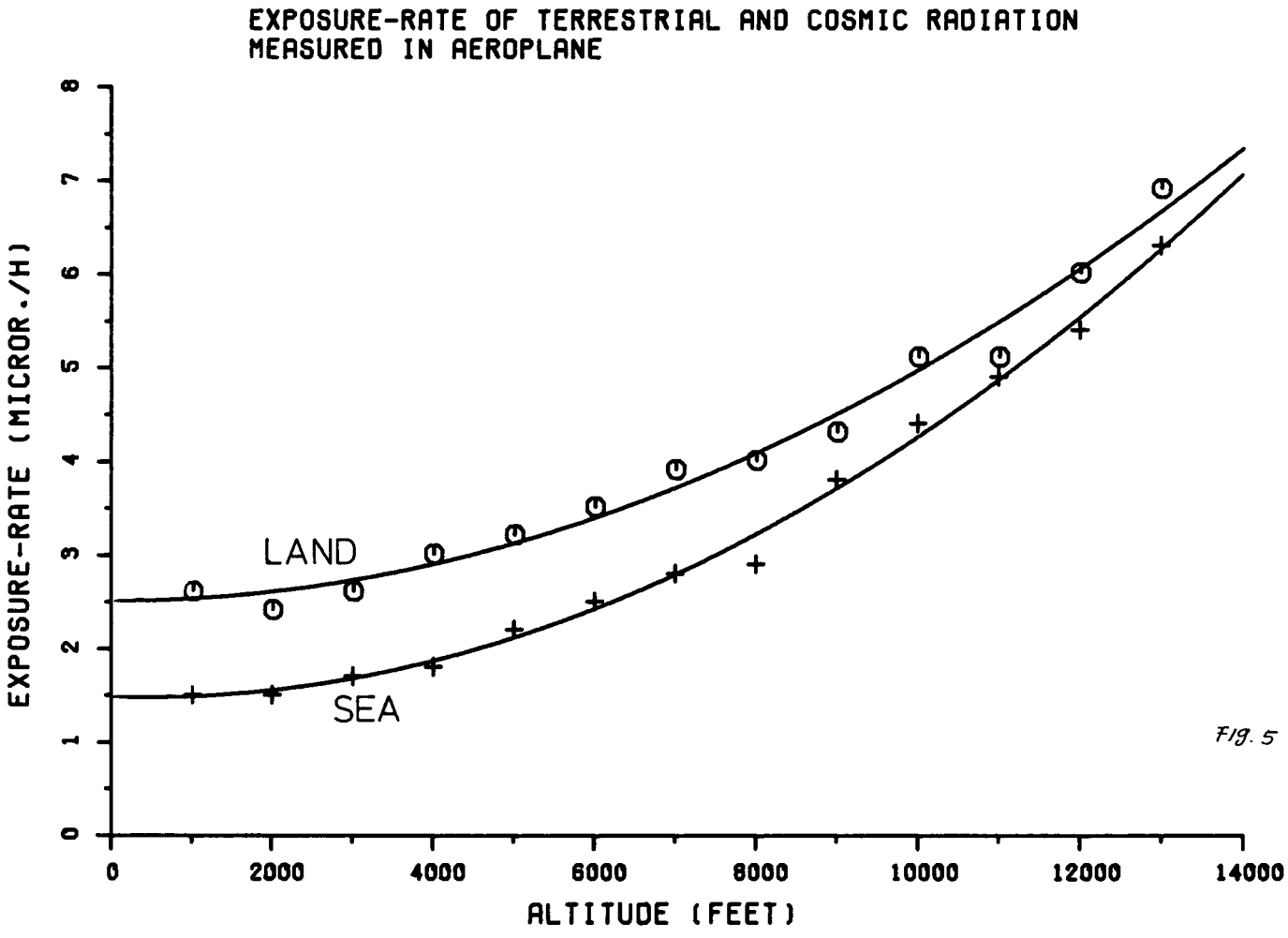
EXPOSURE-RATE OF TERRESTRIAL AND COSMIC RADIATION
MEASURED IN AEROPLANE
EXPOSURE-RATE (MICROR./H)
0
1
2
3
4
5
6
7
8
LAND
SEA
0
2000
4000
6000
8000
10000
12000
14000
ALTITUDE (FEET)

Fig. 5

BLOCK-DIAGRAM SINGLE-CHANNEL EXPOSURE-RATE MONITOR

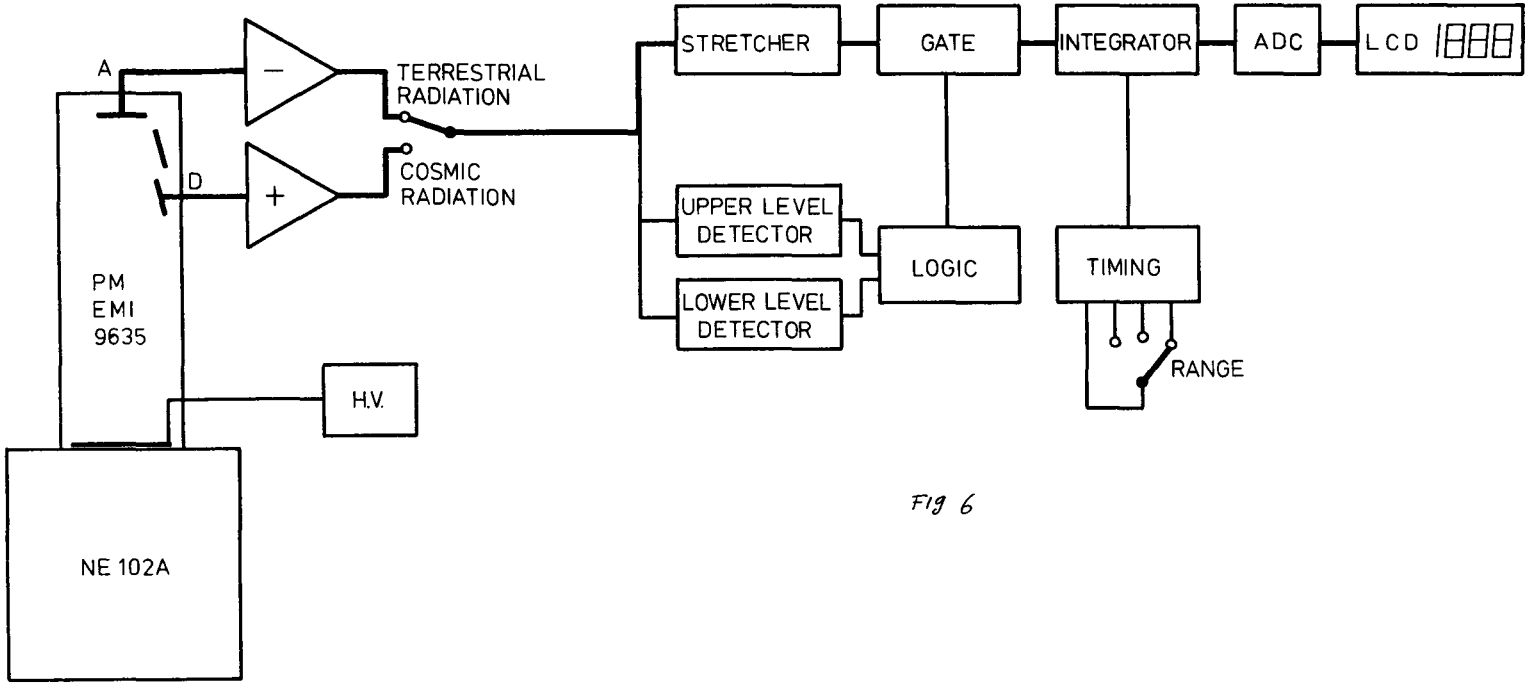

Fig 6

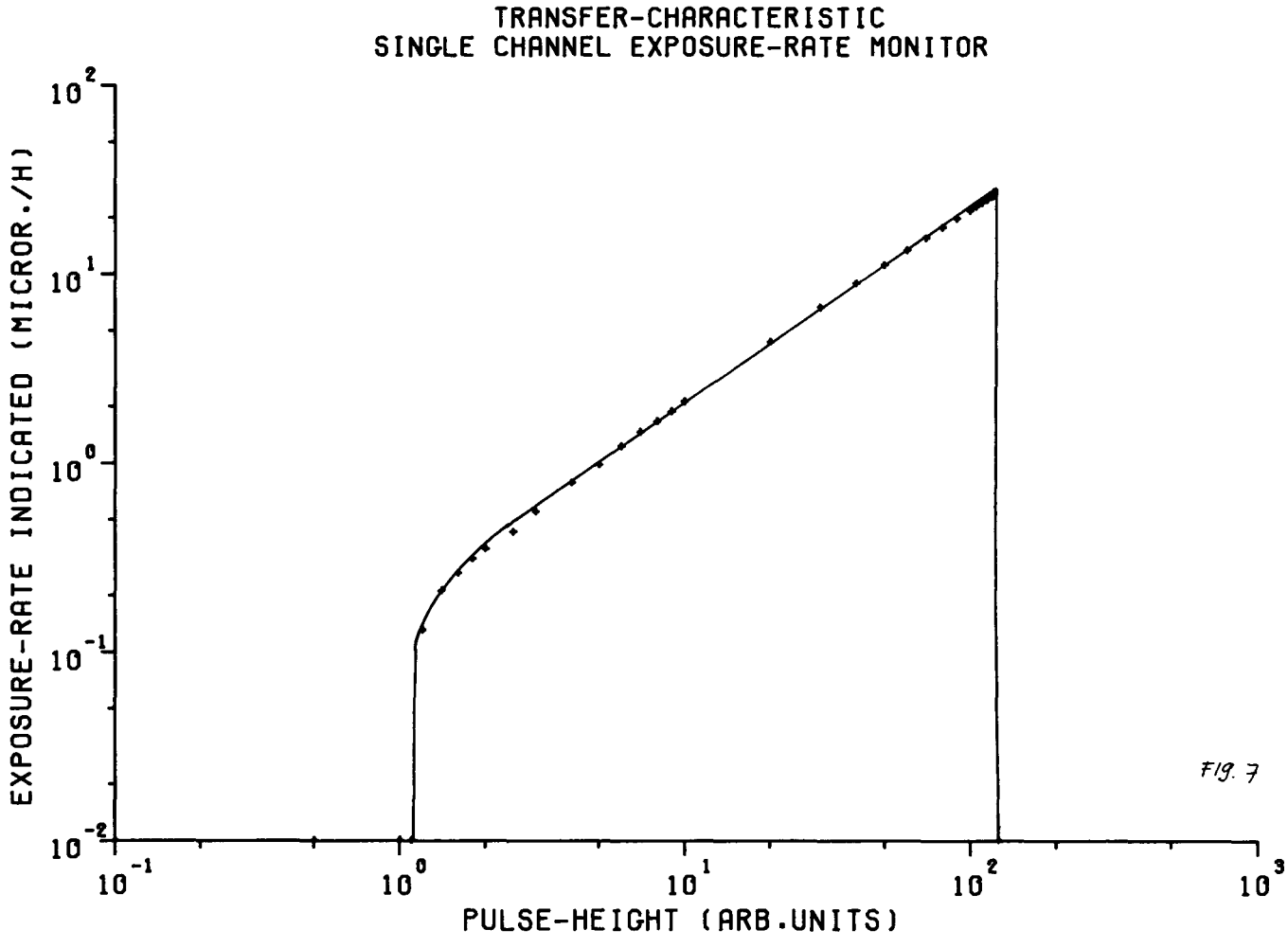
TRANSFER-CHARACTERISTIC
SINGLE CHANNEL EXPOSURE-RATE MONITOR
EXPOSURE-RATE INDICATED (MICROR./H)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
10^{3}
PULSE-HEIGHT (ARB.UNITS)

Fig. 7

EXTERNAL RADIATION OF TERRESTRIAL ORIGIN IN THE REPUBLIC OF IRELAND

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SUMMARY. Measurements of external radiation levels have been made at almost 300 points in the Republic of Ireland using a high pressure ionisation chamber. The results are presented to show the variation in levels produced by underlying geological strata of different compositions. The consequences of this variability in radiation levels in terms of the contributions to collective dose equivalent in different parts of the country are estimated and compared with some other sources of variability in dose equivalent.

RESUME. RAYONNEMENT EXTERNE D'ORIGINE TERRESTRE EN REPUBLIQUE D'IRLANDE. On a mesuré à l'aide d'une chambre d'ionisation à haute pression les niveaux de rayonnement extérieur en près de 300 points en république d'Irlande. Les résultats sont présentés de manière à montrer la variation des niveaux selon les couches géologiques sousjacentes de différentes compositions. Les conséquences de cette variabilité des niveaux de rayonnement en termes des contributions à l'équivalent de dose collective dans différentes régions du pays sont estimées et comparées à d'autres sources de variabilité de l'équivalent de dose.

KURZFASSUNG. AUSSERE STRAHLUNG TERRESTRISCHEN URSPRUNGS IN DER REPUBLIK IRLAND. Der Pegel der äusseren Strahlung wurde an nahezu 300 Orten der Republik Irland mit einer Hochdruck-Ionisationskammer gemessen. Aus der Darstellung der Ergebnisse ist ersichtlich, inwieweit die Pegelschwankungen durch geologische Unterschiede des Untergrundes verursacht werden. Die Auswirkungen der verschieden hohen Strahlenpegel auf ihren Anteil an der Kollektiv-Äquivalentdosis in verschiedenen Landesteilen werden abgeschätzt und mit einigen anderen Ursachen für die Schwankung des Dosisäquivalents verglichen.

Measurement of the environmental gamma radiation levels have been made at 264 locations throughout the Republic of Ireland.

The measurements were taken using a Reuter-Stokes Environmental Monitor, type RSS-111, which consists of a high pressure ionization chamber with a digital readout and also with a strip chart recorder providing a complete record of the gamma radiation level as the instrument was transported around the country by road. The readings were taken with the detector held one metre above ground at points which were close to roads or tracks but were several metres away from the actual road material. The background radiation level due to cosmic radiation was found by taking the measuring instrument out in a boat to a point where the water was in excess of 10 m deep.

The average of all readings taken throughout the country corresponds to a dose of 0.72 milligray per annum (72 mR/yr) and the range is from 0.35 to 1.95 mG/annum, all inclusive of cosmic ray background. When the cosmic ray background of 0.35 mG/yr is taken into consideration it can be seen that the terrestrial component of the gamma flux ranges from zero to 1.6 mG/annum. This result is not in itself surprising and indicates that Ireland is at the lower end of the levels previously reported for European countries which range from 0.27 mG/annum for the Netherlands (1) to 0.8 mG/annum for the G.D.R. (2) for radiation of terrestrial origin.

The lowest readings in the survey were found over the central limestone plain of Ireland (Fig. 1) where the average of 78 readings gave a value of 0.59 mG/annum inclusive of cosmic ray background.

The highest readings in the survey were found along a line of intrusions of volcanic origin extending across the South-East corner of the country (Fig. 1). The average of 12 readings taken over these intrusions gave a value of 1.24 mG/annum for total gamma ray background.

Further work is at present in progress to determine the exact extent of these regions of high natural background, but from the readings of the strip chart recorder, it seems likely that the population resident within the high level zones will be between 10,000 and 20,000.

Precise data are not available for the genetically significant dose in Ireland as a result of X-ray diagnostic and clinical procedures, but it is not likely to exceed that applicable to the United Kingdom and a figure of 0.1 mG/annum would appear reasonable. The whole body average dose due to naturally occurring radioisotopes may be taken as 0.17 mG/annum (2).

The average total dose within Ireland is therefore close to 1 mG/annum and by far the largest source of variation in this figure will be the natural radiation of terrestrial origin.

Some preliminary work has been carried out on the ratio of indoors to outdoors dose rates which appears in many cases to be close to one. However, in many parts of rural Ireland, house construction is of natural stone and it is not yet possible to identify the range of variables.

The main source of the increased level of radiation in the "high" areas is identified as potassium, though detectable quantities of uranium have also been found.

Future work will include measurements of the radon content of the indoors atmosphere and assessments of the radioactive content of local foods, particularly fish originating from the Irish Sea.

This work has been supported by the Association Euratom - CEA and by the Commission of the European Communities.

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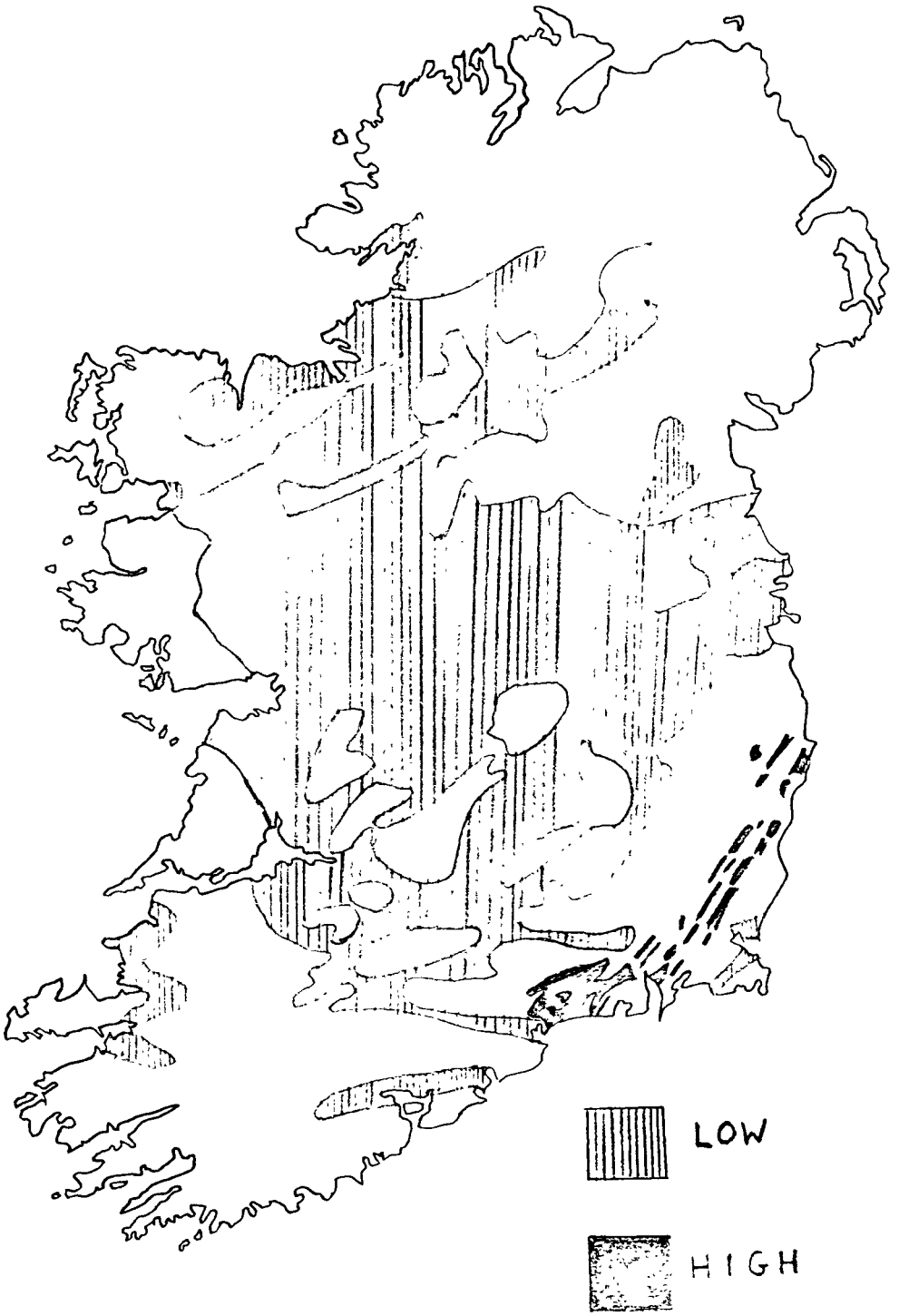
1. A. Biau and J.P. Moroni - A review of existing information on natural radioactivity in Europe (S.C.P.R.I. Luxembourg 1977).

2. United Nations Scientific Committee on the Effects of Atomic Radiation. Report 1977.


LOW
HIGH

TERRESTRIAL AND COSMIC RADIATION IN DENMARK

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SUMMARY. Measurements of the background radiation in Denmark were made with a mobile gamma-spectrometer system and a high pressure ionization chamber. An average value of the terrestrial gamma-ray exposure rate of 4.4 /u R/h was found and a value of 3.6 /u R/h was estimated for the sea level exposure rate from cosmic radiation at an atmospheric pressure of 760 mm Hg, with a value of -4.5% per cm Hg for the total barometric coefficent.

RESUME. RAYONNEMENTS TERRESTRE ET COSMIQUE AU DANEMARK. On a mesuré au Danemark le rayonnement naturel à l'aide d'un sytème de spectromètres gamma mobiles et d'une chambre d'ionisation à haute pression. En moyenne le débit d'exposition au rayonnement terrestre gamma était de 4,4 /u R/h et la valeur du débit d'exposition au rayonnement cosmique a été estimée à 3,6 /uR/h au niveau de la mer pour une pression atmosphérique de 760 mm Hg, avec -4,5% par cm Hg pour coefficient barométrique total.

KURZFASSUNG. TERRESTRISCHE UND KOSMISCHE STRAHLUNG IN DÄNEMARK. Die Messungen der natürlichen Strahlung in Dänemark wurden mit einem beweglichen Gamma-Spektrometer-System und einer Hochdruck-Ionisationskammer durchgeführt. Es wurde ein mittlerer Expositionspegel aus terrestrischer Gamma-Strahlung von 4,4 /u R/h festgestellt. Der Pegel der kosmischen Strahlung in Meereshöhe wurde bei einem Luftdruck von 760 mm Hg zu 3,6 /u R/h abgeschätzt mit einem Druckkoeffizienten von -4,5% pro cm Hg.

1. INTRODUCTION

The penetrating background radiation is composed of a cosmic-ray component and a terrestrial γ-ray component. The cosmic component at ground level (the secondary cosmic rays) is produced when primary cosmic rays interact with atomic nuclei in the upper atmosphere. The terrestrial component is due to the naturally occurring radionuclides and the man-made radionuclides present in the environment.

Both components are subject to variations. The cosmic component is attenuated in the atmosphere, and the thickness of the atmospheric layer (measured in terms of atmospheric pressure) consequently influences the radiation intensity at ground level. The terrestrial component arises mainly from the γ-emitting radionuclides in the top soil layers. Precipitation is a principal cause of variation due to, for example, increased soil humidity, snow cover or wash-out of airborne radon daughters.

This paper presents results of measurements made in Denmark 1978 of the two background components.

2. FIELD MEASUREMENTS

The measurements were made with a high pressure ionization chamber and a mobile Ge(Li) spectrometer system which made it possible to distinguish between ground level exposure rates from cosmic radiation and from terrestrial γ-radiation.

The ionization chamber[1)] is a Reuter-Stokes model RSS-111; it records the total background exposure rate on a magnetic tape unit permitting an assessment of the time variation of the exposure rate. The Ge(Li) spectrometer[2)] registers γ-radiation and gives an assessment of exposure rates from the individual γ-emitting radionuclides. Measurements of the atmospheric pressure were made with an aneroid barometer.

The locations for the measurements are shown in Fig. 1. The individual sites were selected in order that a good approximation to a 2π-geometri would be obtained, as this idealization is assumed when the γ-ray spectra are interpreted quantitatively. At most of these locations the soil was cultivated, leaving the aged fallout uniformly distributed in the plowing layer (0-20 cm). This contrasts with uncultivated soil where the fallout concentration decreases nearly exponentially with depth. At each location two measurements were made at the same site with a time interval of three months.

Washout of radon daughters from the atmosphere was observed on several occasions when measurements were made during rainfall. The effect was seen from the γ-spectra, which displayed unusually large radon daughter contributions and from the ionization chamber measurements, showing significant short-term variations of the total background exposure rate.

3. RESULTS AND DISCUSSION

From the recorded γ-spectra the contributing γ-emitting radionuclides were identified and the corresponding concentrations in the soil estimated. These concentrations yielded the basis for estimating the ground level exposure rates using conversion factors derived from computer calculations of γ-ray transport from soil to air[3,4]. The sum of the individual radionuclide exposure rates yielded the total terrestrial exposure rate. The total background exposure rate was recorded by the high pressure ionization chamber, and an average value was then calculated for the measuring period of the γ-spectrometer. An estimate of the cosmic-ray induced exposure rate was subsequently calculated as the difference between the total background exposure rate and the total terrestrial exposure rate.

The estimated terrestrial exposure rates from the field γ-spectroscopic measurements are presented in Table I, where each value is an average of two measurements. It is noted that the

contribution from the naturally occurring radionuclides varies significantly throughout the country with the low values in the western part of Jutland and the high values on the island of Bornholm. This pattern reflects the geological origin of the top soil. As for the contribution from fallout (^{137}Cs) the two locations, Studsgård and Skydebanen, distinguish themselves from the rest; here measurements were made over uncultivated soil. At Skydebanen the undisturbed exponential distribution of ^{137}Cs leaves larger amounts of deposited source material closer to the soil surface than in the case of cultivated soil, and at Studsgård this effect is further enhanced due to vegetation of heather, which accumulates fallout like lichen, although not to the same extent.

In Fig. 2 the total terrestrial exposure rates from Table I are depicted versus the total background exposure rates obtained by the ionization chamber. The observations are correlated with a highly significant correlation coefficient of 0.99 and are fitted with a line of regression of the type $y = \alpha(x-\beta)$, where $\alpha = 1.03 \pm 0.03$ (1 SD) μRh^{-1} per μRh^{-1} and $\beta = 3.8 \pm 0.1$ (1 SD) μRh^{-1}. The observed slope of 1.03 μRh^{-1} per μRh^{-1} indicates that the field γ-spectrometer system yields acceptable estimates of the terrestrial exposure rate, and the zero intercept of 3.8 μRh^{-1} is interpreted as the cosmic-ray contribution to the background radiation. Several effects cause the observations to scatter around the regression line. The model assumptions used for the interpretation of the γ-ray spectra are never completely fulfilled, for example, variations of soil composition, source distribution and measuring geometry are bound to occur. Furthermore, variations of atmospheric pressure influence the cosmic-ray contribution, so that observations made under conditions of high atmospheric pressure are situated to the left of the regression line, and conditions of low atmospheric pressure are to the right.

The variation of cosmic-ray exposure rate versus atmospheric pressure is shown in Fig. 3. The estimates of cosmic-ray exposure rate are correlated to the measurements of atmospheric

pressure with a highly significant correlation coefficient of -0.69. A regression line of the type $y = \beta + \alpha(x-760)$ yields for the coefficients $\alpha = 0.016 \pm 0.003$ (1 SD) μRh^{-1} per mm Hg and $\beta = 3.56 \pm 0.03$ (1 SD) μRh^{-1}. The latter value of 3.56 μRh^{-1} is in good agreement with the value of the total cosmic-ray ionization in air at sea level adopted by UNSCEAR[5] and the observed variation with pressure (-4.5% per cm Hg) agrees well with published values of the total barometer coefficient[6]

4. CONCLUSION

Simultaneous measurements with a γ-spectrometer system and a high pressure ionization chamber have yielded accurate estimates of terrestrial γ-ray exposure rates - including contributions from naturally occurring radionuclides and deposited fallout - and of ground level exposure rates from cosmic radiation. A value of 3.6 μRh^{-1} was obtained for the latter corresponding to an atmospheric pressure of 760 mm Hg; furthermore a value of -4.5% per cm Hg was obtained for the total barometer coefficient.

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5) UNSCEAR. 1977. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. (New York) 725 pp.

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TABLE I

Terrestrial exposure rates estimated from field γ-spectroscopic measurements (μRh^{-1})

Location	^{40}K	^{238}U series	^{232}Th series	^{137}Cs	Total
Tylstrup	1.9	0.8	0.8	0.1	3.6
Ødum	2.1	0.8	1.1	0.1	4.2
Studsgård	0.7	0.4	0.6	0.8	2.5
Askov	1.4	0.8	0.9	0.2	3.2
Jyndevad	1.0	0.5	0.3	0.1	1.9
Blangstedgård	2.3	0.9	1.4	0.1	4.6
Rısø	2.4	0.9	1.5	0.1	5.0
Skydebanen	1.9	0.7	1.1	0.3	3.9
Ledreborg	2.3	1.0	1.5	0.1	4.9
Tystofte	2.2	1.2	1.3	0.1	4.9
Abed	2.3	1.1	1.7	0.1	5.1
Tornbygård	2.5	1.0	1.7	0.1	5.4
Rødbjergkilde	2.1	1.9	1.8	0.1	5.9
Gudhjem	2.7	1.1	2.1	0.1	6.0

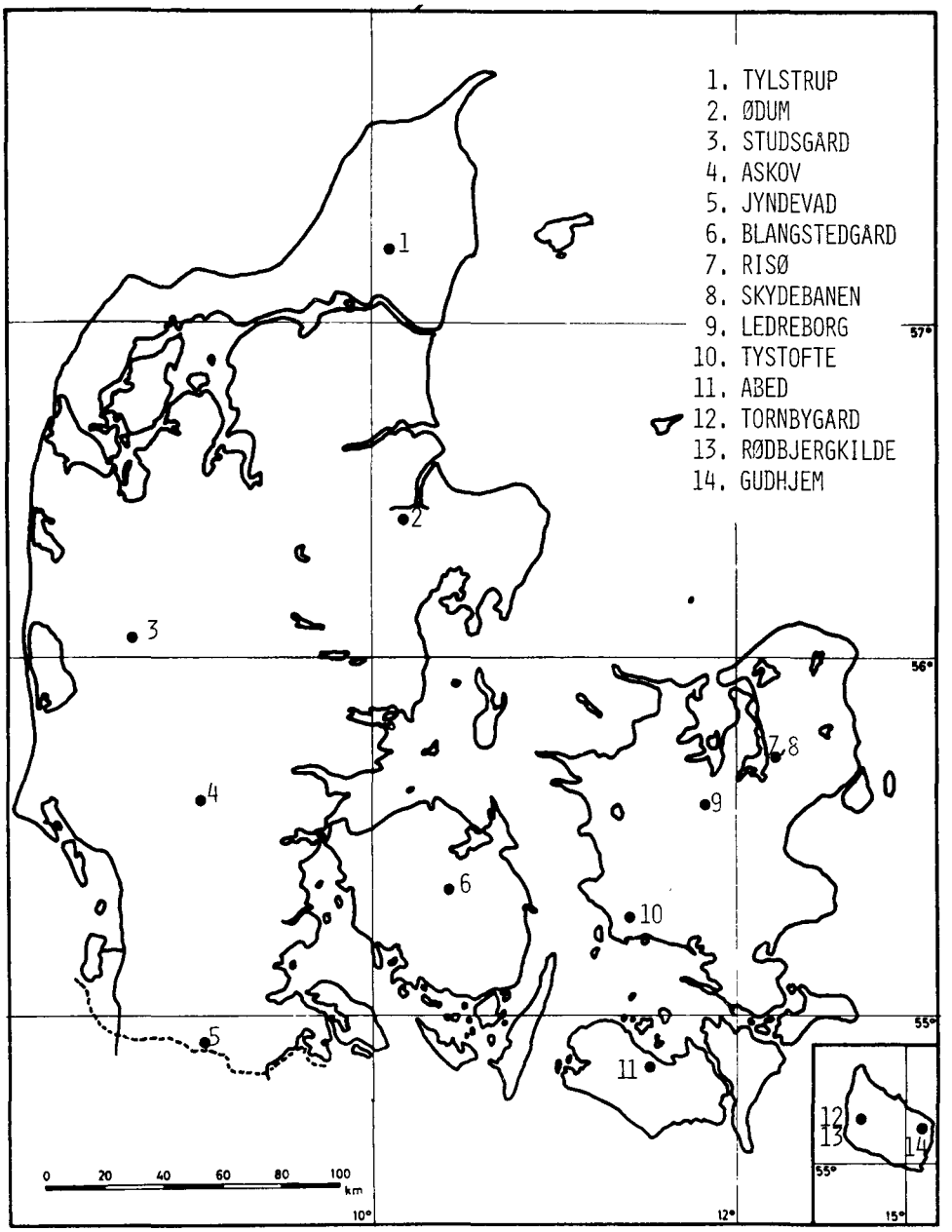

Fig. 1. Locations for measurements.

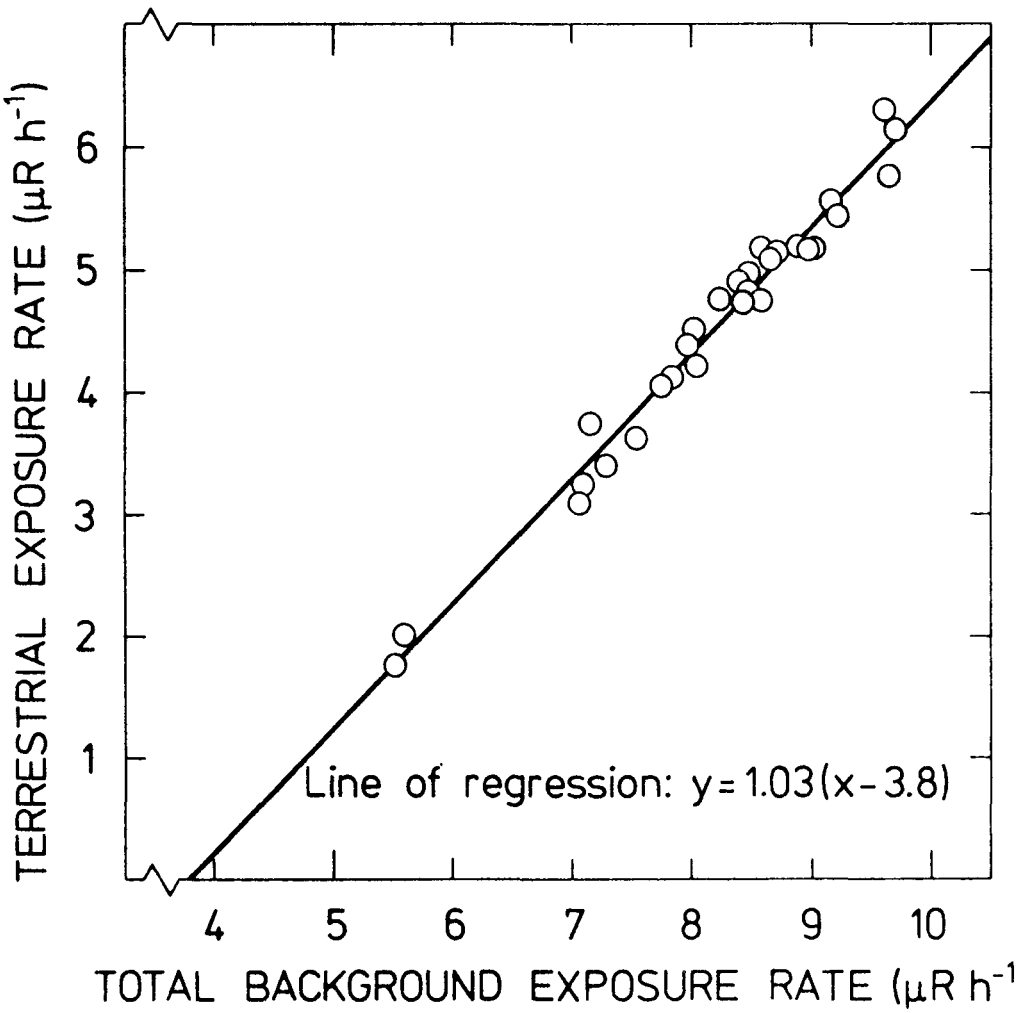

Fig. 2. Terrestrial exposure rate (μRh^{-1}) estimated from field γ-spectroscopic measurements versus total background exposure rate (μRh^{-1}) measured with a high pressure ionization chamber.

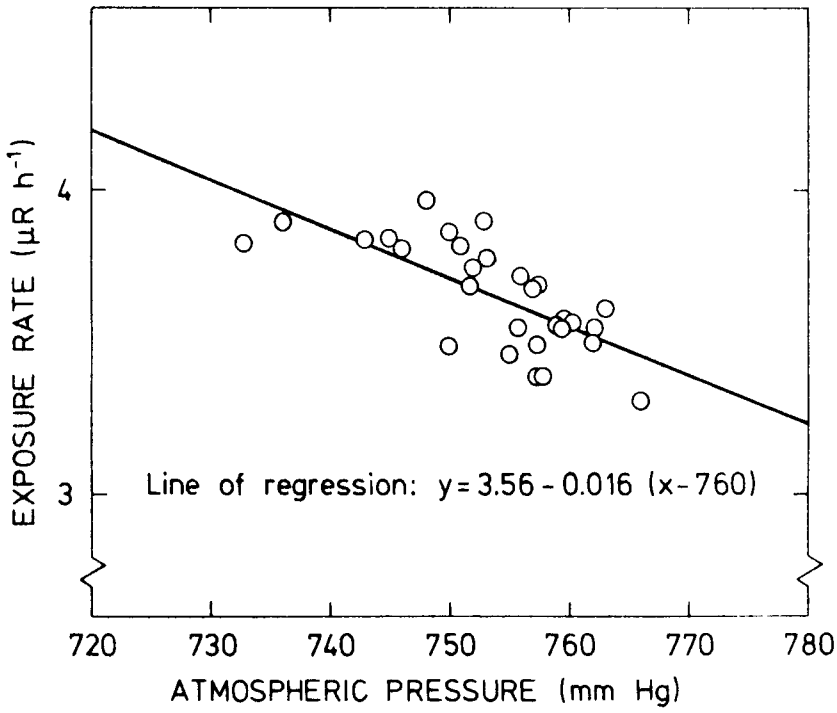

Fig. 3. Estimated cosmic-ray exposure rate (μRh^{-1}) at ground level versus atmospheric pressure (mm Hg).

GAMMA EXPOSURE LEVELS FROM NATURAL RADIOACTIVITY IN GREENLAND

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The Geological Survey of Greenland
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SUMMARY. An extensive radiation monitoring programme was carried out at Kvanefjeld, South Greenland, where elevated exposure rate levels of several hundred /uR/h result from extensive uranium and thorium mineral deposits. To study the relation between exposure rate and Th-U-K surface concentrations a number of field locations were monitored using LiF thermoluminescence dosimeters, a high-pressure ionization chamber and a portable gamma-ray spectrometer. These measurements were accompanied by calibration experiments and a two-media gamma-ray transport calculation. Reliable factors for the conversion of a set of surface concentration values into a gamma-ray exposure rate were established. Futhermore, these factors were used to estimate the exposure levels at other locations in Greenland based on geological prospecting data. The data-recorded radiation exposure levels, make it possible to estimate external gamma radiation doses for the most populated areas in Greenland.

RESUME. NIVEAU D'EXPOSITION GAMMA DUE A LA RADIOACTIVITE NATURELLE AU GROENLAND. Un vaste programme de surveillance radiologique a été mené à Kvanefjeld, Sud Groenland, où de vastes gîtes d'uranium et de thorium engendrent des débits de dose de plusieurs centaines de /uR/h. Pour étudier la relation liant le débit d'exposition aux concentrations de Th-U-K en surface, un certain nombre de sites ont été équipés de dosimètres à thermoliminescence LiF, d'une chambre d'ionisation à haute pression et d'un spectromètre gamma portable. Les mesures ont été complétées par des étallonnages et par un calcul de transport de rayon gamma à 2 milieux. On a établi des facteurs fiables de conversion des concentrations en surface en débit d'exposition gamma. En outre, ces facteurs ont servi à estimer les niveaux d'exposition dans d'autres zones du Groenland partant de données acquises lors de prospection géologique.
Les niveaux relevés d'exposition au rayonnement permettent d'estimer les doses gamma externes pouvant être reçues dans les zones les plus peuplées du Groenland.

KURZFASSUNG. GAMMA-EXPOSITIONSWERTE ZUFOLGE NATÜRLICHER RADIOAKTIVITÄT IN GRÖNLAND. Ein umfassendes Strahlenüberwachungsprogramm wurde in Kvanefjeld , Südgrönland, durchgeführt, wobei, wegen ausgedehnter Uran- und Thoriumvorkommen, erhöhte Expositionspegel von mehreren hundert /uR/h beobachtet wurden. Die Beziehung zwischen den Expositionspegeln und den Th-U-K-Oberflächenkonzentrationen wurde mit LiF-Thermolumineszenzdosimetern, einer Hochdruck-Ionisationskammer und einem tragbaren Gamma-Spektrometer an mehreren Aufpunkten untersucht. Parallel zu diesen Messungen wurden Eichmessungen sowie eine Zweimedien-Gammastrahl-Transportrechnung durchgeführt. Zuverlässige Faktoren für die Umrechnung einer Reihe von Oberflächenkonzentrationswerten in einen Gamma-Expositionspegel wurden bestimmt. Diese Faktoren wurden herangezogen, um die Expositionspegel an anderen Orten mit hoher Radioaktivität in Grönland, nach Prospektionsmessungen ausgewählt, zu ermitteln.
Die festgestellten Expositionspegel ermöglichen eine Abschätzung der aus äusserer Gammabestrahlung in den bewohnten Gegenden Grönlands empfangenen Dosen.

Introduction

Extensive outcrops of radioactive rock are found on the Kvanefjeld plateau near the town of Narssaq, South Greenland. These rocks, which form the northern most part of the Ilimaussaq alkaline intrusion (see Fig. 1), typically contain 1000 parts per million thorium and 300 parts per million uranium. They continue at depth and represent a major low-grade uranium resource.

Many thousand square meters of the terrain surface have gamma-ray exposure rates of several hundred microroentgens per hour, and in places more than 1000 μR/h can be observed. Because of potential mining activity, prolonged human exposure to this anomalous radiation environment can be foreseen. The radiation levels in the area and its surroundings are thus being studied rather closely.

A monitoring programme of environmental gamma exposure rates was carried out on the Kvanefjeld plateau in five typical areas, varying from low-level to high-level radioactivity. Furthermore measurements were made at a number of places in Narssaq and surroundings which represent the daily work-, rest- and living areas of workers[1].

About 5000 determinations of the radioelement concentrations in the terrain surface are available from exploration surveys with portable gamma-ray spectrometers. There has been some interest in interpreting these data in radiological terms. To study the relation between exposure rate and radioelement surface concentrations, the field locations were monitored with thermoluminescence dosimeters (TLD), a high pressure ionization chamber, and a portable gamma-ray spectrometer. These measurements were accompanied by calibration experiments and a two-media gamma-ray transport calculation.

Evaluation of numerical constants made it possible to convert a set of surface concentration values into a gamma-ray exposure rate 1 meter above ground level. Furthermore the mean exposure level of the approx. 120 km^2 Ilimaussaq intrusion was estimated on the basis of the relative aboundance of the various intrusive rocks and their concentrations of radioelements[2]. Finally the conversion factors were used to predict the gamma-ray exposure rates at other places in Greenland where the radioelement concentrations have been determined.

Field sites and geology

The field sites outside Kvanefjeld where the exposure rates have

been calculated are given in the map in Fig. 2 and Table I gives the geological setting of the areas in short form. A more thorough geological description of each area is given by Escher and Watt[3].

The Nordre Strømfjord and Søndre Strømfjord areas are dominated by gneiss, and the calculated exposure rates are based on analysis of the average radioelement concentration of the gneiss. The Sarfartoq carbonatite complex (90 km^2), which is situated within the Søndre Strømfjord area is attractive for tourists and it is easily reached from the nearby Søndre Strømfjord airport. The geological data is given by Secher and Larsen[4].

The Godthåb, Fiskenæsset and Frederikshåb areas are the most populated in Greenland. The exposure rates from these areas have been estimated on the basis of radioelement contents in composite sand samples[5]. It has been shown that composite sand samples in Greenland to a high degree reflect the local bedrock composition[6].

The exposure rates from Scoresby Sund, inner and outer zones, are calculated from radioelement contents in samples collected during an initial uranium exploration programme in the areas[7,8]. Mineralized samples are not included in the calculations.

Field measurements

The measurements on Kvanefjeld were made with the following detectors: 1) thermoluminescence dosimeters (LiF TLD 700, Harshaw), 2) a high-pressure ionization chamber (Environmental Monitoring System RS 111, Reuter Stokes) and 3) a portable gamma-ray spectrometer equipped with a 3 x 3 inch NaI crystal (Geometrics Exploranium model GR-410).

At each of the five main areas selected, locations were established in patterns to cover a representative part of the area. At each location a wooden post was erected to support a TLD package 1 meter above the ground (see Fig. 3). The TLD package incorporates a dosimeter holder equipped with an identification hole code used for automatic processing[9].

The dosimeters remained in their field positions for approx. 3 months. In addition the positions of the post dosimeters were used as fix points for the subsequent measurements carried out with the ionization chamber and the gamma-ray spectrometer.

The ionization chamber records the total background exposure rate on a magnetic tape unit permitting further treatment of the data in a computer.

The radioelement concentrations in the ground were evaluated by means of the portable spectrometer. Counts were accumulated for several minutes with the crystal placed at a field dosimeter position and the non-terrestrial

detector background counts were recorded at sea.

Calibration and data reduction

A certified ^{226}Ra source (1 mCi) calibrated at Amersham was used for calibration of the ionization chamber and for verification of the TL response.

A flat ground at Risø was selected for the calibration experiment in which the source was mounted on a 3 m high wooden post. The ionization chamber and TL dosimeters were successively exposed on top of a 3 m high wooden post placed at a distance of 3 m from the ^{226}Ra source. The calculated exposure rate in the detection position including the determination of the scatter contribution from the earth was in agreement with the instrument reading and the TL response within 1%.

Counts recorded with the portable spectrometer were processed using sensitivity constants and stripping ratios evaluated from calibration measurements on large radioactive concrete pads available at Risø National Laboratory. These pads have known concentrations of thorium, uranium, and potassium and make it possible to calibrate in a source-detector geometry of nearly 2 π[10].

For all field localities, including those at Kvanefjeld, the total exposure rate can be written as:

$$\dot{X} = \dot{X}_{ter} + \dot{X}_{sky} + \dot{X}_{cos}$$

where $\dot{X}_{ter}$ is the exposure rate produced by upward-directed gamma-radiation from the radioelements in the ground.

From the measurements on the Kvanefjeld plateau the following conversion factors for the determination of $\dot{X}_{ter}$ were evaluated[11]:

0.25 μR/h per ppm Th
0.50 μR/h per ppm U
1.25 μR/h per % K

$\dot{X}_{sky}$ is the exposure rate contribution from terrestrial gamma-ray skyshine. At a height of 1 meter above an infinite homogeneously distributed source, 13% of the total terrestrial exposure rate contribution ($\dot{X}_{ter} + \dot{X}_{sky}$) is skyshine[11], i.e. $\dot{X}_{sky}$ is given by:

$$\dot{X}_{sky} = 0.15 \times \dot{X}_{ter}$$

In general the field sites in Greenland are considered infinite and homogeneous gamma sources. However for the Kvanefjeld rocks, the U, Th concentrations vary appreciably over short lateral distances so that the Kvanefjeld plateau cannot be considered an infinite source. The measurements in this particular area indicated that $\dot{X}_{sky}$ had a constant value of 16 μR/h anywhere on the plateau[11].

$\dot{X}_{cos}$ is the cosmic-ray contribution to the exposure rate, which amounts to approx. 4 μR/h.

Results

The evaluated exposure rates are given in Table II, and Fig. 4 gives the ionization chamber data plotted against the TLD data.

The ionization chamber and TLD responses express the present total exposure and represent cosmic radiation including the skyshine component in addition to terrestrial gamma radiation from fallout and natural radionuclides as well as the scatter contribution due to field geometry.

The reasonable agreement between TLD and ionization chamber data indicates an almost identical response for the two detector systems.

Referring to the measurements in Narssaq town (near Kvanefjeld) it is notable that the exposure rate measured on the roads is considerably higher than that measured elsewhere in the town area. This is because construction materials for the roads in Narssaq consist of material derived partly from the Kvanefjeld area.

The Kvanefjeld (Ilimaussaq) and Sarfartoq areas which have an increasing tourism show high exposure levels whereas the most populated places around Godthåb, Fiskenæsset and Frederikshåb appear to be areas of low natural radioactivity.

Conclusion

An extensive radiation monitoring programme was carried out at Kvanefjeld, South Greenland, with the aim of estimating the level of external radiation exposure to personnel working in this area. A number of field locations were monitored using LiF thermoluminescence dosimeters a high-pressure ionization chamber and a portable gamma-ray spectrometer. On the basis of these measurements reliable factors for the conversion of a set of surface concentration values into a gamma-ray exposure rate were evaluated and used to calculate the exposure levels at other places in Greenland based on available geological data.

The radiation exposure level data recorded, make it possible to esti-

mate the external personnel gamma radiation doses which may be obtained during future field work in Greenland.

Acknowledgement

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Table I

Geology of the areas in Greenland where the exposure-rate levels have been determined.

Area	Geology
1. Nordre Strømfjord	Early Proterozoic gneiss terrain
2. Søndre Strømfjord	Early Proterozoic gneiss terrain
2a. Sarfartoq	Phanerozoic carbonatite complex
3. Godthåb	Archaean granulite facies gneiss
4. Fiskenæsset	Archaean granulite amphibolite facies gneiss
5. Frederikshåb	Archaean amphibolite facies gneiss
6. Kvanefjeld, Ilimaussaq	Alkaline igneous complex of middle Proterozoic age
7. Scoresby Sund, Inner zone	Gneiss and granite of Caledonian and pre-Caledonian age
8. Scoresby Sund, Outer zone	Marine sandstone and shale of upper Palaeozoic and Mesozoic age

Table II

Average radioelement concentrations and exposure rates determined for various areas in Greenland.

Area	ppm Th	ppm U	%K	μR/h calc.	μR/h TLD	μR/h Ionization chamber
Kvanefjeld I	2521	837	7.9	1079	1067	x)
" II	476	223	3.1	254	255	251
" III	20.6	7.1	1.4	30.5	25.3	23.0
" IV	418	248	1.7	251	253	240
" V	1314	454	1.7	578	575	x)
Ilimaussaq	45	25	3	35.6		
Inside houses in Narssaq Town	6.2	1.9	0.9	8.2	8.8	8.8
Roads in Narssaq Town	17.8	5.8	2.0	15.3		14.5
Nordre Strømfjord	10.0	0.5	1.0	8.6		
Søndre Strømfjord	9.7	4	1.4	11.1		
Sarfartoq	50	10	3	28.4		
Godthåb	1.6	0.2	0.8	5.8		
Fiskenæsset	3.2	0.4	1.3	7.0		
Frederikshåb	3.4	1.1	1.4	7.6		
Scoresby Sund, Inner Zone	18	3	3.8	16.4		
" " , Outer Zone	6.9	3.2	1.7	10.3		

*) Saturation of ionization chamber

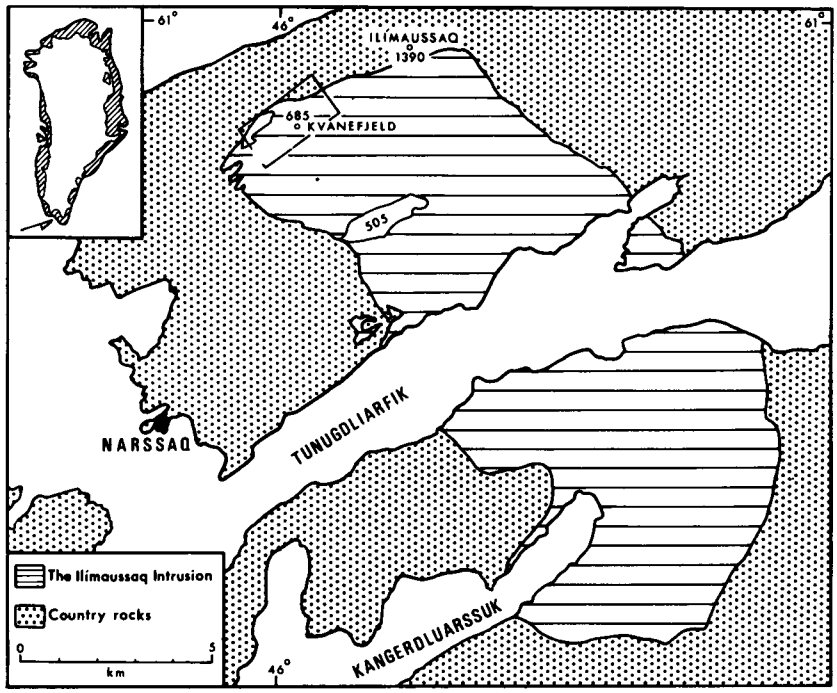
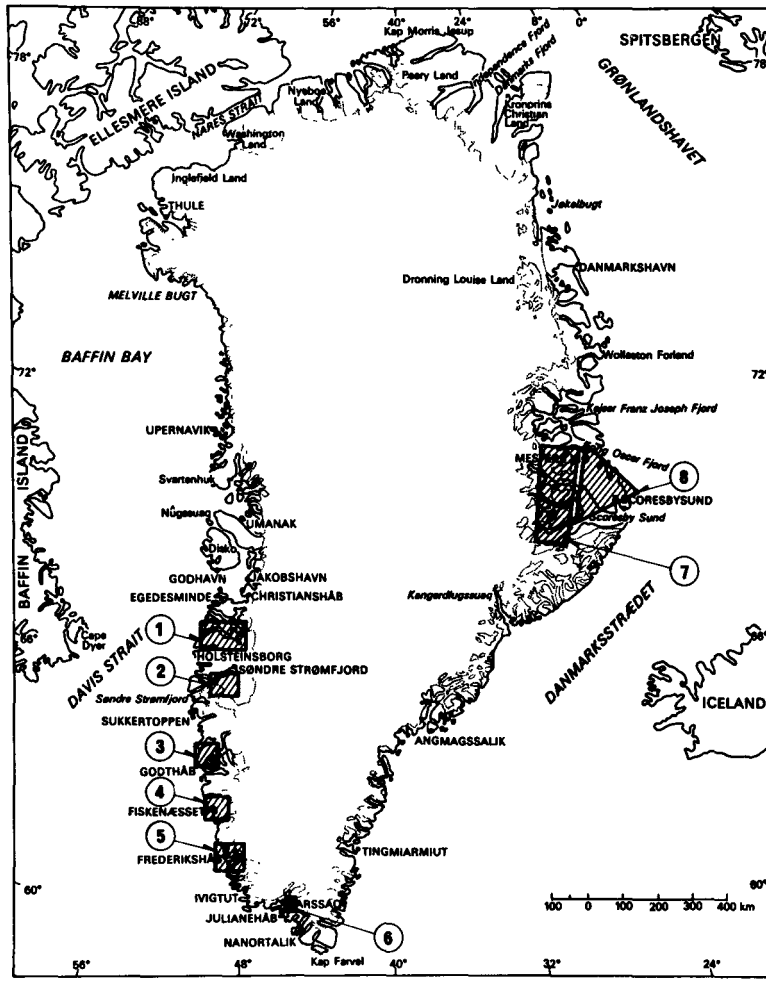


Fig. 1. Location of the Ilimaussaq intrusion and the Kvanefjeld uranium deposit, South Greenland.



1. NORDRE STRØMFJORD AREA
2. SØNDRE STRØMFJORD AREA
3. GODTHÅB AREA
4. FISKENÆSSET AREA
5. FREDERIKSHÅB AREA
6. KVANEFJELD AREA
7. SCORESBYSUND, INNER ZONE
8. SCORESBYSUND, OUTER ZONE

Fig. 2. Map of Greenland with sites where the exposure rates have been determined.

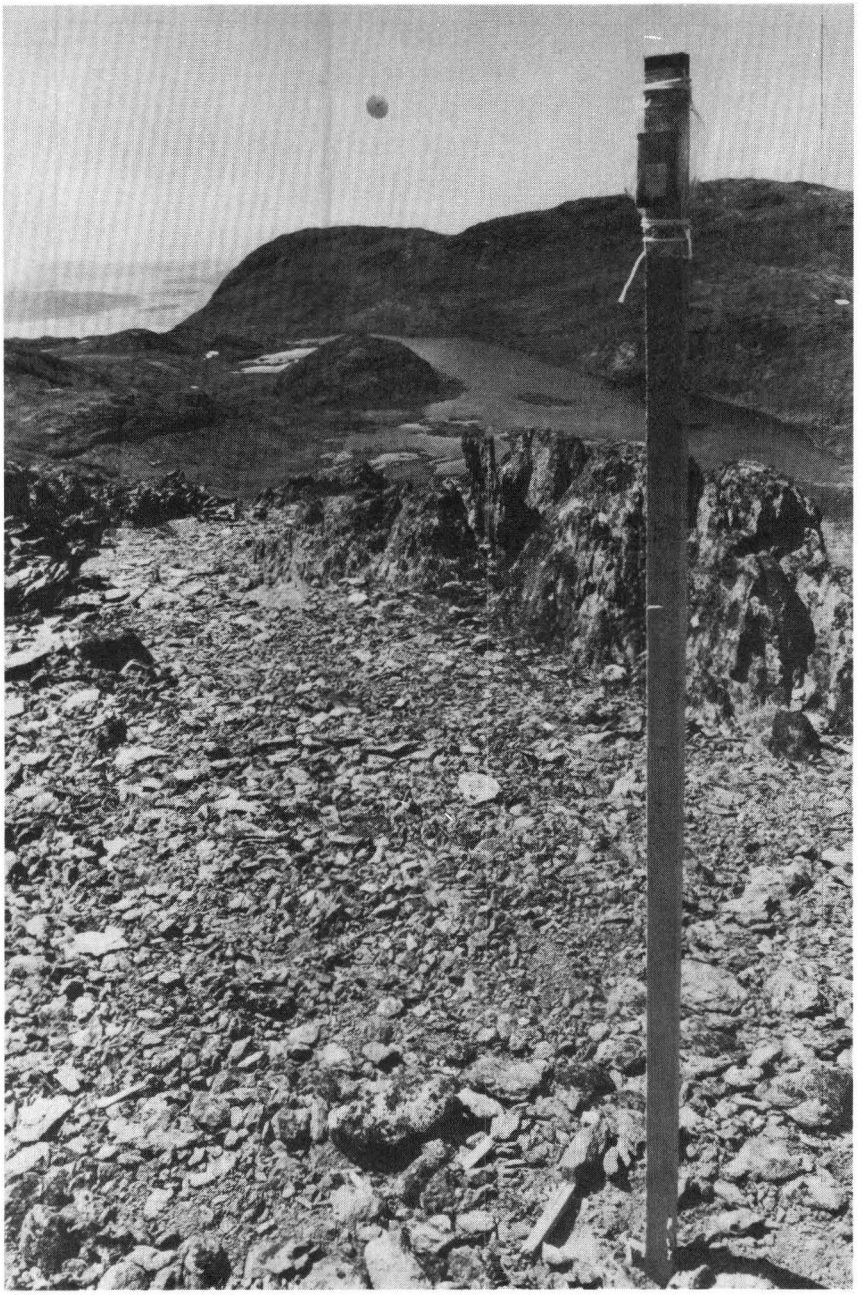


Fig. 3. TLD package on the Kvanefjeld plateau.

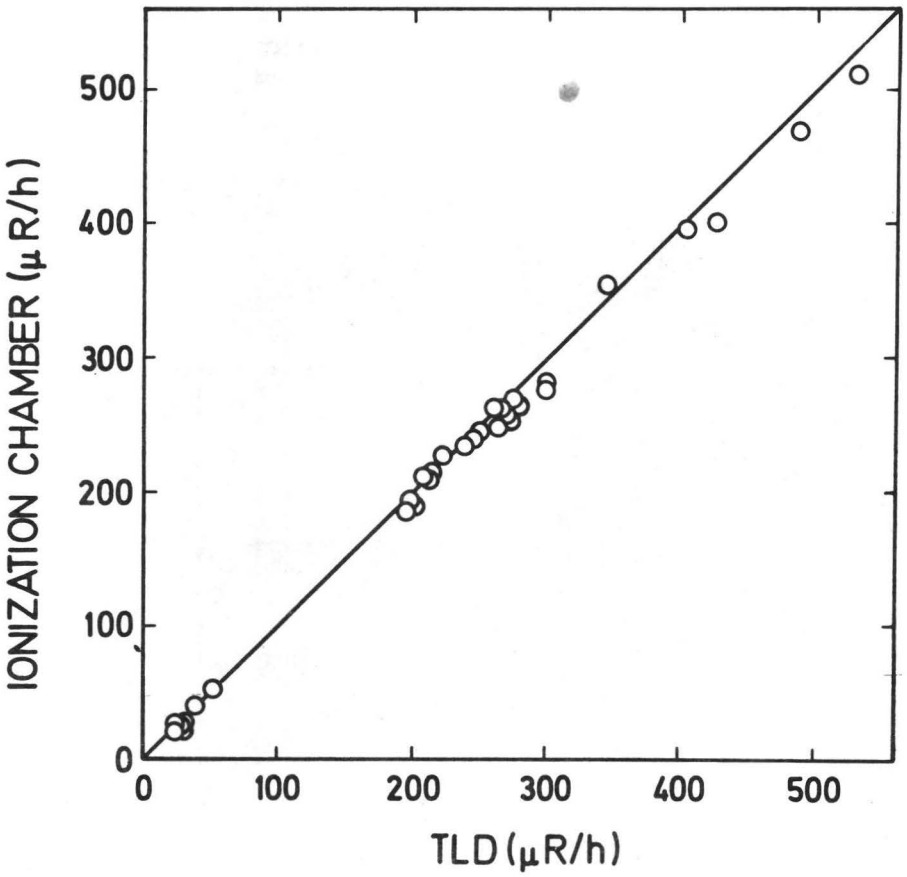


Fig. 4. Ionization chamber data plotted against the TLD data.

TERRESTRIAL AND COSMIC RADIATION IN SCANDINAVIA

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SUMMARY. The Scandinavian bedrock consists mostly of Pre-Cambrian rocks in which granites of different ages constitute a considerable part. In parts of Sweden Pre-Cambrian rocks are overlain by Cambrian-Silurian sedimentary rocks. The upper Cambrian sequence includes alum shales which are overlain by Ordovician limestone. The granites may contain high specific activities of uranium and thorium and the alum shales contain high levels of uranium. Some studies of specific activities of uranium, thorium and potassium-40 in rocks and unconsolidated sediments are revieved.
The bedrock in Finland is almost entirely Pre-Cambrian and is very similar to the bedrocks in Sweden. The major part of Norway consists of deformed sedimentary sequencies. However, granites occur in certain parts and alum shales are present in the Oslo region. All the above-mentioned rock types also occur in Sweden. The alum shale in Sweden has been used for industrial purposes since the sixteenth centruy and this has resulted in waste piles giving radiological problems for existing and planned settlements.
The Swedish Geological Survey has made radiometric measurements from aircraft over about 40% of Sweden. These measurements together with results of geological investigations are now being combined to produce socalled GEO radiation maps. The purpose of these maps is to show where the ground contains such high levels of radionuclides that new building must be restricted and existing buildings should be investigated with regard to radon concentration indoors and gamma radiation outdoors.
The response of various instruments to cosmic radiation has been studied in order to be able to correct for this contribution to the total radiation levels both indoors and outdoors.

RESUME. RAYONNEMENTS TERRESTRE ET COSMIQUE EN SCANDINAVIE. Le socle scandinave consiste essentiellement en roches précambriennes où les granits de différents âges prédominent. Par endroit en Suède, des roches sédimentaires cambrio-siluriennes recouvrent les roches précambriennes. Les couches du cambrien supérieur comprennent des schistes alunifères recouverts de calcaire ordovicien. Les granits peuvent avoir une forte activité spécifique en uranium et les schistes alunifères une forte teneur en uranium. On passe en revue quelques études de l'activité spécifique de l'uranium, du thorium

et du potassium-40 dans les roches et les sédiments non consolidés. En Finlande, le socle quasi entièrement du précambrien, ressemble beaucoup au suèdois. La majeure partie de la Norvège consiste en couches sédimentaires déformées. On trouve cependant du granit dans certaines parties et des schistes alunifères dans la région d'Oslo. Tous les types de roches précités se rencontrent aussi en Suède. Les schistes alunifères y ont été utilisés à des fins industrielles depuis le seizième siècle; il en est résulté des monceaux de déchets qui posent des problèmes de radioprotection aux implantations existantes et prévues. Le service géologique suèdois a fait des mesures radiométriques par avion sur environ 40% du pays. Ces mesures sont actuellement combinées aux résultats de prospections géologiques en vue de dresser des cartes de rayonnements dites GEO et de révéler là où le niveau de radionucléides est si élevé qu'il faut restreindre de nouvelles constructions et là où il faudrait mesurer la concentration en radon à l'intérieur des bâtiments existants et le rayonnement gamma à l'extérieur de ceux-ci. La réponse des divers instruments au rayonnement cosmique a été étudiée pour déduire sa contribution des niveaux globaux de rayonnement, tant à l'intérieur qu'à l'extérieur.

KURZFASSUNG. TERRESTRISCHE UND KOSMISCHE STRAHLUNG IN SKANDINAVIEN. In Skandinavien besteht das Grundgestein grösstenteils aus präkambrischem Fels, der einen hohen Anteil an Graniten aus verschiedenen Erdzeitaltern aufweist. In einigen Teilen Schwedens sind die Gesteine aus dem Präkambrium von Sedimentgesteinen aus dem Kambrium und dem Silur überlagert. Die obere Kambrium-Schicht enthält Alaunschiefer, die von Kalkstein aus dem Ordovizium überlagert sind. Die Granite können Uran und Thorium mit hohen spezifischen Aktivitäten enthalten; die Alaunschiefer weisen hohe Uranpegel auf. Es wird ein Überblick über einige Untersuchungen spezifischer Aktivitäten von Uran, Thorium und Kalium-40 in Fels und unverfestigten Sedimenten gegeben. Das Grundgestein in Finnland entstammt fast ausschliesslich dem Präkambrium und ist dem schwedischen Grundgestein sehr ähnlich. Das norwegische Gebiet besteht überwiegend aus verformten Sedimentschichten. In einigen Teilen Norwegens treten jedoch Granite auf, und in der Osloer Region sind Alaunschiefer festzustellen. Alle oben angeführten Gesteinstypen sind auch in Schweden anzutreffen. Der schwedische Alaunschiefer wird seit dem sechzehnten Jahrhundert für industrielle Zwecke verwendet, was zu Abfallanhäufungen geführt hat, die für die bestehenden und geplanten Siedlungen radiologische Probleme mit sich bringen.

Im Rahmen der geologischen Vermessung Schwedens wurden vom Flugzeug aus ungefähr 40% des schwedischen Gebiets radiometrisch erfasst. Diese Messungen werden zur Zeit neben den Ergebnissen geologischer Untersuchungen für die Anfertigung sog. GEO-Strahlungskarten verwendet. Diese Karten sollen aufzeigen, an welchen Stellen der Boden derart hohe Radionuklidpegel aufweist, dass die Errichtung von Neubauten eingeschränkt werden muss und die bereits vorhandenen Gebäude auf die Radonkonzentration in den Wohnungen und die Gammastrahlung ausserhalb der Wohnungen untersucht werden sollten.
Das Ansprechen verschiedener Instrumente auf die kosmische Strahlung wurde geprüft, um deren Beitrag zu der Gesamtheit der Strahlenpegel innerhalb und ausserhalb der Wohnungen berücksichtigen zu können.

Rolf Sievert, already 30 years ago, found regions in Sweden with enhanced gamma radiation levels. However, his measurements covered limited regions and the radiation protection problem for the general population was not then considered to be serious. It is only a few years since there was found to be an appreciable, and in some cases serious, radiation problem due to the extent to which the radon exhaled from the soil passes through the building materials into the houses, even in houses built on normal ground.

When houses are built on ground containing higher contents of radium than is usual the concentrations of radon daughters may be unacceptable for the individuals living in the houses. This situation is not as unusual as was believed some years ago.

An illustration of this is the problem associated with houses built on abandoned wastes from alum shale mining in past centuries in Sweden (1). The radon concentration indoors was found to be about 900 Bq/m^3 (24 pCi/l) as an average for six houses. The highest value found for a house was 2 000 Bq/m^3 (55 pCi/l), calculated as an average for a year. For comparison, indentical houses built on normal ground also built of normal building materials were studied. The radon concentrations indoors in those houses were about 60 Bq/m^3 (1.6 pCi/l).

Besides this direct contribution to the radiation dose from the ground, the major dose contribution associated with the radionuclides in the ground is that to persons in houses, since the building materials are often taken from the ground. Often local building materials are used and this causes variations between the average radiation doses to persons in houses in different regions.

The term "Scandinavian countries" is usually taken to mean Norway, Sweden and Denmark. In this paper I shall deal with the terrestrial radiation in Norway, Finland and Sweden. Since my knowledge of Sweden is better than for the other countries I shall take my examples from Sweden.

The radioactivity of bedrocks and soils

The Scandinavian bedrock consists mostly of Pre-Cambrian rocks in which granites of different ages constitute a considerable part (Fig 1). In parts

of Sweden Pre-Cambrian rocks are overlain by Cambrian-Silurian sedimentary rocks. The Upper-Cambrian sequence includes alum shales which are overlain by Ordovician limestone.

The alum shale got its name when alum, $KAl(SO_4)_2 \cdot 12H_2O$, was produced from the shale. Alum was used in the dying of textiles and was produced in Sweden from the end of the sixteenth century until the beginning of the twentieth century. Alum shale has also been used for burning of lime. During a short period it was also used for producing oil and some other organic substances.

The contents of uranium, thorium and potassium-40 in rocks and unconsolidated sediments are reviewed in Table I received from Geological Survey of Sweden. The contents of the radionuclides have been determined from gamma spectrometric measurements in the field. The number and distribution over the country of the measurements are intended to give representative results for the bedrocks and soils occurring in Sweden. The dose equivalents above the ground have been given, calculated from the concentrations according to the model given in the 1977 UNSCEAR report (2).

It will be seen that the contents of the naturally occurring radionuclides in limestone, sandstone and shales are low. Granites may contain high concentrations of uranium and thorium. Alum shale contains relatively large amounts of uranium. The grey-brown Middle-Cambrian alum shales generally have contents of uranium of between 10 and 50 grams per tonne. The contents of uranium in the black Upper-Cambrian and Lower-Ordovician alum shales are much higher, varying between 50 and 350 grams per tonne.

The contents of the radionuclides in soils are shown in Table II (3). In normal soils the concentrations vary within small intervals, but in regions where the glacial till contains bedrocks of relatively high specific activity the variations may be greater. Examples are tills containing materials from some granites or alum shales. Some clays contain so much thorium and uranium that the bricks produced from them show unusually high radioactivity.

The average activity of radium in samples from the waste pile on which the houses mentioned in the introduction were built was 2 900 Bq/kg (78 pCi/g) according to Table III (1). For comparison the activity concentration of natural ballast material for production of concrete in the same region (4)

and of agricultural soil (5) have been given. These measurements were made using gamma spectrometry with the sample placed in a hat container over a NaI crystal coupled to a multichannel analyzer. No high values were found in these investigations. However, the soil samples were few and the high gamma levels found above some fields suggest that higher values might be found in some parts of the region.

The gamma radiation from the ground

The absorbed dose in air from the gamma radiation above the ground has been calculated from the activity concentrations in the ground from Tables I, II and III and is shown in Table IV. The model used is taken from the 1977 UNSCEAR Report (2) which refers to the absorbed dose rate in air one metre above the ground.

The dose rates above most types of rocks, as measured directly by gamma detectors, are up to 200 nGy/h (20 μrad/h). However, the dose rates above uranium- and thorium-rich granites might be as much as 500 nGy/h (50 μrad/h) according to the calculations. However, field measurements made by the Swedish Geological Survey (3) above granites have shown even higher levels, up to 4 000 nGy/h (400 μrad/h) for some sites.

Above alum shale the absorbed dose rate in air might be up to 1 900 nGy/h (190 μrad/h) according to the calculations.

The dose rates above soils are usually lower than above rocks. Some tills, however, contain split alum shale and produce high gamma levels. Values of 1 000 nGy/h (100 μrad/h) have been measured.

In Norway the environmental radiation has been investigated with an ionization chamber placed in a car (6). The exposure was integrated while driving the car so that measurements could be made representing an average value for an area. Norway was divided into eight districts according to Fig 2. In Table V the gamma dose rates in air are presented. Relatively large local variations were found. The lowest value, 20 nGy/h (2 μrad/h), was found at a local spot in district VII where the rocks were mostly sedimentary. The highest value 1 130 nGy/h (113 μrad/h), was found in district I. The contribution from the cosmic radiation has been subtracted.

The extreme average values for the districts vary by a factor of almost two from the average for all the districts. In the districts with relatively high gamma radiation levels granites and shales were dominant.

In Fig 3 the number of measurements is plotted against the gamma dose rates in air for all districts in Norway, for towns and for open roads. The higher values for towns, where buildings and paving give additional gamma radiation, are comparable with those found in measurements made by car in Sweden during the 1950s.

The network of 24 gamma stations distributed over Sweden show gamma levels from 40 nGy/h (4 μrad/h) up to 100 nGy/h (10 μrad/h). The values have been corrected for the contribution of fallout and the cosmic radiation.

The response to the cosmic radiation has been studied for the various instruments used, in order to be able to subtract it from the total gamma radiation measured outdoors and indoors.

The gamma stations are, where possible, placed 2.5 metres above a flat piece of land - mostly above a lawn. The station with the lowest value, 40 nGy/h (4 μrad/h), is situated over a lawn under which the bedrock is limestone. The highest values are found in granite regions.

The continuous measurements at the 24 gamma stations illustrate the fact that the gamma levels vary by up to 20 per cent (7) for different meteorological conditions, due to the variations in the radon exhalation from the soil, and by even more when the ground is covered by snow.

Radon exhalation

The radon exhalation varies with the kind of bedrock but also with the condition of the material. Alum shale, for example, is a very fine-grained and dense type of rock. The radon exhalation is therefore normally very small. On weathering, however, the shale fragments and the radon gas is released at a higher rate. The radon exhalation from burnt alum shale is three times that from crushed but not burnt alum shale.

The distribution of various kinds of bedrocks and soils

Pre-Cambrian bedrock constitutes the entire country of Finland, the major part of Sweden, and is present over large areas of Norway. The remainder of Norway and the westernmost part of northern Sweden consist of deformed and metamorphosed sedimentary sequences. Granites of varying ages occur in all three countries, and alum shales are present in a number of areas in Sweden, and in the Oslo region of Norway.

The occurrence of alum shale in Sweden is shown in Fig 4 (3). The activity concentration of uranium varies between different areas, partly because of the various kinds of alum shale mentioned above. For example, the activity concentration of the black alum shale in southern Sweden at points 5 and 6 in Fig 4 is between 50 and 150 grams per tonne, while the values in Västergötland (point 2) are about twice as high (3).

As has already been pointed out, alum shale is often covered by limestone and till, this is illustrated in Fig 5. The activity concentration of limestone is very low. The levels of gamma radiation are therefore usually very low in those regions where the ground surface is undisturbed. The till, however, may contain high contents of weathered alum shale and may therefore give rise to both high gamma radiation levels and high radon exhalation. This can also be the case in regions where no bedrock of alum shale is to be found. The fragmented alum shale has been transported by the inland ice and carried long distances by melt water rivers to be deposited at the edge of the receding glaciers.

The next figure, Fig 6, shows the occurrence of granites with unusually high contents of radioactive substances in Sweden (3). The exposure rates shown on the map vary between 20 and 60 μR/h. Some months ago, measurements made by the Geological Survey of Sweden showed levels of up to 400 μR/h above granites in a region where new building was planned for 15 000 persons and where roads etc had already been built. This problem might also occur in some granite regions in Finland and Norway.

The alum shale in Norway, in the Oslo region, seems to contain relatively low specific activities of radionuclides. In Finland no alum shale is to be found.

Planning for new building areas

In areas where the ground contains very high activity concentrations, care must be taken when new building is planned. Houses already existing on such ground should be checked with regard to the concentration of radon daughters indoors.

The Swedish Committee on Measures Against Radiation Hazards in Buildings has discussed this problem in its preliminary report (8). The Committee has commissioned the Geological Survey of Sweden to produce so called GEO radiation maps. Radiometric measurements from aircraft over about 40 per cent of Sweden have been carried out for other purposes. These measurements are now being combined with results of geological investigations to produce the GEO radiation maps.

The purpose of these maps is to show where the ground contains such high levels of radionuclides that new building must be restricted and existing buildings should be investigated with regard to the radon daughter concentration indoors and the gamma radiation outdoors.

The GEO radiation maps are planned to be the first basic data when planning the land for building purposes. More detailed mapping is needed for the detailed planning of regions which the GEO radiation maps have shown to be problem regions.

As mentioned above, the radon from the ground contributes to the radon concentration indoors, and therefore to the collective absorbed dose from the air indoors, to a degree which cannot be neglected when the air exchange rates are decreased to very low values. This is the case for rather widespread regions of the Scandinavian countries because of the relatively high contents of uranium normally found in the ground.

Average absorbed doses

The absorbed dose rates in air from gamma radiation outdoors have been estimated to be 73 nGy/h (7.3 μrad/h) with a range of 20 to 1 200 nGy/h (2 - 120 μrad/h) (6) for Norway. The corresponding values for Sweden are 80 nGy/h (8 μrad/h) with a range of 18 to 4 000 nGy/h (1.8 - 400 μrad/h). This corresponds to the dose equivalents 0.53 mSv/a (53 mrem/a) from the

ground for Norway and to 0.58 mSv/a (58 mrem/a) for Sweden with a conversion factor from absorbed dose rate in air to dose equivalent of $7.2 \cdot 10^{-3}$ mSv mGy^{-1} (2) and for an occupancy factor of 100 per cent outdoors. The total dose equivalent including the contribution from the cosmic radiation is about 0.83 mSv/a (83 mrem/a) for Norway and 0.88 mSv/a (88 mrem/a) for Sweden.

The average absorbed doses from radon daughters outdoors have not been estimated due to lack of information. The average effective dose equivalent from the inhalation of radon daughters for 100 per cent occupancy time outdoors might be somewhere between 0.1 and 1 mSv/a but very great variations are to be expected between various regions. The variation over 24 hours of the concentration of radon in air 0.3 metres above the ground is illustrated by Fig 7 (9). The collective dose from the contribution of the radon in the ground to the radon daughter concentration indoors has not been estimated.

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Table I.

Uranium (U), thorium (Th) and potassium (K) contents and radiation dose (mSv/yr) for different rock types in Sweden. (3)

	U gram/ton	Th gram/ton	K %	mSv/yr
Granite, normal	2 - 10	5 - 20	2 - 6	0.4 - 1.4
Granite, uranium and thorium rich	8 - 40	10 - 90	4 - 6	0.9 - 3.9
Gneiss	2 - 10	5 - 20	2 - 6	0.4 - 1.4
Diorite	0.1 - 2	1 - 10	1 - 3	0.1 - 0.6
Sandstone	0.5 - 5	1 - 10	1 - 5	0.1 - 0.9
Limestone	0.5 - 2	0.1 - 2	0.1-0.5	<0.1 - 0.2
Shale	1 - 10	2 - 12	2 - 5	0.3 - 1.1
Alum shale M. Camb.	10 - 50	2 - 10	2 - 6	0.7 - 2.7
Alum shale U. Camb.-L.Ord.	50 -350	2 - 10	3.5 - 6	2.4 -14.5

1 gram uranium per ton is equivalent to 12.3 Bq/kg of ^{238}U
1 gram thorium per ton is equivalent to 4.0 Bq/kg of ^{232}Th
1 % potassium is equivalent to 310 Bq/kg of ^{40}K

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Table II.

Uranium (U), thorium (Th) and potassium (K) contents and radiation dose (mSv/yr) for different soil types in Sweden. (3)

	U gram/ton	Th gram/ton	K %	mSv/yr
Till	3.5 - 7.0	12 - 25	2.7 - 3.4	0.5 - 1.1
Gravel	2.5 - 3.5	13 - 24	3.0 - 3.4	0.6 - 0.9
Sand	1.5 - 3.0	8 - 19	2.5 - 2.7	0.6 - 0.7
Silt	6.0 - 8.0	11 - 25	2.7 - 3.0	0.7 - 1.1
Clay	4.0 - 8.5	17 - 37	2.9 - 3.7	0.8 -1.4

1 gram uranium per ton is equivalent to 12.3 Bq/kg of ^{238}U
1 gram thorium per ton is equivalent to 4.0 Bq/kg of ^{232}Th
1 % potassium is equivalent to 310 Bq/kg of ^{40}K

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Table III a. Activity concentrations of samples from old wastes from alum shale mining (1).

Sample No.	Bq/kg			Photon emmission rate $kg^{-1}s^{-1}$	pCi/g			ΣpCi/g
	^{232}Th	^{226}Ra	^{40}K		^{232}Th	^{226}Ra	^{40}K	
1	190	1990	1250	4830	5.1	54	34	130
2	270	2540	1630	6240	7.3	69	44	170
3	300	2780	1870	6850	8.1	75	51	190
4	230	3250	1650	7610	6.2	88	45	210
5[1)]	350	3940	2230	9460	9.5	106	60	260
Average	270	2900	1730	7000	7.2	78	47	190

1) Selected sample of alum shale

Table III b. Activity concentrations of natural ballast materials for concrete production and normal agricultural soil (1).

Sample	Number	Bq/kg			Photon emmission rate $kg^{-1}s^{-1}$	pCi/g			ΣγpCi/g
		^{232}Th	^{226}Ra	^{40}K		^{232}Th	^{226}Ra	^{40}K	
Ballast materials									
Hjo, Tibro	2	67	44	1100	390	*1.8*	*1.2*	*30*	*10.6*
Skövde	2	71	41	1130	400	*1.9*	*1.1*	*31*	*10.9*
Skara	3	58	41	880	340	*1.6*	*1.1*	*24*	*9.2*
Lidköping	2	53	41	800	320	*1.4*	*1.1*	*22*	*8.6*
Country-wide Average	306	72	48	814	385	*1.9*	*1.3*	*22*	*11.1*
Agricultural soil (4)									
Skaraborg county	9	74	67	850	-	*2.0*	*1.8*	*23*	-
The whole country	268	44-115	33-96	480-850	-	*1.2-3.1*	*0.9-2.6*	*13-23*	-

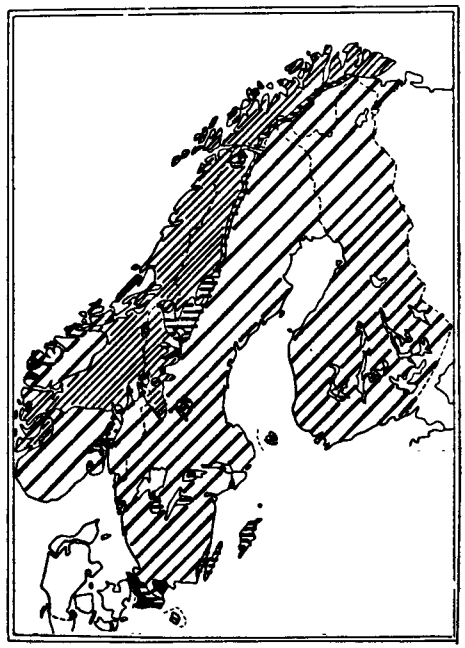
Table IV. Absorbed dose in air one meter above the ground calculated from the activity concentrations in the soil or rock given in Tables 1, 2 and 3.

	Calculated	
	nGy/h	μrad/h
ROCK TYPES		
Granite, normal	50 - 190	5 - 19
Granite, uranium- and thorium-rich	120 - 530	12 - 53
Gneiss	50 - 190	5 - 19
Diorite	16 - 80	1.6 - 8
Sandstone	18 - 120	1.8 - 12
Limestone	4 - 17	0.4 - 1.7
Shale	37 - 150	3.7 - 15
Alum shale M.Camb.	80 - 370	8 - 37
Alum shale U.Camb. - L.Ord.	310 - 1 900	31 - 190
SOIL TYPES		
Till	90 - 150	9 - 15
Gravel	90 - 130	9 - 13
Sand	60 - 100	6 - 10
Silt	100 - 150	10 - 15
Clay	110 - 190	11 - 19
Waste from old alum shale mining	1 500	150

Table V. The table shows the doses from the γ-radiation in open air in the districts of Norway[1] (6).

District	Per cent of total population	γ-dose rate in air (nGy/h)		
		Towns	Open areas	Averaged
I	5	68	76	71
II	8	91	75	84
III	5	77	75	76
IV	11	90		90
V	15	95	62	85
VI	9	63	56	59
VII	6	48	36	43
VIII	10	48	55	53
IX	6	58	50	55
X	12	75	72	73
XI	7	85	77	83
All districts	94	75	63	73

1) The populated weighted average outdoor dose rates are calculated from data on the resident population in the districts and of population pattern.



Pre-Cambrian rocks
(older sedimentary rocks, rocks of volcanic, gneiss, granites etc)

Caledonian mountain range
(undifferentiated)

Cambrian-Silurian sedimentary rocks
(sandstone alum shales, Limestone etc)

Sedimentary rocks of Devonian
to Tertiary age

Fig 1. Sketch map of the bed-rocks of Norway, Sweden and Finland.

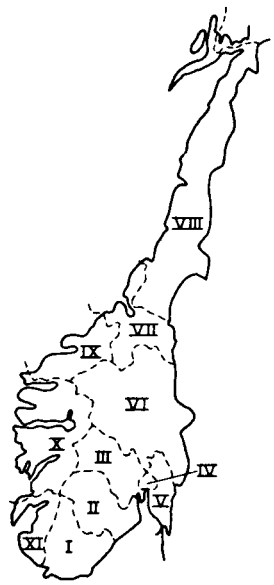


Fig 2. A map of Southern Norway where the different districts are indicated. (6).

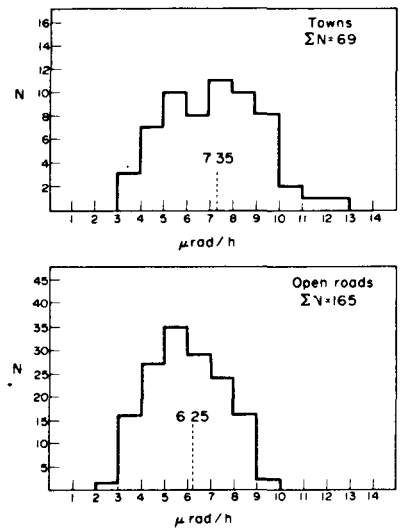


Fig 3. The number, N, of measurements vs the γ-dose rate in air in all districts (6).

DISTRIBUTION OF ALUM SHALE IN SWEDEN

1 CALEDONIAN FRONT
2 VÄSTERGÖTLAND
3 NÄRKE
4 ÖSTERGÖTLAND
5 ÖLAND
6 SKÅNE

0 100km

FIG. 4

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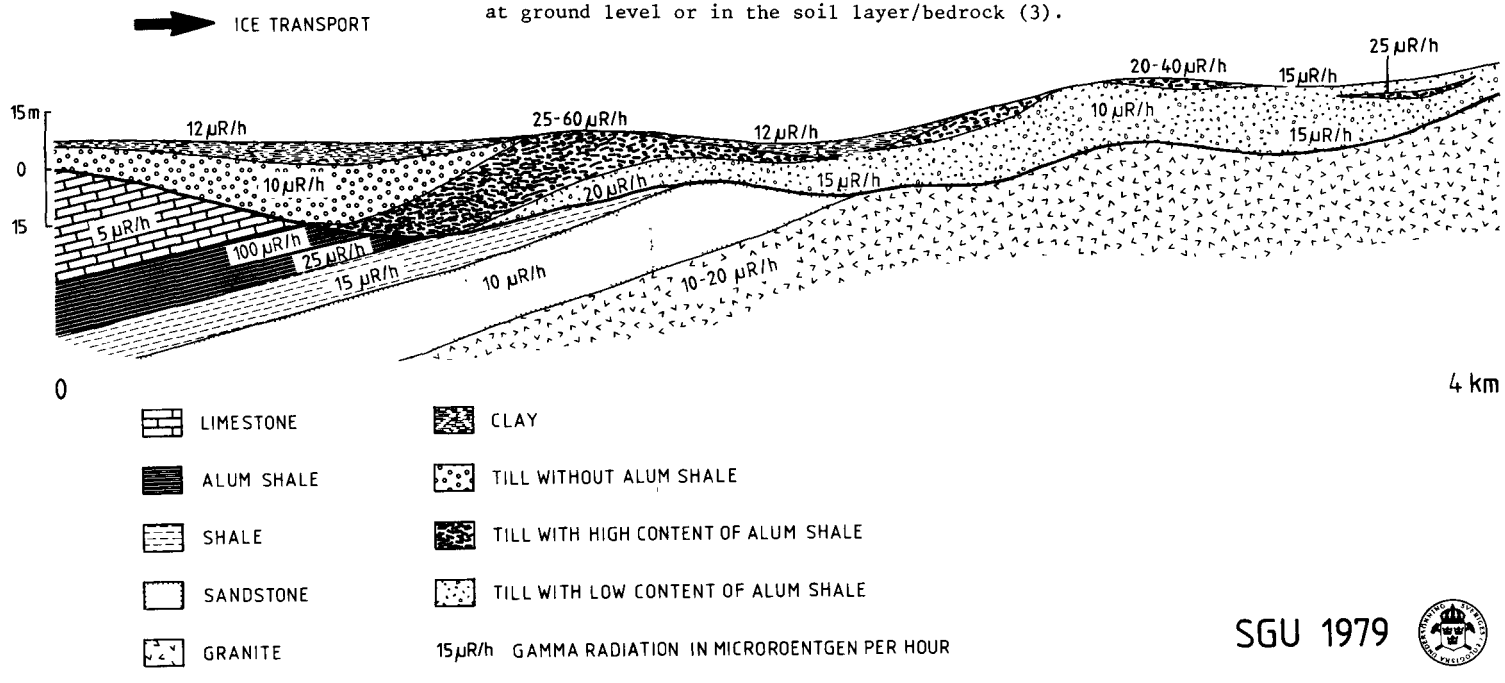
Fig 5. Illustration of how the alum shale has been transported and mixed into the till and the exposure rate in the bedrock and the soil. The figure is schematic and the scales of height and length vary depending on the environment. The gamma radiation is assumed to be measured at ground level or in the soil layer/bedrock (3).



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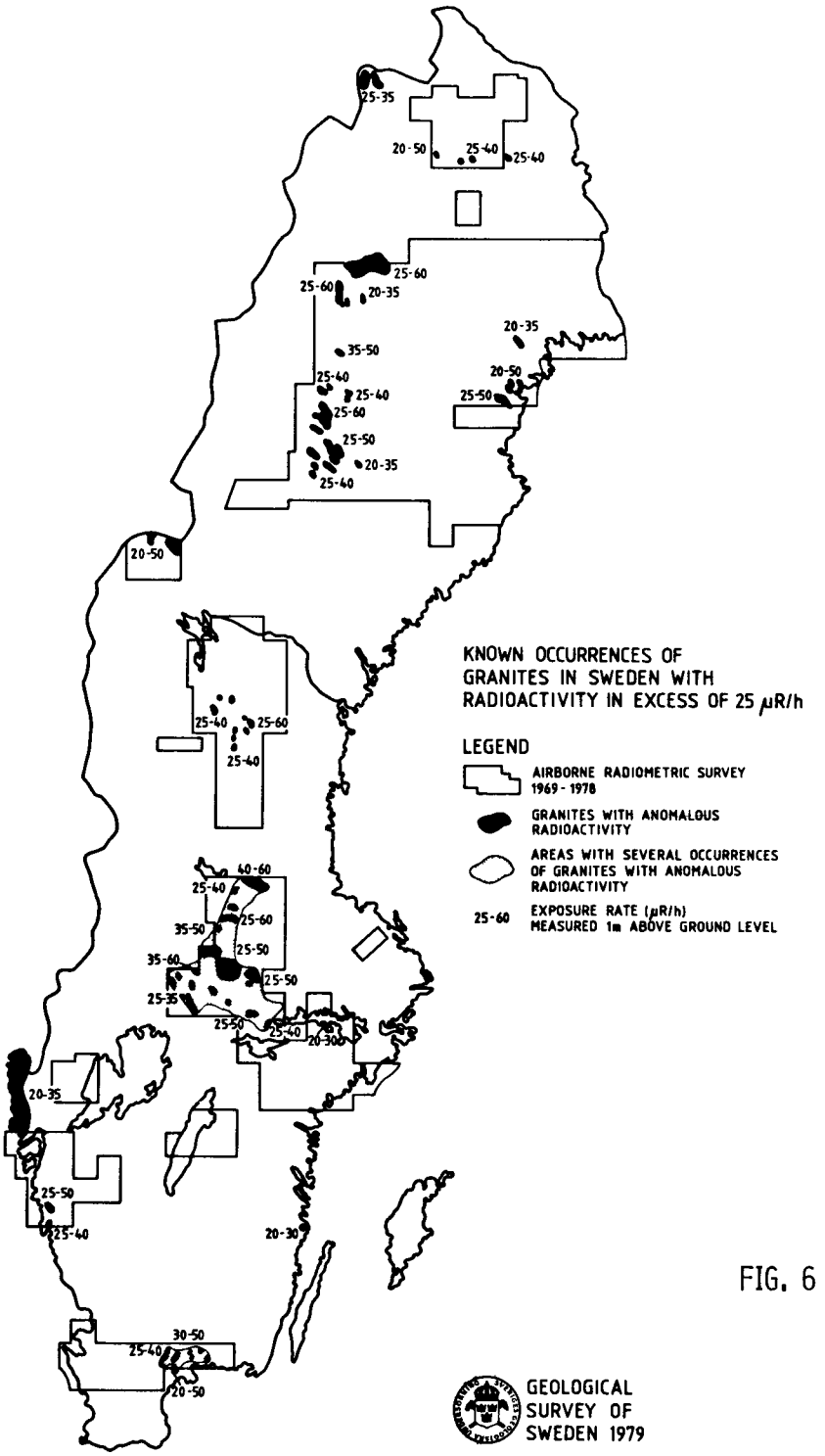
KNOWN OCCURRENCES OF
GRANITES IN SWEDEN WITH
RADIOACTIVITY IN EXCESS OF 25 μR/h
LEGEND
AIRBORNE RADIOMETRIC SURVEY
1969 - 1978
GRANITES WITH ANOMALOUS
RADIOACTIVITY
AREAS WITH SEVERAL OCCURRENCES
OF GRANITES WITH ANOMALOUS
RADIOACTIVITY
25-60 EXPOSURE RATE (μR/h)
MEASURED 1m ABOVE GROUND LEVEL
GEOLOGICAL
SURVEY OF
SWEDEN 1979

FIG. 6

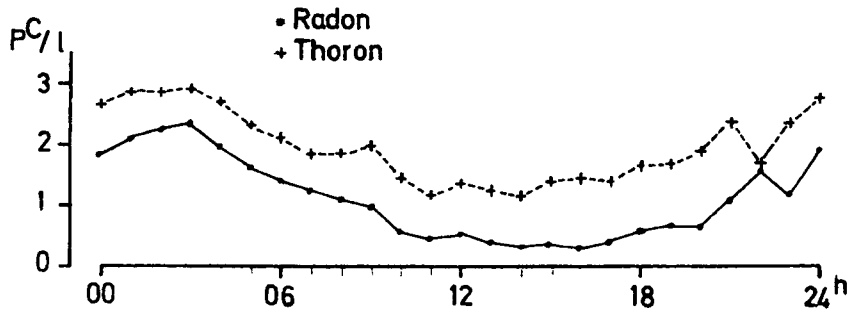

Fig 7. Diurnal variations of radon and thoron concentrations at the height 0.3 m (9).

VARIATION OF THE TERRESTRIAL LOCAL DOSE RATE ON A JOURNEY THROUGH SCANDINAVIA

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During a journey from Aachen to Helsinki through Finland, Norway, Sweden, Denmark and back to Aachen with a private car the local dose rate was measured using one and the same calibration factor. In order to determine the change of the local dose rate offside the roads in comparison to the local dose rate on the roads, measurements offside the roads and outside the car were carried out, whenever a high alteration was registered. The measurements were carried out using a scintillation counter Sz 25/64 BD and a rate meter scaler LB 1821 from the laboratory of Prof. Dr. Berthold.

The scintillation counter lay flat on the floor of the car, on the right hand side between the front- and the back seat. The scintillation cristal was directed to the right. Parallel measurements in and out of the car, in a height of 1 m, in several different places showed that the car shields approx. 1o % of the radiation.

Fig. 1 shows the itinerary and the average terrestrial local dose rate in the car for single sections of the road. In brackets, some values of measured dose rates offside the road and outside the car are mentioned. The highest measured value was approx. 3oo mrem/a close to the wall in the Temppeliaukio church in Helsinki. The lowest values of approx. 25 mrem/a were measured near Trondheim and the Lüneburg heather.

It shows that the change of the terrestrial local dose rate on the roads agrees well with the geological changes. The higher terrestrial local dose rates measured in regions with granite and gneiss corres-

* Data introduced from the floor

ponds to the values measured in the black forest and the bavarian forest in the Federal Republic of Germany /1/, /2/.

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/2/ Der Bundesminister des Innern:
Die Strahlenexposition von außen in der Bundesrepublik Deutschland durch natürliche radioaktive Stoffe im Freien und in Wohnungen unter Berücksichtigung des Einflusses von Baustoffen;
Bonn (1978)

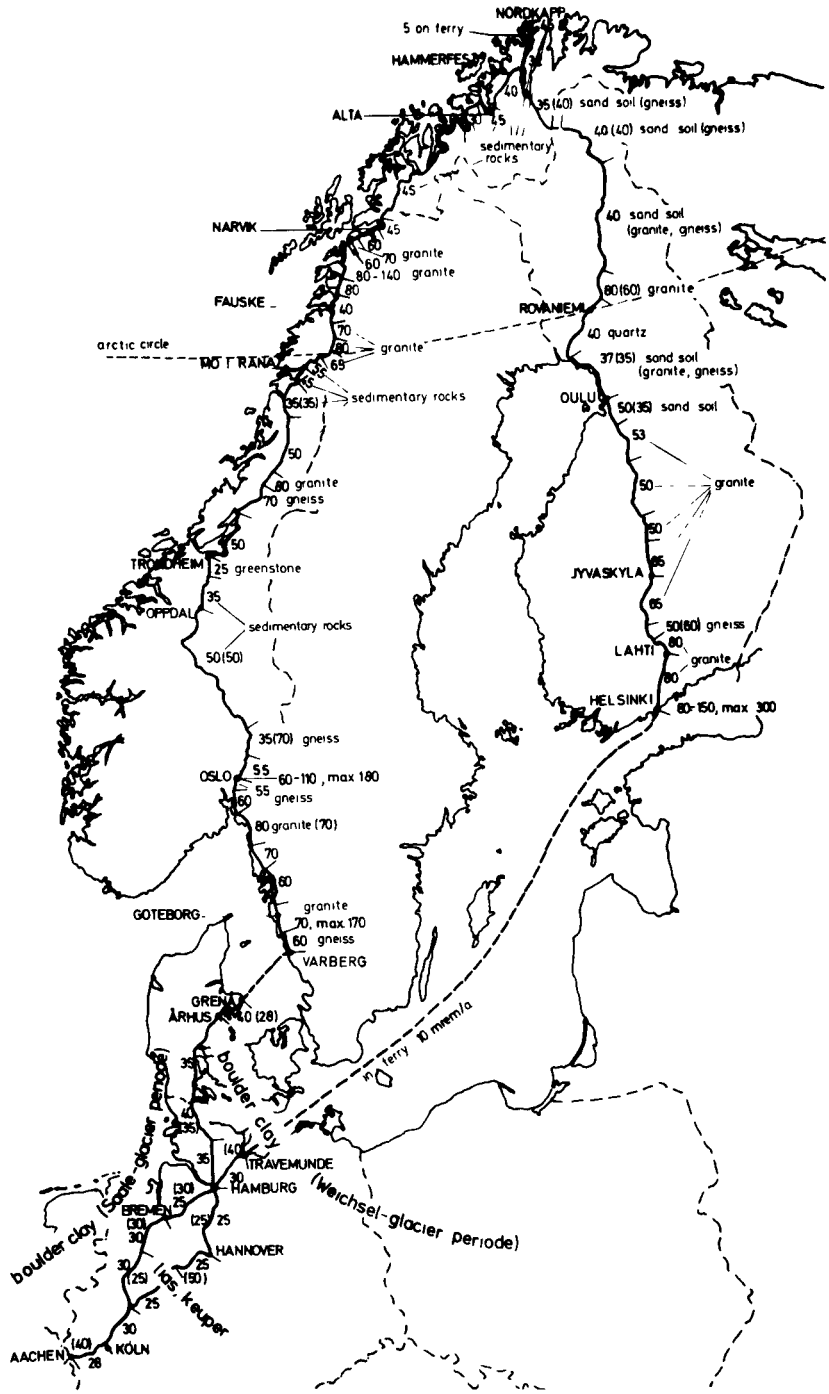

Fig. 1: Local dose rate on roads in Northern Germany, Danmark, Sweden, Norway, Finnland in mrem/a in 1979, shieldings by the car approx. 10% ( ) measured data offside the roads, outside car

NIVEAUX DE L' EXPOSITION NATURELLE EN FRANCE

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RESUME. Le Service Central de Protection contre les Rayonnements Ionisants a été amené dans le cadre d'un contrat avec la Commission des Communautés Européennes, à compléter de façon importante la carte de l'irradiation naturelle externe en France.
A cet effet, le Service Central de Protection contre les Rayonnements Ionisants a ajouté au réseau de points de mesure constitué par les stations de référence fonctionnant depuis 1967, un réseau constitué par des particuliers bénévoles choisis avec la coopération des Directions Départementales de l'Action Sanitaire et Sociale. Ainsi, près de 4000 dosimètres thermoluminescents ont été installés pour des périodes de six mois, dans tous les départements de la métropole, à l'intérieur et à l'extérieur des habitations.
Les auteurs exposent les difficultés pratiques rencontrées pour réaliser une telle étude et présentent les principaux résultats.
A titre de comparaison, deux campagnes de mesures à l'aide d'un radiamètre à scintillateur plastique ont été réalisées à Paris et dans le département de l'Yonne.

KURZFASSUNG. NATÜRLICHE STRAHLENBELASTUNG IN FRANKREICH. Der "Service Central de Protection contre les Rayonnements Ionisants" (S.C.P.R.I.) hat im Rahmen eines Vertrags mit der Kommission der Europäischen Gemeinschaften die Karte der natürlichen Bestrahlung von aussen in Frankreich wesentlich vervollständigt.
Hierzu hat der Service Central de Protection contre les Rayonnements Ionisants das seit 1967 in Betrieb stehende Messstellennetz aus Bezugsstationen um ein von freiwilligen Mitarbeitern betriebenes Netz erweitert, die gemeinsam mit der Direction Départementale de l'Action Sanitaire et Sociale ausgewählt wurden. In diesem Rahmen wurden nahezu 4000 Thermolumineszenz-Dosimeter für Zeiträume von 6 Monaten in allen Départements des französischen Mutterlandes in Wohnungen und draussen aufgestellt.
Die mit einer solchen Untersuchung verbundenen praktischen Schwierigkeiten werden erörtert und die wichtigsten Ergebnisse vorgestellt.
Zu Vergleichszwecken wurden zwei Messkampagnen mit Hilfe eines Strahlungsmessgeräts mit Kunststoffszintillator in Paris und im Département Yonne durchgeführt.

SUMMARY. EXPOSURE TO NATURAL RADIATION IN FRANCE. A contract award by the Commission of the European Communities has involved the S.C.P.R.I. (Service Central de Protection contre les Rayonnements Ionisants) in completing in some detail the map of external natural radiation for France.

For this purpose, the S.C.P.R.I. added to the network of measurement locations comprising the reference points in operation since 1967, a network established with the assistance of private individuals selected in cooperation with health authorities in the various départements (counties). Nearly 4000 TLDs have thus been installed for periods of 6 months, in all départements in France, both inside and outside dwellings. The authors present the practical difficulties in performing such a study, and present the principal results.

For the purposes of comparison, two sets of measurements have been performed with a plastic scintillator, in Paris and in the département of Yonne.

Nous avons exposé à maintes reprises, et notamment au cours de la présente réunion (1), les réserves qu'il convenait de faire quant à la signification de prétendues mesures précises de l'exposition aux rayonnements d'origine naturelle, nous n'y reviendrons pas ici. Depuis plus de 15 ans, le SCPRI a acquis une longue expérience au niveau de ses différents réseaux de surveillance des expositions éventuelles de tous ordres tant sur le territoire français qu'à l'échelle internationale dans le cadre de la mission que lui a confiée l'OMS (IRC). Les résultats des mesures effectuées ont en particulier confirmé l'allure générale, par ailleurs connue dans ses grandes lignes, de la carte de France de l'exposition naturelle.

Néanmoins, afin de disposer de mesures réalisées dans des conditions aussi comparables que possible entre les différents pays de la Communauté Européenne, cette dernière a confié une étude d'harmonisation des résultats au SCPRI qui a mis en place un réseau spécialement étudié à cet effet.

TECHNIQUE DE MESURE

Le dosimètre thermoluminescent présente des caractéristiques particulièrement adaptées aux mesures d'irradiation ambiante et notamment dans l'environnement des sites nucléaires.

Une intercomparaison a été organisée en 1976 par GAIL DE PLANQUE et T.P. GESEIL (2) concernant 26 pays utilisant essentiellement le dosimètre thermoluminescent (TLD) pour les mesures de faibles doses résultant de l'irradiation ambiante. Cette intercomparaison montre que le TLD, Fluorure de Lithium, Fluorure de Calcium ou Sulfate de Calcium est sensible à toutes les composantes du rayonnement naturel, cosmique et tellurique. On connaît bien, cependant, les défauts des TLD avec lesquels il n'est possible de conserver une excellente qualité de mesure, lorsqu'on utilise un grand nombre de dosimètres (les réponses intrinsèques de chaque dosimètre différant sensiblement les unes des autres), qu'en pratiquant des contrôles et des tris continus.

Le dispositif que nous avons adopté pour réaliser ces mesures comprend :

- un film dosimétrique dans son étui comportant un écran de 1 mm de Plomb et un écran de 0,2 mm de Cuivre
- une pastille de Fluorure de Lithium

- une pastille de Fluorure de Calcium.

Ces éléments sont placés dans une pochette en vinyle hermétiquement soudée.

Le film constitue un témoin des conditions dans lesquelles a été gardé le dispositif dosimétrique.

L'ensemble peu encombrant peut être distribué en grand nombre à des particuliers qui à l'aide de consignes faciles et uniformes le mettent en place pour une durée de six mois dans leur appartement ou leur jardin.

A chaque envoi est joint un questionnaire (fig.1) destiné à déterminer dans quelles conditions sont placés les dosimètres (intérieur ou extérieur, nature des matériaux de construction, nature du sol ...). Ce questionnaire a été réalisé en collaboration avec le Commissariat à l'Energie Atomique (CEA) qui, dans le même cadre, a entrepris une opération de mesures davantage axée sur l'étude de la contribution des matériaux de construction (3).

Les tâches ont été réparties de la façon suivante : le CEA s'est chargé d'explorer un petit nombre de départements à la fois avec de nombreux points de mesure (de 300 à 500 par département), tandis que le SCPRI couvrait la majeure partie de la France à raison d'environ 40 dosimètres par département.

MISE EN PLACE DES DOSIMETRES

A cet effet, au réseau des stations du SCPRI déjà équipées de dosimètres, mis en place depuis 1966, a été ajouté dès février 1977, le réseau de dosimètres installés chez des particuliers avec la collaboration des Directions Départementales de la Protection Civile (DDPC) et des Directions Départementales des Affaires Sanitaires et Sociales (DDASS) dans l'ensemble des départements français. En 1978, 86 départements français ont ainsi participé à la mise en place des dosimètres durant les périodes d'avril à octobre et de juin à décembre.

Il est demandé à chaque DDASS de choisir 20 personnes résidant dans le département, acceptant de garder l'un des dosimètres dans une des pièces de son logement et l'autre à l'extérieur, le questionnaire décrit plus haut permettant de localiser sans équivoque l'implantation des dosimètres au moment de leur interprétation.

Les personnes qui ont accepté de recevoir des dosimètres appartiennent dans la majorité des cas aux services locaux de la Santé Publique : chefs de service, médecins inspecteurs, assistantes sociales, inspecteurs de salubrité, etc, toutes personnes conscientes des nécessités d'une enquête officielle.

Une telle mise en place ne se réalise cependant pas sans difficultés. Il y a lieu en particulier de veiller de très près au choix judicieux des points d'implantation, afin d'éviter notamment le regroupement d'un nombre trop important de dosimètres en un même lieu à l'échelon d'une région.

Afin d'être à l'abri d'inégalités d'exposition au rayonnement naturel durant le transport, les dosimètres sont expédiés par colis postal dans chaque département au Directeur Départemental des Affaires Sanitaires et Sociales ou à son représentant délégué qui se charge de la répartition. Afin de s'assurer qu'au cours du transport les dosimètres ne voyagent pas à côté de colis contenant des sources radioactives, une enveloppe contenant deux dosimètres témoins est jointe à l'envoi. Elle doit être retournée au SCPRI par le responsable départemental dès la réception du colis. A l'arrivée au SCPRI, ces dosimètres témoins sont immédiatement développés pour vérifier qu'aucune irradiation accidentelle ne s'est produite au cours du transfert. De même, au retour, une enveloppe contenant deux dosimètres est adressée au responsable départemental, qui la joindra à son expédition. A ce jour, aucune exposition anormale durant le transport n'a été enregistrée.

Ainsi, nous disposons pour 1978 de 865 valeurs de dose enregistrées à l'extérieur d'habitations dans 79 départements et 946 à l'intérieur d'habitations dans 68 départements.

RESULTATS

Les résultats correspondant aux périodes d'avril à octobre ou juin à décembre 1978 sont rassemblés dans les tableaux présentés ci-après. Les Tableaux I et II résument les principales données pour chaque département, nombre de points de mesure, valeurs minimale, maximale et moyenne exprimées en millirads par an, contribution du rayonnement cosmique comprise.

A l'extérieur des habitations (Tab.I) pour 865 valeurs de dose réparties dans 79 départements, on trouve une moyenne de 99 millirads par an.

La valeur annuelle moyenne maximale est observée dans le département de la Corse avec 186 millirads/an et le maximum ponctuel dans ce même département (280 millirads/an).

A l'intérieur des habitations (Tab.II) pour 946 valeurs relevées dans 68 départements on obtient une moyenne de 105 millirads/an, ce qui semble confirmer que, dans l'ensemble, la contribution des radioéléments naturels présents dans les matériaux de construction est légèrement plus importante que l'effet d'écran produit par les murs vis à vis du rayonnement d'origine tellurique.

C'est dans le département de la Haute Vienne qu'apparaissent la plus forte moyenne (191 millirads/an) et la plus forte valeur ponctuelle (376 millirads/an).

On note également des valeurs moyennes relativement élevées dans les départements de la Corse, du Morbihan, des Hautes Pyrénées, de la Haute Loire, de la Corrèze et des Vosges avec respectivement 180, 169, 167, 156, 155 et 150 millirads/an.

Dans le tableau III sont présentées les différentes valeurs moyennes pour chaque département ainsi que les rapports entre les doses à l'intérieur et à l'extérieur des habitations.

Les figures 2 et 3 montrent l'ensemble des résultats sur toute la France à l'extérieur et à l'intérieur des habitations.

L'expérience ainsi amorcée a, dans l'ensemble, donné toute satisfaction; elle est poursuivie en 1979 dans de bonnes conditions. Cette méthode relativement simple à mettre en oeuvre pourra être utilisée pour d'autres pays : c'est le cas actuellement pour la Belgique.

Ainsi que l'on pouvait s'y attendre, la carte de l'irradiation ambiante externe en France se révèle conforme à la configuration géologique du pays. C'est ainsi que la Corse, le Massif Central, le Massif Armoricain, les Vosges ont des niveaux d'irradiation dépassant notablement la moyenne. Les Landes, l'Ain, l'Aube se situent par contre en dessous de 50 millirads/an.

CONCLUSIONS

La première ébauche d'une carte de l'exposition naturelle en France, réalisée dans des conditions assurant une cohérence satisfaisante avec les mesures effectuées dans les autres pays de la Communauté, n'apporte aucune surprise : elle concorde avec les données géologiques, comme on pouvait s'y attendre.

Quant à l'exposition à l'intérieur des habitations, elle diffère peu de l'exposition extérieure; cette situation est assez différente de celle retrouvée dans d'autres pays, notamment en RFA; cela tient sans doute au caractère plus traditionnel de l'habitat en France.

Ces résultats ne représentent cependant qu'une première étude, encore très partielle. L'acquisition de nouveaux résultats, dans les mois à venir, permettra de compléter définitivement la carte, et de juger par ailleurs la stabilité de l'exposition au cours du temps.

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TABLEAU I : Doses relevées à l'extérieur des habitations

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Ain	5	44	60	48
Aisne	15	56	130	89
Allier	13	112	186	148
Alpes de Haute Provence	17	58	118	82
Hautes Alpes	9	70	174	99
Alpes Maritimes	5	52	180	71
Ardèche	2	52	58	55
Ardennes	18	58	88	71
Ariège	7	120	176	140
Aube	15	48	70	55
Aude	6	62	114	91
Aveyron	15	62	112	85
Calvados	9	62	122	89
Cantal	2	126	138	132
Charentes	6	62	144	94
Charentes Maritimes	19	62	194	98
Cher	14	64	88	76
Corrèze	12	130	200	150
Corse	12	120	280	186
Côte d'Or	8	68	126	92
Côtes du Nord	14	104	208	133
Dordogne	17	52	172	108
Doubs	14	80	144	113
Drôme	4	40	40	40
Eure	12	48	78	63
Finistère	10	88	250	115
Gard	14	58	262	108

TABLEAU I (suite 1)

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Haute Garonne	10	104	154	130
Gers	18	46	196	123
Gironde	10	60	126	99
Hérault	15	86	168	103
Ille et Vilaine	19	76	146	118
Indre	11	68	222	91
Indre et Loire	8	44	132	80
Isère	8	44	104	68
Jura	12	40	92	77
Landes	18	36	88	47
Haute Loire	2	122	156	129
Loire Atlantique	2	98	114	106
Loiret	15	48	102	75
Lot	17	40	136	97
Lot et Garonne	3	78	88	81
Maine et Loire	15	50	88	71
Manche	3	78	88	83
Marne	9	74	96	79
Haute Marne	12	78	146	84
Mayenne	19	58	108	86
Meurthe et Moselle	18	104	182	134
Meuse	15	78	118	96
Morbihan	19	102	228	176
Moselle	7	70	104	89
Nièvre	15	52	160	89
Nord	1	44	44	44
Orne	20	50	82	66
Pas de Calais	2	38	60	49

TABLEAU I (suite 2)

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Puy de Dôme	4	98	166	129
Hautes Pyrénées	18	110	170	148
Pyrénées Orientales	7	88	140	121
Bas Rhin	14	74	110	85
Haut Rhin	11	92	194	127
Haute Saône	9	82	158	121
Saône et Loire	18	60	210	106
Haute Savoie	9	56	86	73
Seine Maritime	15	50	76	63
Seine et Marne	14	54	80	66
Deux Sèvres	11	58	132	97
Tarn	14	66	104	77
Var	4	48	70	60
Vaucluse	3	46	66	54
Vendée	16	64	162	101
Vienne	12	66	224	101
Haute Vienne	15	114	278	167
Vosges	8	94	168	128
Yonne	21	74	158	98
Territoire de Belfort	10	48	106	76
Région Parisienne	15	62	118	75

3)

TABLEAU II : Doses relevées à l'intérieur des habitations

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Aisne	14	54	126	83
Allier	19	70	256	128
Alpes de Haute Provence	16	50	192	85
Hautes Alpes	10	64	148	93
Alpes Maritimes	7	50	124	82
Ardennes	21	36	100	71
Ariège	13	106	172	134
Aube	16	40	98	62
Aude	10	112	174	135
Aveyron	20	58	118	80
Bouches du Rhône	1	66	66	66
Calvados	13	48	136	86
Cantal	2	110	120	115
Charentes	13	46	172	113
Charentes Maritimes	17	74	158	106
Cher	18	52	98	80
Corrèze	16	106	242	155
Corse	12	88	290	180
Côte d'Or	9	66	150	95
Côtes du Nord	19	94	232	133
Dordogne	19	54	164	112
Doubs	19	74	192	124
Eure	11	46	92	63
Finistère	9	54	158	82
Gard	12	48	160	94
Haute Garonne	11	122	166	126
Gers	19	90	252	147

TABLEAU II (suite 1)

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Gironde	16	4[illegible]	188	107
Hérault	14	8[illegible]	170	120
Ille et Vilaine	19	7[illegible]	188	129
Indre	12	7[illegible]	142	115
Indre et Loire	9	7[illegible]	112	93
Jura	18	4[illegible]	76	58
Landes	17	5[illegible]	84	73
Haute Loire	6	10[illegible]	176	156
Loire Atlantique	16	9[illegible]	166	126
Lot	16	9[illegible]	142	128
Lot et Garonne	7	5[illegible]	100	79
Maine et Loire	15	5[illegible]	128	76
Manche	5	56	98	77
Marne	15	66	126	81
Haute Marne	17	58	144	90
Mayenne	19	60	106	85
Meurthe et Moselle	20	84	214	139
Meuse	16	58	152	108
Morbihan	20	114	246	169
Moselle	13	72	156	100
Nièvre	17	80	210	108
Orne	20	46	132	69
Puy de Dôme	4	100	178	139
Pyrénées Atlantiques	1	84	84	84
Hautes Pyrénées	16	110	198	167
Pyrénées Orientales	9	108	164	136
Bas Rhin	19	64	122	89
Haute Saône	9	78	142	116

TABLEAU II (suite 2)

Département	Nombre de points	Minimum (mrad/a)	Maximum (mrad/a)	Moyenne (mrad/a)
Saône et Loire	18	84	188	108
Haute Savoie	12	48	98	64
Seine Maritime	16	46	86	57
Seine et Marne	15	50	80	72
Deux Sèvres	14	42	132	83
Tarn	18	60	124	82
Var	6	46	72	60
Vendée	19	48	134	101
Vienne	19	70	114	90
Haute Vienne	16	124	376	191
Vosges	9	94	194	150
Yonne	22	58	144	99
Territoire de Belfort	11	52	100	80

Tableau III : Comparaison des résultats moyens à l'intérieur et à l'extérieur des habitations.

Département	Dose annuelle moyenne à l'extérieur des habitations (mrad/a)	Dose annuelle moyenne à l'intérieur des habitations (mrad/a)	Rapport $\frac{\text{Dose intérieure}}{\text{Dose extérieure}}$
Aisne	89	83	0,93
Allier	148	128	0,86
Alpes de Haute Provence	82	85	1,04
Hautes Alpes	99	93	0,94
Alpes Maritimes	71	82	1,15
Ardennes	71	71	1
Ariège	140	134	0,96
Aube	55	62	1,13
Aude	91	125	1,48
Aveyron	85	80	0,94
Calvados	89	86	0,97
Cantal	132 *	115 *	0,87
Charentes	94	113	1,20
Charentes Maritimes	98	106	1,08
Cher	76	80	1,05
Corrèze	150	155	1,03
Corse	186	180	0,97
Côte d'Or	92	95	1,03
Côtes du Nord	133	133	1,00
Dordogne	108	112	1,04
Doubs	113	124	1,10
Eure	63	63	1,00
Finistère	115	82	0,71
Gard	108	94	0,87
Haute Garonne	130	126	0,97
Gers	123	147	1,20

Tableau III (suite 1)

Département	Dose annuelle moyenne à l'extérieur des habitations (mrad/a)	Dose annuelle moyenne à l'intérieur des habitations (mrad/a)	Rapport Dose intérieure / Dose extérieure
Gironde	99	102	1,08
Hérault	103	120	1,17
Ille et Vilaine	118	129	1,09
Indre	91	115	1,24
Indre et Loire	80	93	1,16
Jura	77	58	0,75
Landes	47	73	1,55
Haute Loire	129 *	156	1,21
Loire Atlantique	106 *	126	1,19
Lot	97	128	1,32
Lot et Garonne	81 *	79	0,98
Maine et Loire	71	76	1,07
Manche	83 *	77	0,93
Marne	79	81	1,03
Haute Marne	84	90	1,07
Mayenne	86	85	0,99
Meurthe et Moselle	134	139	1,04
Meuse	96	108	1,13
Morbihan	176	169	0,96
Moselle	89	100	1,12
Nièvre	89	108	1,21
Orne	66	69	1,05
Puy de Dôme	129 *	139 *	1,08
Hautes Pyrénnées	148	167	1,13
Pyrénnées Orientales	121	136	1,12
Bas Rhin	85	89	1,05
Saône et Loire	106	108	1,02

Tableau III (suite 2)

Département	Dose annuelle moyenne à l'extérieur des habitations (mrad/a)	Dose annuelle moyenne à l'intérieur des habitations (mrad/a)	Rapport Dose intérieure / Dose extérieure
Haute Saône	121	116	0,96
Haute Savoie	73	116	1,59
Seine Maritime	63	57	0,90
Seine et Marne	66	72	1,11
Deux Sèvres	97	83	0,86
Tarn	77	82	1,06
Var	60 *	60	1,00
Vendée	101	101	1,00
Vienne	101	90	0,89
Haute Vienne	167	191	1,14
Vosges	128	150	1,17
Yonne	98	99	1,01
Territoire de Belfort	76	80	1,05

* résultats peu significatifs (moins de 5 points de mesure)

- Figure 1 - Questionnaire relatif aux mesures par intégration chez des particuliers

Ne rien écrire dans cette colonne

QUESTIONNAIRE RELATIF AU DOSIMETRE

PLACE A L'INTERIEUR DES HABITATIONS

Remplir le questionnaire en écrivant en lettres capitales et en mettant une croix dans les cases correspondant aux réponses.

Numéro du dosimètre : .....

	Jour	Mois	Année
Date de mise en place			
Date de relevé			

ND (1 2 3 4)

COM (6 10)

CX (12 17 21)

CY (23 28 32)

DS (34 38)

FS-EV (40 44)

MEP (46 50)

REL (52 56)

AR-DS (58 62)

DEV (64 68)

- NOM de la personne chez laquelle est placé le dosimètre

M., Mme, Melle : .....
.....

- Localisation géographique

Commune : .....
Lieu dit : .....
(éventuellement)
N° : ..... Rue : .....
Code postal : .....

EMPLACEMENT DU DOSIMETRE

I. Le dosimètre doit être placé dans une salle de séjour, dans un meuble en bois. Avez vous pu le placer :

- dans une salle de séjour *oui* [1] *non* [2] — SDS (70)
- dans un meuble en bois *oui* [1] *non* [2] — MEB (72)

Si vous avez répondu non à la dernière question, préciser la nature du support du dosimètre :
.....

AMEB (74)

II. Le meuble (ou support) où est placé le dosimètre est-il :

- contre un mur de gros-oeuvre [1]
- contre une cloison [2]
- dans une autre position [3]

PLS (76 77)

III. Indiquer l'étage où est placé le dosimètre :

- sous-sol [1]
- rez-de-chaussée [2]
- en étage [3]

lequel : .....

ND (1 4)

ET (6)

NUET (8 9)

T.S.V.P.

- Figure 1 (suite)

Ne rien écrire dans cette colonne

CONSTRUCTION (Répondre à l'une ou l'autre des rubriques V et VI selon que votre habitation est un pavillon ou un immeuble)

V. Votre habitation est un pavillon oui [ ] 1

Est-il construit :
- sur vide sanitaire [ ] 1
- sur sous-sol [ ] 2

Est-ce que sa construction est de type industriel ?

oui [ ] 1 *non* [ ] 2

SSPAV [ ] 13

TYCO [ ] 11 PAVIND [ ] 15

VI. Votre habitation est un immeuble oui [ ] 2

Précisez le nombre d'étages : .....

NBET [ ][ ] 17 18

VII. Année de construction de votre habitation : .....

Si vous ne savez pas exactement, pouvez vous préciser si c'est :

- avant 1900 [ ] 1
- de 1900 à 1945 [ ] 2
- après 1945 [ ] 3
- date inconnue [ ] 4

DCONS [ ][ ][ ] 20 22

MATERIAUX (Cette rubrique concerne la pièce où se trouve le dosimètre)

VIII. Gros-oeuvre (murs)

- bois [ ] 1
- briques [ ] 2
- béton [ ] 3
- parpaings [ ] 4
- granite [ ] 5
- meulière [ ] 6
- pierre de taille (calcaire) [ ] 7
- pierre du pays * [ ] 8
- autre matériau : .....
- constitution inconnue [ ] 9

GOEV [ ][ ][ ][ ][ ] 24 28

AMG [ ] 29

IX. Cloison

- bois [ ] 1
- briques [ ] 2
- plâtre [ ] 3
- parpaings [ ] 4
- béton [ ] 5
- autre matériau : .....
- constitution inconnue [ ] 6

CLOIS [ ][ ][ ][ ][ ] 31 35

X. Plancher

. Support :

- hourdis [ ] 1
- dalle de béton [ ] 2
- planches sur partie de bois [ ] 3
- autre :.....

AMC [ ] 36

PLASU [ ][ ] 38 39

. Revêtement :

- parquet [ ] 1
- carrelage [ ] 2
- dallage * [ ] 3
- moquette [ ] 4
- plastique [ ] 5

autre matériau : .....

PLARE [ ][ ] 41 42

* Si vous pouvez préciser la nature de la pierre (schiste, grès, etc...) indiquer la à la rubrique autre matériau.

IPESB [ ][ ][ ][ ][ ][ ]– 44 49

- Figure 1 (suite)

Ne rien écrire dans cette colonne

QUESTIONNAIRE RELATIF AU DOSIMETRE PLACE A L'EXTERIEUR DES HABITATIONS

Remplir le questionnaire en écrivant en lettres capitales et en mettant une croix dans les cases correspondant aux réponses.

Numéro du dosimètre : .....

Date de mise en place / Date de relevé			
	Jour	Mois	Année

- NOM de la personne chez laquelle est placé le dosimètre

  M., Mme, Melle : .....
  .....

- Localisation géographique

  Commune : .....
  Lieu dit : .....
  (éventuellement)
  N° : ..... Rue : .....
  Code postal : .....

ID 1 2 3 4

COM 6 10

CX 12 17 21

CY 23 28 32

DS 34 38

FS-EV 40 44

MEP 46 50

REL 52 56

AR-DS 58 62

DEV 64 68

(1) Distance sol dosimètre

Le dosimètre doit être approximativement placé à 1 mètre du sol. Si et seulement si cette distance est notablement différente, veuillez l'indiquer ici : .....

DSOL 70 71

(2) Distance mur - dosimètre

Le dosimètre doit être placé à une distance minimale de 5 m. d'un mur. Si cette distance est notablement inférieure, veuillez l'indiquer ici : .....

DMUR 72

(3) Nature du sol à l'aplomb du dosimètre

terre cultivée	1	*béton*	5
terre battue	2	*gravier*	6
pelouse	3	*dallage* *	7
macadam	4		

autre matériau : .....

NSO (panaché 8) 74

(4) Nature du support sur lequel est fixé le dosimètre

bois	1	*béton*	4
métal	2	*pierre*	5
plastique	3	*autre*	6

NSUP (panaché 7) 75

ERESB 76 80

* si vous pouvez préciser la nature de la dalle (schiste, grès, marbre, etc...) indiquer la à la rubrique autre matériau.

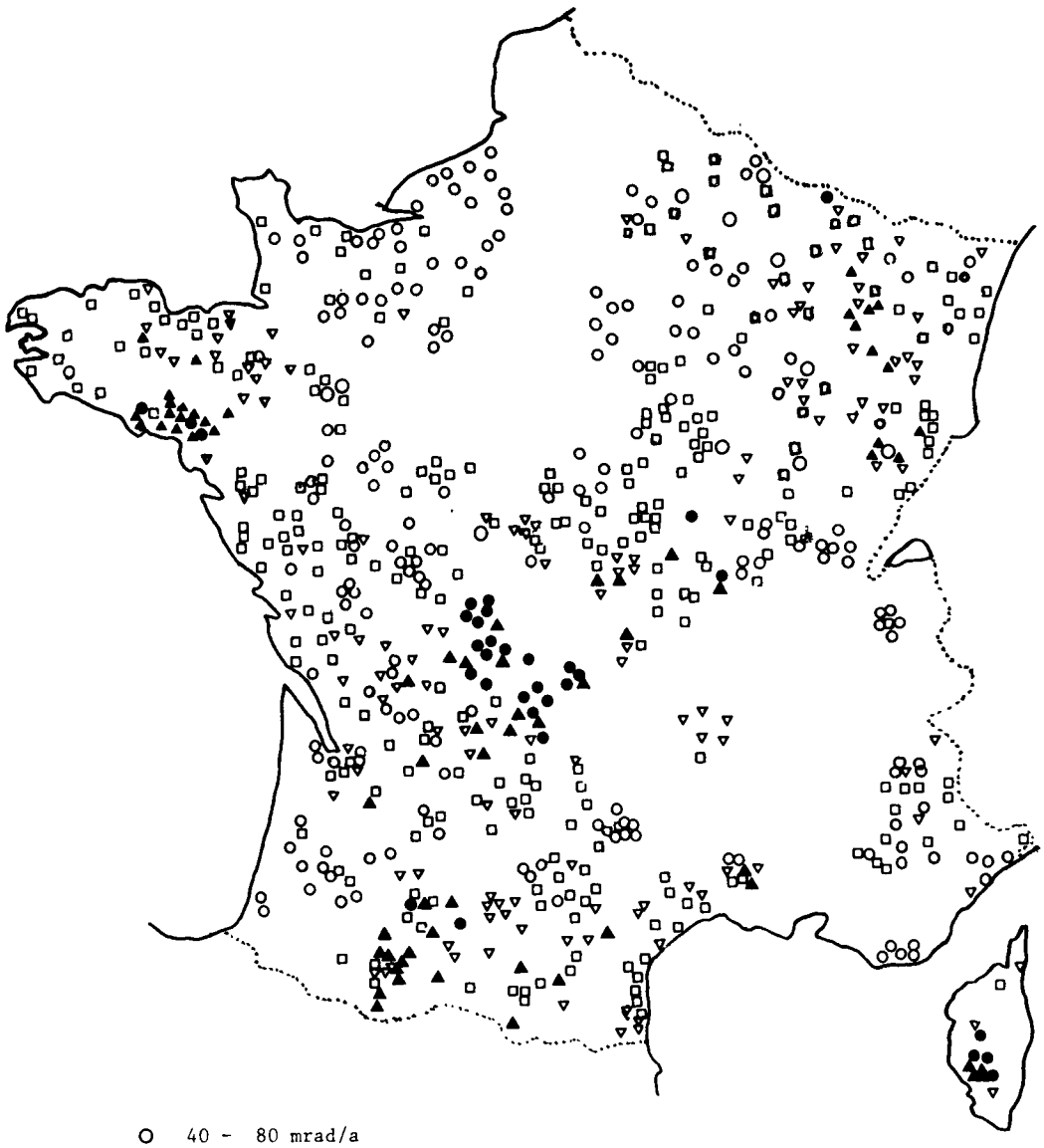

Fig. 2 : Irradiation naturelle à l'extérieur des habitations.
Dose intégrée sur une période de 1 an (mrad).

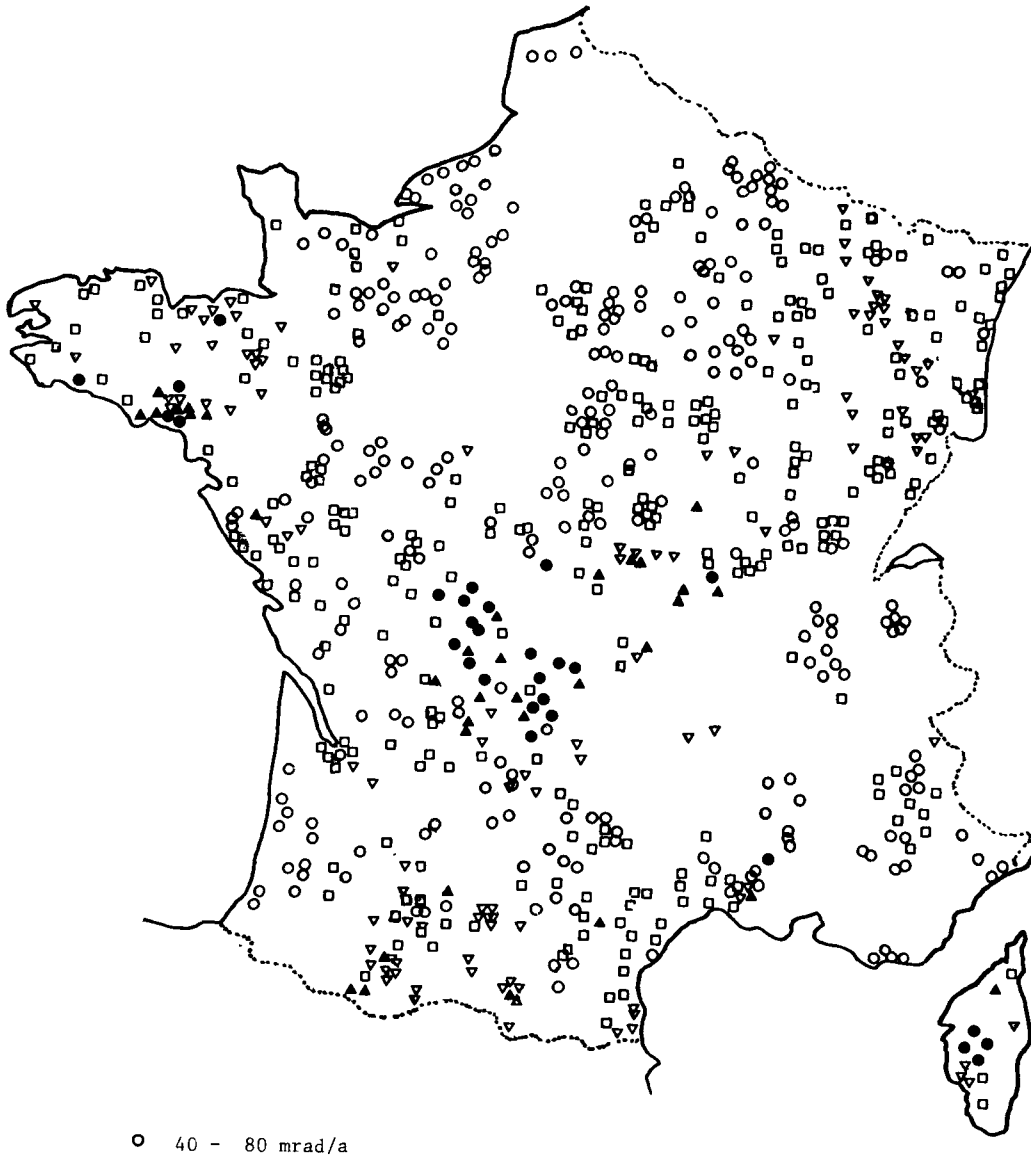

Fig. 3 : Irradiation naturelle à l'intérieur des habitations.
Dose intégrée sur une période de 1 an (mrad).

EXTERNAL NATURAL RADIATION EXPOSURE IN AUSTRIA

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1. Cosmic and terrestrial radiation.

In collaboration with the Austrian ministry for health a survey was carried out during the years 1970-1974 to measure the external doses of natural radiation in communities with a population of more than 3000 people. Measurements were made in 600 towns and villages spread over the whole territory of Austria.
As measuring equipment a Geiger Müller tube with a pulse counter was used.In each community the instrument was carried trough the streets to measure the dose for an exposure time of at least 30 minutes (in larger cities measuring times of several hours had to be used).So for every place an average value of the radiation dose independent of local fluctuations was measured.
Results ("Strahlenkarte Österreichs 1975"):
Mean dose for each member of the Austrian population:

78 mR/y total dose
43 mR/y terrestrial dose

Maximum: 190 mR/y (in a village,built on granite) total dose.
Minimum: 34 mR/y (in villages on alluvial soils) total dose.

2. Indoor doses of external radiation.

A second investigation in collaboration with the ministery for health was carried out during the years 1975-1978.More than 6000 single measurements in 1900 buildings in all parts of our country were made.As measuring equipment a RSS 111 high pressure ionization chamber was used to measure the total dose.
Results:

Average doses for buildings made of various material:
Brick ... 12,4 µR/h,concrete ... 9,3 µR/h,wood ... 8,6 µR/h
natural stones ... 12,5 µR/h.

* Data for Austria introduced from the floor

Mean indoor dose for each member of the Austrian population:

1o7 mR/y

Maximum: 338 mR/y (brick house in granite formation).

Minimum: 44 mR/y (concrete houses in alluvial formations).

Mean values for the Austrian"Bundesländer":

Burgenland	94,6 mR/y	Salzburg	88,5 mR/y
Kärnten	131,8 mR/y	Steiermark	113,o mR/y
Oberösterreich.	1o8,6 mR/y	Tirol	1o5,1 mR/y
Niederösterr. .	1o6,9 mR/y	Vorarlberg	89,4 mR/y
		Wien	1o6,9 mR/y

3. Specific activity of natural soils and their radiation.

In 1977 we made measurements of the specific activity of characteristic soils by means of gamma spectrometrie.Furthermore,on every place,where samples had been taken,the terrestrial radiation was measured.These values were compared with the calculated dose rates from the samples.Both results agree well.

Some average values are:

Granite:	U 238 series .... 1,84 pCi/g Th 232 series ... 1,32 pCi/g K 4o .....32,7 pCi/g	Dose rate 1 m above granite = 12,1 uR/h
Sandstone:	U 238 series .. o,6 pCi/g Th 232 series.. o,8 pCi/g K 4o .....12,6 pCi/g	Dose rate 1 m above sandstone = 5,3 uR/h
Limestone:	U 238 series .. o,84 pCi/g Th 232 series . o,o8 pCi/g K 4o ..... 2,2 pCi/g	Dose rate 1 m above limestone = 2,2 uR/h

From these and some other results for the terrestrial dose above geological formations we calculate the average value for the terrestrial dose obove natural soils in Austria to be 42,o mR/y (4,8 uR/h).

The contribution to this dose are 4o % from the Th 232 series, 4o % from K 4o and 2o % from the U 238 series.

SOME CONSIDERATIONS ON THE CALCULATION OF THE NATURAL RADIATION DOSE FOR DIFFERENT BODY ORGANS. AVERAGE VALUES FOR SWITZERLAND.

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Summary:

From measurements of the terrestrial and cosmic radiation made in our country some years ago and from considerations on the average concentration of the natural occuring radionuclides in the human body, a mean value of the dose rate for the natural radiation at three different organs was determined. The equivalent dose rate for the red bone marrow amounts to about 143, for the ovaries to about 114 and for the testes to about 100 mrem/year (average gonadal dose 107 mrem/year - 0,107 mSv/y).

Considerable efforts have been made in our country over the last few years to determine the average gonad doses and bone marrow doses resulting from radiodiagnostic examinations of patients.

At present, an average bone marrow dose for adults, as well as for new-born and 12 year-old children, is being determined using 3 different phantoms. The first results will appear in spring 1980.

To permit specific comparision between organ doses due to background and those caused by medical or industrial use of ionizing radiation, the background dose, generally determined in the open, must be converted into "organ doses". This corresponds also to the recommendations of the ICRP-report Nr. 26.

* Data for Switzerland introduced from the floor

The absorbed dose in the organs resulting from the gamma components of terrestrial radiation was determined using the MIRD technique. If the dose in the open air is specified as 100%, then the following dose values were calculated:

- Dose in red marrow : 82,5%
- Testicular doses : 68,5%
- Ovarian dose : 57,0%

For Switzerland, approx. 70 mrem/year can be assumed as the average value for the terrestrial components determined in the open air. To calculate this value, a conversion factor R/rad = 0,957 was used. The additional dose rate of approx. 3 mrem/year produced in the open air by the fall-out components at the time of the measurements (1961), was then deducted from this.
To determine the actual radiation burden produced by the terrestrial components in the three above-mentioned organs, a "housing factor" was calculated for the country, taking into account 12 different "living situations".
One of these subfactors, namely, that for the "additional contribution of massive houses", was derived from the excellent series of measurements carried out by Kolb and collaborators in West Germany (the few Swiss measurements were insufficient, and the houses and house materials in South Germany and Switzerland are very similar).
The Swiss "housing factor" is 1.13 (giving a 13% increase in the organ dose rate).

Intracorporeal components

If a homogeneous distribution of the radionuclides in our body (K-40, Po-210, U-238 series, C-14, Rb-87, Ra-226 and Ra-228 with the decay products Rn-222 and Rn-220, Na-22, Be-7 etc) is assumed, the total equivalent dose rate would be approx. 35 mrem/year.

If, however, the nuclide concentrations actually present in the testes, ovaries and in the red marrow are considered, then we obtain the following values:

- testes (mean weight 35 g): approx. 24 mrem/year
- ovaries (weight of both ovaries 11 g): approx. 19 mrem/year
- red marrow (1'500 g): approx. 42 mrem/year

Cosmic components

Below 2'000 m, the majority of the dose is caused by muons and electrons (ratio 4:1). The cosmic radiation dose recorded in Switzerland in 1961, was due exclusively to these components *). The quality factor rad/rem is dependent on the radiation quality, that is of the altitude of the different regions. We have calculated Q = 1.06 as the average value for Switzerland.
The equivalent dose rate produced by cosmic radiation thus amounts to 36 mrem/year (0.7 mrad/y for the n, included).

The values of the average organ dose rate for the natural radiation for Switzerland (mrem/year) are shown in table 1.

As is well known, when comparing medically caused and natural radiation burdens it must be taken into consideration that, for the same total dosage, the biological effects that might be produced by medical applications can be more intensive than those produced by natural radiation.

The dose rate is, in fact, very different: for example, the average natural dose of 100 mrad for the ovaries accumulates over a year, while the same dose produced in the same organ by a colon X-ray picture is given in a fraction of a second.

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*) *Commencing in 1979, TLD measurements were carried out in various parts of Switzerland. The recorded values are somewhat lower than those for 1961.*

Or, for instance: the dose rate of natural radiation in the bone marrow amounts to approx. $5 \cdot 10^{-6}$ mrad/sec; while that of an X-ray of the pelvis (60 kV, 100 mA, 0,6 sec exposure time) is 100 mrad/sec.

On the basis of experiments on animals, we have therefore selected a minimum biological factor of 3. That means: for an X-ray picture of the lumbar vertebral column (centred on the 5th vertebra), the bone marrow dose amounts on average to approx. 125 mrad per picture and the testicular dose approx. 48 mrad. If, for practical purposes these values are compared to those for "natural radiation", then it must be taken into consideration that, for example, the "natural" bone marrow dose of approx. 143 mrad/year could have a biological effect which is at least three times less than that of the "medical" 125 mrad mentioned in the example.
Using the formula for the calculation of the Genetically Significant Dose (GSD), it can easily be deduced that the GSD for the natural radiation is the (mean) annual dose produced by this radiation in the gonads (D = 107 mrem for Switzerland, which is one reason why it was necessary to derive the most precise value possible for the gonad dose from the measured data for natural radiation). The investigation carried out in Switzerland to determine the Genetically Significant Dose as a result of radiodiagnostic examinations give a value of approx. 43 mrem/year. From the purely physical point of view, the medically produced GSD for the population would therefore be approx. 2,5 times smaller than the naturally produced GSD. In relation to the expected biological consequences, however, the radiodiagnostic examinations (on average 1.4 examinations per person per year in Switzerland) results as much as the double (or more) as the annual natural GSD (43·3 = 130 mrem/year, i.e. even more than 107 mrem/year).

BACKGROUND RADIATION

AVERAGE DOSE RATES FOR SWITZERLAND (mrem/year)

ORGAN	COMPONENT			TOTAL
	TERRESTRIAL	INTERNAL	COSMIC	
ACTIVE RED BONE MARROW	65	42	36	143
OVARIES	54	24	36	114
TESTES	45	19	36	100
MEASURED IN THE OPEN	70	-	36	106

AVERAGE GONADAL DOSE : ca. 107 mrem/year

A REVIEW OF INDOOR EXPOSURE FROM NATURAL RADIATION

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SUMMARY. In most circumstances, building materials are the primary source of exposure indoors. The most important radiological consequences of the natural radioactivity in building materials are irradiation of the body by gamma rays and irradiation of the lung tissues by radon-222 decay products. These consequences are explored in terms of the exposures incurred by occupants as determined by certain properties of the materials themselves and of structures as a whole. Secondary sources of indoor radiation are also touched upon.

RESUME. REVUE DE L'EXPOSITION DUE AU RAYONNEMENT NATUREL DANS LES BATIMENTS. Le plus souvent, les matériaux de construction forment la principale source d'exposition à l'intérieur des bâtiments. Les conséquences radiologiques les plus importantes de la radioactivité naturelle dans les matériaux de construction sont l'irradiation du corps par les rayons gamma et celle des tissus pulmonaires par les descendants du radon-222. Ces conséquences sont examinées en terme d'exposition des occupants déterminée par certaines propriétés des matériaux eux-mêmes et des structures dans leur ensemble. Les sources secondaires de rayonnement à l'intérieur des bâtiments sont aussi abordées.

KURZFASSUNG. UBERSICHT ÜBER DIE EXPOSITION AUS NATÜRLICHER STRAHLUNG IN WOHNUNGEN. In den meisten Fällen sind Baustoffe die Hauptquelle für die Exposition in Wohnungen. Die wichtigsten radiologischen Auswirkungen der natürlichen Radioaktivität in Baustoffen sind die Bestrahlung des Körpers durch Gammastrahlen und die Bestrahlung der Lungengewebe durch die Zerfallsprodukte des Radons-222. Es wird untersucht, inwieweit diese Strahlenbelastung der Bewohner auf bestimmte Eigenschaften der Baustoffe selbst oder aber auf die Struktur der Gebäude zurückzuführen ist. Sekundäre Expositionsquellen in Wohnungen werden ebenfalls erfasst.

Introduction

My main source of information for this paper is the NEA report on radioactivity in building materials published by OECD (1). I also want to draw on an experiment that a colleague and I recently performed (2) and on a commentary on the report prepared by another colleague (3). My thesis is that measurement, modelling, and meditation are useful and essential in this area of radiation protection.

The theme of the session allows me to ignore cosmic rays, but the UNSCEAR report shows (4) that the annual absorbed dose in tissue is almost 0.3 mGy at sea level and that substantial thicknesses of masonry are required to bring about significant shielding. The dose from cosmogenic radionuclides in indoor air is entirely negligible.

In developed countries, where persons spend most of their time indoors, the strongest source of natural radiation overall is usually building materials. (I shall refer to a possible exception later.) Attention must be fixed on irradiation of the whole body by gamma rays and irradiation of the lung tissues by the short-lived daughters of ^{222}Rn. It is scarcely necessary to note that ^{40}K, ^{226}Ra and its decay products, and the series headed by ^{232}Th are the prime radionuclides.

For reasons that will become obvious, one must, when considering the exposure of occupants, take into account the material immediately subjacent to a building. This may include the fill or hardcore and the soil and even the underlying rock.

At the back of one's mind when contemplating exposure to natural radiation is the question : when should the ICRP system of dose limitation be applied? The Commission advises that it is applicable to exposures elevated above "normal" by human activities or choice of environment, and adds that judgement may be required (5).

In the absence of consensus, the NEA Group decided to estimate the extra exposure to gamma rays and radon daughters that persons incur through living indoors rather than in the open. There may be a primitive notion of normality behind this decision, but it is indicative of the general state of thinking about natural radiation. I shall therefore not pursue the matter of incremental exposure indoors but consider absolute values.

Gamma rays

There are a number of ways of determining the dose rate indoors from gamma rays. Investigators have variously used analytical, numerical, and experimental methods and combinations thereof. The dose rate varies from

place to place in a room and depends on the configuration and properties of the structural elements and on the radioactivity in the materials. At the risk of over-simplifying, one might suggest that the dose rate of interest is that which prevails at the centre of the room, and that the materials and methods used for masonry dwellings in western Europe are perforce similar. One would wish nevertheless to admit a distinction between the configuration of single-family houses and apartment blocks. I would assert therefore that radioactive content of the building materials is the key parameter, and that one might be able to establish a rough estimate of the dose rate merely by considering its value in two circumstances.

The information available indicates that the representative specific activities of extractive types of materials used traditionally in building are : 500 Bq kg^{-1} for ^{40}K, 50 for ^{226}Ra, 50 for ^{232}Th. These values are severely rounded to avoid any suggestion of undue accuracy. The nuclides of interest contribute to the irradiation of persons approximately in the ratio of their exposure rate constants, namely 1:10:15, it being assumed that radioactive equilibrium prevails. This would allow one to define an effective specific activity, S_{eff}, as the sum of the products of individual specific activities and ratios. For typical masonry, S_{eff} is about 1750 Bq kg^{-1}.

A review of the available information leads to the conclusion that the relationships shown below between the dose rates in air at the centre of a room and the effective specific activity of the building materials may be defensible. $\dot{D}$ is in units of $\mu Gy\ h^{-1}$.

$$\dot{D}_{flat} = 4 \times 10^{-5}\ S_{eff}$$

$$\dot{D}_{house} = 3 \times 10^{-5}\ S_{eff}$$

The use of two expressions, one for a room in a purpose-built flat and the other for a single-family house, maintains the distinction to which I have referred. If the specific activities of the nuclides are the same in all the masonry elements of a room, these equations apply directly : if not, the specific activities should be weighted, for each element, by its relative projected area. Gamma rays from outside are assumed not to penetrate into the room.

I shall return later to an ad hoc test of one of these formulas, but it should be noted that this approach would lead one to predict dose rates of

0.07 and 0.05 μGy h^{-1} in air for typical flats and houses. The mean of these values, 0.06 μGy h^{-1}, is virtually identical with the estimate in UNSCEAR for masonry buildings in general. If the conventional value of the ratio between dose in air and dose in the organs of interest is adopted, namely 0.7, and also the conventional value of the fraction of time spent indoors, 0.8, the annual dose in tissue from exposure inside flats becomes 0.35 mGy and inside houses 0.25 mGy.

Radon daughters

I shall pursue a simplified approach in estimating exposure to radon daughters. Daughter concentration in a dwelling is influenced by many factors, but I shall focus on a few important influences such as radium content, radon emanating fraction, the configuration of the structural elements, and the ventilation rate. I shall largely ignore plateout of radon daughters, the surface finishes and moisture content of structural elements, and factors such as air conditioning, variable ventilation, and meteorology.

The radon daughter concentration in a room attributable to its structural elements may be estimated, in an approximate manner, from this expression :

$$C_E = \frac{5 \times 10^2 \, S_{Ra} \, \eta \, \lambda \, d \rho A}{j^{3/2} \, V}$$

C_E is in units of mWL. The other symbols stand for the following quantities. S_{Ra} is the specific activity of ^{226}Ra in the material of the element; η is the ^{222}Rn emanating fraction, the fraction of radon formed that escapes; λ the decay constant of radon-222; d the half-thickness of the element; ρ the density; A the area of the element; and V the volume of the room. All of these are in SI units, but the ventilation rate j is in inverse hours. The summation symbol signifies that all the elements are to be considered.

The value of the emanating fraction η is about 0.03 for ordinary concrete and 0.1 for gypsum. For other materials, one might also use 0.1, although it would tend to be high. With a cavity wall, the appropriate value of d is half the thickness of the inner leaf.

The equation is thought to have some validity for the ventilation rates prevailing in dwellings, say 0.5 to 3 air changes per hour. Outside this range, one should estimate the activity concentration of the ^{222}Rn gas and couple that with a value of the equilibrium factor indicated by the

ventilation rate.

Unlike gamma rays, extraneous radon daughters are important. If a dwelling has a ground floor, the most powerful source of radon may be infiltration from the soil. Radon and daughters are, of course, present in ventilating air, and account must be taken of the shift towards greater radioactive equilibrium that occurs when it enters the dwelling.

To illustrate the relative contributions of these three sources to indoor concentrations, I considered typical masonry dwellings in a typical geological area with typical outside air concentrations. For a nominal ventilation rate of 1 air change per hour, I computed the contributions from the structural elements, the ground, and the incoming air (Table I). Since the table is illustrative in nature, it is more illuminating to examine the percentage columns.

A clear pattern emerges. In single-family houses and ground-floor flats, the ground is likely to be the strongest source of radon daughters. In high-rise apartments, its contribution must, in principle, be negligible, but opinions differ about the degree to which flats are isolated from the stairwell, lift-shaft, and service trunking. It would appear, that in typical masonry dwellings, the ventilating air might contribute as much, if not more than, the structural materials. (Indeed, I feel that I have overestimated the contribution from the elements.) There may be pointers here to the priorities for radon reduction.

Despite their approximate nature, it is tempting to convert the values of total concentration to annual exposure. For an indoor occupancy factor of 0.8, they represent 0.14 and 0.07 WLM in a year for houses and flats respectively. In the UNSCEAR report, sketchy experimental data are used to suggest a value of 0.21 WLM for masonry buildings in general.

Verification

It is very hard to verify models of radon concentration in dwellings. The gas is transported so readily that it is almost impossible to quantify the inputs to a room. Concentrations are usually higher than expected due, no doubt, to undiscovered routes of entry. However, we have been able experimentally to test the models for flats that I have mentioned (2).

We had access to a new apartment block built of concrete and phosphogypsum. It was unoccupied, undecorated, and unfurnished, but structurally complete. The relevant properties of the building materials and structural elements were easily established. The specific activity of ^{226}Ra in the phosphogypsum was about 450 Bq kg^{-1}. The experiment was mounted high up in the

building in a room with 3 gypsum partitions 65 mm thick. (The three ordinary concrete elements were 150 mm thick.) The room was closed, and all other doors and windows in the apartment and in contiguous apartments were opened wide, so that the radon and daughter concentrations everywhere in the air surrounding and ventilating the room were virtually equal. We had, as it were, isolated the room from secret sources of radon. The ventilation rate stabilized at 0.86 h^{-1}.

The simple models that I have described led us to expect certain radon gas and daughter concentrations and gamma-ray dose rates. The degree to which the predictions matched the observations is evident from Table II; the outcome is reasonably encouraging, but the opposite signs of daughter and gas differences indicate that there may be appreciable plateout.

One point must be stressed. The values here are attributable solely to the structural elements of the room. The gross radon daughter concentration was actually 3.6 mWL, which is about twice the value that one would expect in an ordinary flat and roughly equal to the value for an ordinary house.

Waterborne radon

Although the emphasis in this session is on building materials, mention must be made of one, at least, of the other sources of radon in dwellings, the water supply. It is paradoxical that waterborne radon has more impact through inhalation than ingestion.

If the activity concentration in the supply is known, the radon daughter concentration indoors is readily modelled. For representative radon concentrations in surface and ground waters of 3.70×10^{2} and 3.70×10^{4} Bq m^{-3}, the daughter concentrations in indoor air would be about 0.005 and 0.5 mWL in typical circumstances. For the bulk of buildings, therefore, water is a second-order source, but under certain hydrological conditions, it may be the dominant contributor.

Summary

The crude estimates of exposure to natural radiation that have been made here are summarized in Table III.

It will be clear to the reader of the UNSCEAR report that the values in this table are not radically different from those in the UN document. The cosmic-ray entry is merely the UNSCEAR value rounded somewhat, but it has been possible to differentiate between exposures in single-family houses and in blocks of flats. The significant difference is that the

gamma-ray and radon daughter values here derive from models, whereas the UNSCEAR values derive from measurements. I suggest that the broad agreement argues for the utility of modelling.

In my opening proposition, I said that measurement and modelling were essential as well as useful. What do I mean by essential in this context? I have in mind : (i) the need to describe the prevailing distribution of exposures; (ii) the need to estimate the effects of changes in exposure conditions; (iii) the need to forecast the exposures in novel situations. General surveys do not fulfil all these requirements. Only modelling can be used for predictive purposes, and specific measurements are necessary to supply values of parameters in models, and to verify the predictions. Modelling needs to be far more elaborate, of course, than I have been illustrating.

I also proposed that we should meditate on indoor exposure. The prayerful association is deliberate, because it will be extremely difficult to decide how to apply the ICRP dose control system to natural radiation. It seems to me that the solution must stem from a definition of normal exposure that has a sociological as well as statistical dimension. We may then be able to apply controls in abnormal situations.

References

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TABLE I. RADON DAUGHTER SOURCES AND CONCENTRATIONS IN MASONRY DWELLINGS IN TYPICAL CIRCUMSTANCES

Sources	Houses [a]		Flats [b]	
	mWL	%	mWL	%
Structural elements	0.7	20	0.8	50
Ventilating air	0.8	25	0.8	50
Ground underneath	1.8	55	0	0
All sources	3.3	100	1.6	100

[a] Single-family houses and ground-floor flats

[b] Flats other than ground-floor

TABLE II. EXPECTED AND OBSERVED RADIATION LEVELS IN AN ISOLATED CONCRETE AND PHOSPHOGYPSUM ROOM DUE TO THE STRUCTURAL ELEMENTS

Quantity	Unit	Exp.	Obs.	Diff.
^{222}Rn daughter concentration	mWL	1.7	1.5	+13%
^{222}Rn gas concentration	Bq m^{-3}	11.1	13.0	-14%
Gamma-ray dose rate in air	μGy h^{-1}	0.08	0.064	+25%

TABLE III. CRUDE ANNUAL EXPOSURES TO NATURAL RADIATION INDOORS

Contributors	Houses[a]	Flats
Cosmic rays, mGy	0.3	0.3
Gamma rays, mGy	0.25	0.35
Radon daughters, WLM	0.14[b]	0.07[c]

[a] Single-family houses

[b] And ground-floor flats

[c] Other than ground-floor flats

DER GEHALT NATÜRLICHER RADIONUKLIDE UND DIE RADON-EXHALATIONSRATE VON BAUSTOFFEN UND MESSUNGEN DER GAMMASTRAHLENDOSISLEISTUNG IN WOHNUNGEN IN DER BUNDESREPUBLIK DEUTSCHLAND

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KURZFASSUNG. Es soll ein Überblick gegeben werden über die Ergebnisse der seit 1972 in der Bundesrepublik Deutschland laufenden Untersuchungen (gefördert vom Bundesminister des Innern) über die Strahlenexposition von aussen durch natürliche radioaktive Stoffe in Wohnungen unter Berücksichtigung des Einflusses von Baustoffen.
Über 500 Baustoffproben wurden auf ihren spezifischen Gehalt an Kalium-40, Radium-226 und Thorium-232 untersucht. Für verschiedene Baumaterialien (wie z.B. Bimsprodukte, Chemiegipse, Granit, Flugascheerzeugnisse usw.) wurden überdurchschnittlich hohe Werte gefunden. Der Zusammenhang zwischen der spezifischen Aktivität von Baustoffen und der Ortsdosisleistung in Räumen und der daraus folgenden Möglichkeit der Angabe von Grenzwerten wird an verschiedenen Berechnungsmodellen diskutiert. In einer Zusammenfassung werden dann die Ergebnisse von etwa 30.000 Einzelmessungen der Gammastrahlen-Dosisleistung in Wohnungen angegeben. Der Mittelwert in der Bundesrepublik Deutschland betrug 8,0 /uR/h, der Maximalwert 33,8 /uR/h. Ein Beitrag zur Strahlenexposition der Bewohner, den das aus den Baustoffen exhalierende Radon mit seinen Folgeprodukten liefert, wird zur Zeit noch untersucht. Über erste Ergebnisse dieser Messungen mit einer Abschätzung der zu erwartenden Lungenbelastung wird berichtet.

SUMMARY. MEASUREMENT OF THE RADIONUCLIDE CONTENT AND RADON EXHALATION RATES OF BUILDING MATERIALS AND MEASUREMENTS OF GAMMA RADIATION DOSES IN DWELLINGS IN THE FEDERAL REPUBLIC OF GERMANY. The aim is to review the research (supported by the Federal Minister of the Interior) carried out in the Federal Republic of Germany since 1972 into levels of exposure to external radiation from naturally radioactive substances in dwellings, with particular reference to the influence of building materials.
Over 500 samples of building materials have been analysed to determine their specific potassium-40, radium-226 and thorium-232 content, and above-average values have been recorded for various building materials (e.g. pumice products, by-product gypsum, granite, fly ash products).

The relationship between the specific radioactivity of building materials and dose rates within buildings, together with the consequent possibility of laying down maximum permissible values, are discussed in the light of several calculation models. This is followed by a brief review of results of approximately 30 000 individual measurements of gamma radiation dose rates inside dwellings. The average doserate for the Federal Republic of Germany was 8.0 /uR/h and the maximum 33.8 /uR/h. The additional exposure of occupants of dwellings attributable to radon and its daughter products in building materials is still being investigated. The initial results of these measurements are reported on and an estimate is also made of the lung exposure to be expected.

RESUME. TENEUR EN RADIONUCLEIDES NATURELS ET TAUX D'EMANATION DE RADON PAR LES MATERIAUX DE CONSTRUCTION; MESURE DU DEBIT DE DOSE DANS LES HABITATIONS EN REPUBLIQUE FEDERALE D'ALLEMAGNE. Le rapport donne un aperçu des résultats des études en cours en République fédérale d'Allemagne depuis 1972 (à l'instigation du ministère de l'intérieur) sur l'exposition au rayonnement externe dû aux matières radioactives naturelles dans les habitations compte tenu des matériaux de construction.
On a étudié la teneur spécifique en potassium-40, radium-226 et thorium-232 de plus de 500 échantillons de matériaux. Certains matériaux (produits à base de ponce, plâtres chimiques, granit, produits à base de cendre volante etc.) accusent des valeurs supérieures à la moyenne. A l'aide de divers modèles de calcul, on examine la corrélation entre l'activité spécifique des matériaux et le débit de dose d'ambiance à l'intérieur des locaux, et la possibilité d'en dériver des limites. Le rapport contient une synthèse des résultats de quelque 30 000 mesures de débit de dose gamma dans les logements. La valeur moyenne pour la République fédérale d'Allemagne était de 8,0 /uR/h, et le maximum de 33,8 /uR/h. Les contributions à l'exposition radiologique des occupants, du radon et de ses produits de filiation qu'émanent les matériaux de construction est encore à l'étude. Le rapport publie les premiers résultats de ces mesures, ainsi qu'une évaluation de la charge pulmonaire prévisible.

Im Rahmen der vom Bundesminister des Innern geförderten Forschungsvorhaben untersuchten mehrere wissenschaftliche Institutionen in den vergangenen sieben Jahren die Strahlenexposition von außen in der Bundesrepublik Deutschland durch natürliche radioaktive Stoffe im Freien und in Wohnungen unter Berücksichtigung des Einflusses von Baustoffen (1, 2). Eine Übersicht über die umfangreichen Untersuchungen soll im folgenden gegeben werden.

Dank der leichten Handhabung der eingesetzten Meßgeräte (Typ H 7201) wurden in den Jahren 1973 und 1974 etwa 30 000 Messungen der Gammastrahlen-Dosisleistung in Wohnungen durchgeführt. Aufgegliedert nach den einzelnen Bundesländern zeigt die Tabelle I die mittleren Dosisleistungswerte.

Tabelle I: Mittlere Dosisleistungswerte in Wohnungen und im Freien in den Bundesländern

Land	Dosisleistung in Wohnungen	im Freien	Differenz	Quotient
	$(\overline{\dot{D}_H})$	$(\overline{\dot{D}_F})$	$\overline{\dot{D}_H - \dot{D}_F}$	$\overline{\dot{D}_H / \dot{D}_F - 1}$
	µR/h	µR/h	µR/h	%
Baden-Württemberg	7,9	6,2	1,7	34
Bayern	8,5	6,9	1,6	31
Berlin (West)	7,0	5,8	1,3	26
Bremen	5,3	4,2	1,1	30
Hamburg	5,6	5,6	0,05	2
Hessen	9,0	6,0	3,0	55
Niedersachsen	6,5	4,8	1,6	39
Nordrhein-Westfalen	7,6	5,9	1,7	33
Rheinland-Pfalz	10,3	6,8	3,7	59
Saarland	12,1	7,9	4,2	62
Schleswig-Holstein	6,0	5,2	0,8	19
Bundesrepublik (Mittelwert sämtlicher Einzelmessungen)	8,0	6,0	1,8	36

Um eine bessere Übersicht zu erhalten, werden in den folgenden vier Abbildungen die nach verschiedenen Kriterien ausgewerteten Meßergebnisse von nur vier Bundesländern angegeben.

Abbildung I zeigt die Abhängigkeit der gemessenen Dosisleistung vom Baujahr der Wohnhäuser. Die mittlere Dosisleistung in Neubauten ist im allgemeinen etwas geringer als in älteren Häusern (durchschnittlich 5 - 6 mrem/a). Nur im Saarland ist in den nach 1900 gebauten Häusern die Dosisleistung um fast ein viertel höher als in den älteren Häusern, was auf die Verwendung von Schlackensteinen zu Beginn dieses Jahrhunderts zurückzuführen ist.

In der Abbildung II werden die Ergebnisse nach den einzelnen Gebäudearten unterteilt. Die Dosisleistung in Fertighäusern liegt bis auf Bayern in allen Bundesländern niedriger als im Freien. Da die Dosisleistung im Freien bei den Fertighäusern allgemein niedriger ist als bei den Massiv- bzw. Fachwerkhäusern, muß ein Einfluß der Bebauung auf die im Freien ermittelte Dosisleistung angenommen werden.

Die Umrechnung (3) dieser Dosisleistungen in die jährliche Keimdrüsendosis ergibt in Wohnungen für Bremen den Minimalwert von 33 mrem/a und den Maximalwert von 76 mrem/a für das Saarland. Der Zusatzbeitrag der Wohngebäude zur Umgebungsstrahlung im Freien beträgt hier 7 mrem/a für Bremen und 26 mrem/a für das Saarland. Eine auf die Bevölkerungszahl normierte Summenverteilung ergab folgende Aufgliederung:

- 80 Prozent der Bevölkerung ist einer terrestrischen Strahlung zwischen 4,3 µR/h und 10,5 µR/h in Wohnungen ausgesetzt
- je ein Prozent der Bevölkerung ist einer terrestrischen Strahlung von weniger als 2,7 µR/h und mehr als 16 µR/h in Wohnungen ausgesetzt
- der Variationsbereich der jährlichen Ortsdosis in Wohnungen in der Bundesrepublik für 98 Prozent der Bevölkerung beträgt 116 mR.

Der Mittelwert sämtlicher Einzelmessungen der Dosisleistung in Wohnräumen ergab 8,0 µR/h, die gewichteten Mittelwerte nach der Einwohnerzahl und nach der Häufigkeit des Baujahres betrugen 7,9 µR/h.

In der Abbildung III sind die prozentualen Abweichungen der Dosisleistung im Haus gegenüber der im Freien und in Abbildung IV die absolute Abweichung der entsprechenden Jahresdosis für die verschiedenen Haustypen in vier Bundesländern aufgetragen. Die abschirmende Wirkung der Fertighäuser tritt außer in Bayern in allen anderen Ländern (bis zu 10 %) deutlich hervor. Im Saarland beträgt die positive Abweichung bei den Massivhäusern 63 Prozent, was sicher zum Teil auf die Verwendung der bereits erwähnten Schlackensteine zurückzuführen ist. Ähnlichen Einfluß haben die Bimssteine, die häufig in Hessen und Rheinland-Pfalz beim Hausbau eingesetzt werden. In den beiden Ländern beträgt der Zusatzbeitrag bei Massivhäusern 59 Prozent.

Die Mittelwerte für die Bundesrepublik in den verschiedenen Häusergruppen sind in der Tabelle II zusammengestellt.

Tabelle II: Keimdrüsendosis durch die terrestrische Strahlung in der Bundesrepublik

	Mittelwerte in Wohnungen $\dot{D}_H$ mrem/a	Mittelwerte im Freien $\dot{D}_F$ mrem/a	Differenz $\dot{D}_H - \dot{D}_F$ mrem/a	Quotient $(\dot{D}_H/\dot{D}_F - 1)$ %
Gebäude-Baujahr				
bis 1900	52	41	10	33
1901 - 1948	53	38	14	43
ab 1949	47	36	10	33
Gebäudearten				
Massivhaus	50	37	11	37
Fachwerkhaus	51	39	11	34
Fertighaus	29	31	- 3	- 6
Holzhaus	32	31	0	2
Bundesrepublik (Mittelwert sämtlicher Einzelmessungen)	50	37	11	36

In der Tabelle III sind die Unterschiede der Meßergebnisse in Abhängigkeit vom jeweils verwendeten Hauptbaumaterial aufgetragen. Die Angaben dürfen nur als grobe Richtwerte angesehen werden, die regional stark schwanken können.

Tabelle III: Änderung der Strahlenexposition in Wohnungen zu der im Freien in Abhängigkeit vom verwendeten Baustoff

Baustoff	Unterschied
Bimsstein	+ 50 %
Schlackenstein	+ 47 %
Klinker	+ 36 %
Ziegel und Naturstein	+ 35 %
Lehm	+ 35 %
Kalksandstein	+ 24 %
Beton	+ 24 %
Blähbeton	+ 18 %
Holz	- 4 %

Eine Abschätzung des Einflusses der einzelnen Baustoffe auf die Höhe der Umgebungsstrahlung in Wohngebäuden wird in der Tabelle IV angegeben.

Tabelle IV: Einfluß der Baumaterialien auf die Höhe der Umgebungsstrahlung in Wohngebäuden

Baustoff	Radioaktivitäts-gehalt	zusätzliche Strahlenbelastung (mrem/a)
Naturgips Holz Kunststoff	sehr niedrig	0
Kalkstein Sandstein	niedrig	0 - 10
Ziegel Beton	mittel	10 - 20
Granit techn. Gips	höher	20 - 80
Schlackenstein Bimsstein	sehr hoch	80 - 170

In den Jahren 1972 bis 1976 wurde in vielen Einzelmessungen der Kalium-, Radium- und Thoriumgehalt von Baustoffen durch Gammaspektrometrie bestimmt. Weit über 500 verschiedene Stoffe, die beim Hausbau verwendet werden, wurden von sechs Institutionen untersucht (1, 2). Die Vergleichbarkeit der jeweiligen Ergebnisse wurde durch Kalibrierungsvergleiche sichergestellt. Die Tabelle V gibt die Mittelwerte und die zugehörigen Maximalwerte des spezifischen Gehaltes an Kalium, Radium und Thorium in verschiedenen Baustoffgruppen an.

Tabelle V: Mittelwerte (und Maximalwerte) der Konzentration natürlich radioaktiver Stoffe in Gruppen verschiedener Baumaterialien, die in der Bundesrepublik Deutschland verwendet werden

Baumaterialgruppe	Kalium (nCi/kg)	Thorium (nCi/kg)	Radium (nCi/kg)
Bausand, Baukies	7 (18)	0,4 (1,0)	0,4 (0,8)
Sandstein	5 (24)	0,5 (1,4)	0,5 (1,1)
sonstige Natursteine	13 (25)	0,8 (1,4)	0,7 (1,2)
Naturgips	2,4 (5)	0,5 (-)	0,7 (-)
andere Kunststeine	10 (20)	0,8 (2,5)	0,9 (2,6)
sonstige Zuschlagstoffe und Putzm.	6 (16)	0,4 (0,9)	0,6 (1,6)
Basalt	38 (55)	1,4 (2,6)	1,1 (2,3)
Zement	4 (7)	1,4 (5,2)	1,4 (5,3)
Schiefer, Granit	40 (96)	2,2 (5,2)	1,5 (3,6)
Wandkeramik	13 (31)	1,9 (4,9)	1,7 (2,7)
Ziegel, Klinker	17 (69)	2,6 (10,0)	2,2 (6,7)
Bimssteine	24 (30)	2,3 (4,6)	2,2 (3,6)
Schlackensand und Schlackensteine	9 (16)	2,8 (5,6)	2,2 (3,2)
techn. erz. Gips	2 (6)	0,5 (-)	14,0 (28,0)

Bemerkenswert ist der hohe Radiumgehalt für technisch erzeugten Gips und der relativ hohe Anteil von Thorium und Radium in Ziegel-, Bims- und Schlackensteinen.

Die Relation zwischen der spezifischen Aktivität der Baustoffe und der durch sie hervorgerufenen Ortsdosisleistung in Wohnungen ist kompliziert. Sie ist u.a. abhängig von verschiedenen geometrischen Faktoren wie Wanddicke, Größe der Wohnräume, Größe der Fenster und Türen, außerdem von der Vielzahl der in den Räumen verwendeten Baustoffe und zusätzlich von Größen wie der Abschirmwirkung der Gebäude gegenüber der terrestrischen Strahlung bzw. der Dosisaufbaufaktoren der einzelnen Materialien.

Ein konservatives Näherungsverfahren wurde vom Leningrader Forschungsinstitut für Strahlenhygiene (4) erstellt. Anstelle eines realen Hauses wurde ein Hohlraum in einem unendlich dicken Baumaterial angenommen. Dieses Modell liefert unter der Annahme einer zusätzlichen Strahlenexposition von maximal 150 mrad/a folgende maximal zulässigen Konzentrationen der Radionuklide in dem jeweiligen Baustoff.

Radionuklid	maximale Konzentration (MK) in nCi/kg
K-40	130
Ra-226	10
Th-232	7

Falls alle drei Radionuklide in dem Baustoff vorhanden sind, gilt

$$\frac{C_K}{MK_K} + \frac{C_{Ra}}{MK_{Ra}} + \frac{C_{Th}}{MK_{Th}} \leq 1$$

wobei C die jeweilige spezifische Aktivität in nCi/kg angibt.

Um auch im Einzelfall eine Berechnung der Dosisleistung in Wohnungen durchzuführen, wurde von E. Oberhausen und G. Keller ein Plattenmodell vorgeschlagen (1, 5). Es wird eine Kreisscheibe endlicher Dicke betrachtet und die Ortsdosisleistung in einem Punkt im Abstand z berechnet. Dann gilt:

$$J_{L,RN}=I_{\gamma,RN}\cdot a\cdot \rho_b\cdot 2\pi\sum_{j=1}^{n} ß_j\cdot B_{j,b}\cdot\int_{y=0}^{D}[E_i(-\mu_j\cdot y\sqrt{1+\frac{R^2}{(z+y)2}})-E_i(\mu_j\cdot y)]\cdot dy$$

Es bedeutet:

$J_{L,RN}$: Dosisleistung des Radionuklides

$I_{\gamma,RN}$: Gammastrahlenkonstante des Radionuklides

a : Spezifische Aktivität des Radionuklides im Baustoff

ρ_b : Dichte des Baustoffes

D : Dicke des Baustoffes

E_i : Exponentialintegrale

μ_j : Schwächungskoeffizient in dem Baustoff für die Energie j

R : Radius der betrachteten Kreisscheibe

z : Abstand des Meßpunktes von der Kreisscheibe

$B_{j,b}$: Dosisaufbaufaktor für die Energie j, den Baustoff b, die Dicke D (dose buildup factor)

$ß_j$: Bewertungsfaktor

Die energieabhängigen Dosisaufbaufaktoren, die in die Formel eingehen, sind für realistische Wanddicken keineswegs vernachlässigbar, ihre Berechnung jedoch recht kompliziert (5). Messungen der Dosisleistung in einem Modellraum aus Chemiegipswänden ergaben eine sehr gute Übereinstimmung mit den berechneten Werten nach dem Plattenmodell.

Der Beitrag zur Strahlenexposition in Wohnräumen durch das aus dem Baumaterial emanierende Edelgas Radon wird z.Z. in der Bundesrepublik noch untersucht. Die Radon-Exhalationsrate von einigen nicht vorbehandelten Baustoffen ist in Tabelle VI angegeben (6).

Tabelle VI: ^{222}Rn-Exhalationsrate aus Baustoffen

Baustoff	^{226}Ra-Konzentration (nCi/kg)	^{222}Rn-Exhalationsrate (pCi/m^2·h)
Kalksandstein	0,3	40
Bimsstein	1,8	83
Roter Klinker	1,0	7
Hüttenstein	1,9	58
Leichtbeton	0,2	13
Chemiegips	7,0	650

Durch Tapezieren, Streichen, Verputzen und Isolieren der Wände und Decken wird die Radon-Exhalationsrate der Baustoffe stark verändert. Zusätzlich kann sie noch durch die Radon-Konzentration in der Außenluft und durch die Zulieferung des Radons aus dem Erdreich beeinflußt werden. Entscheidend für die Lungenexposition ist die Konzentration des Radons und seiner Folgeprodukte in der Innenluft der Räume. Diese wird außer von der Exhalationsrate der Baustoffe noch von der Ventilationsrate bestimmt. In der Bundesrepublik Deutschland wird seit Anfang 1979 im Rahmen eines Forschungsvorhabens die Konzentration des Radons und seiner Folgeprodukte in Wohnräumen untersucht. Ein erstes Ergebnis aus 100 Messungen zeigt die Abbildung V (7). Hierbei wurde aus den gemessenen spezifischen Aktivitätskonzentrationen der kurzlebigen Radonfolgeprodukte die mittlere Inhalationsdosis für die Lunge nach UNSCEAR'77 berechnet. Der Mittelwert der Inhalationsrate dieser Messungen ergab $0{,}61 \pm 0{,}35$ rem/a.

Wie die Ergebnisse dieser Untersuchungen zeigen, ist der zusätzliche Beitrag zur Strahlenexposition der Bevölkerung durch die Verwendung von Baumaterialien mit relativ hohem Gehalt an natürlichen radioaktiven Stoffen sowie die Konzentration des Radons und seiner Folgeprodukte in Wohnungen keineswegs vernachlässigbar. Gerade Maßnahmen zur besseren Wärmedämmung wie z.B. extrem dichte Fenster und Türen oder die Verwendung von besser isolierenden Steinen müssen auch unter dem Aspekt der eventuellen Erhöhung der Strahlenexposition der Bevölkerung gesehen werden.

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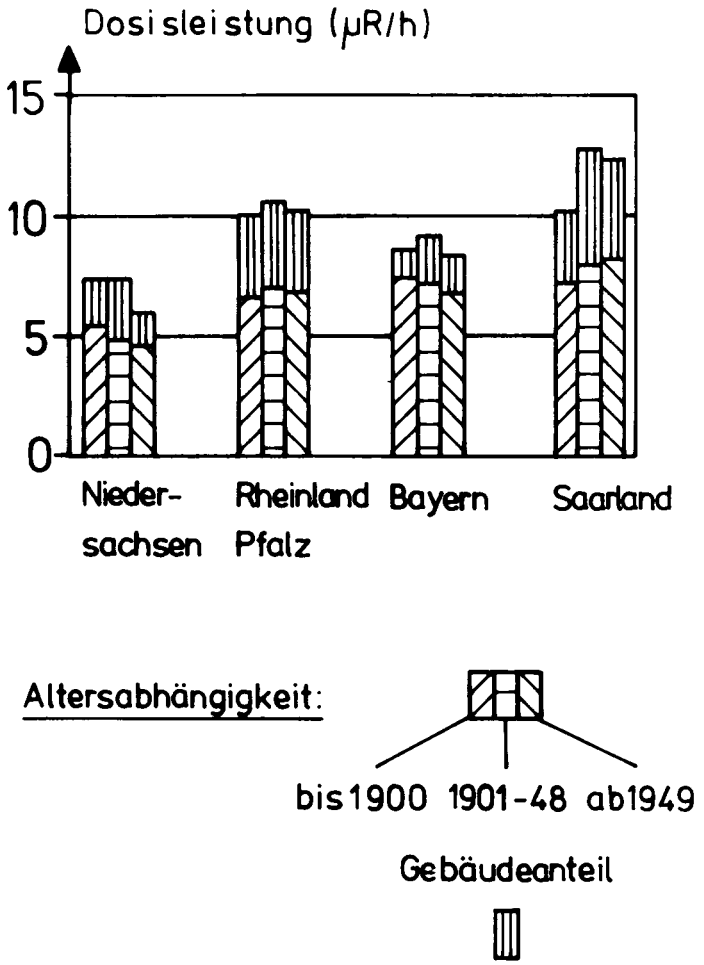

Abbildung I: Terrestrische Strahlung in Wohnungen in Abhängigkeit vom Alter der Gebäude

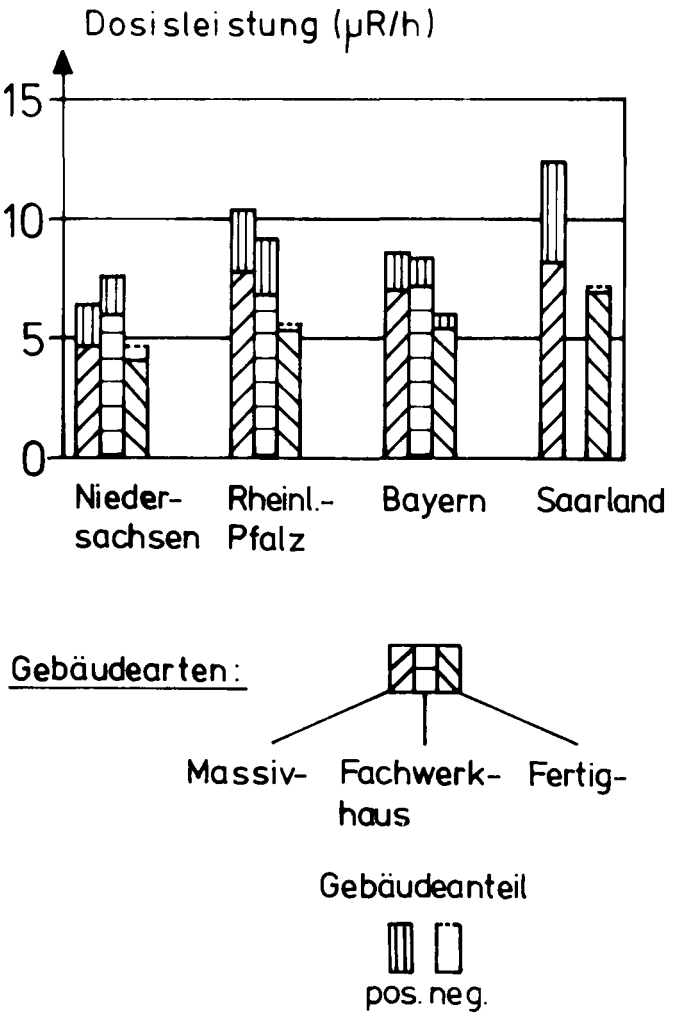

Abbildung II: Terrestrische Strahlung in Wohnungen in Abhängigkeit von der Gebäudeart

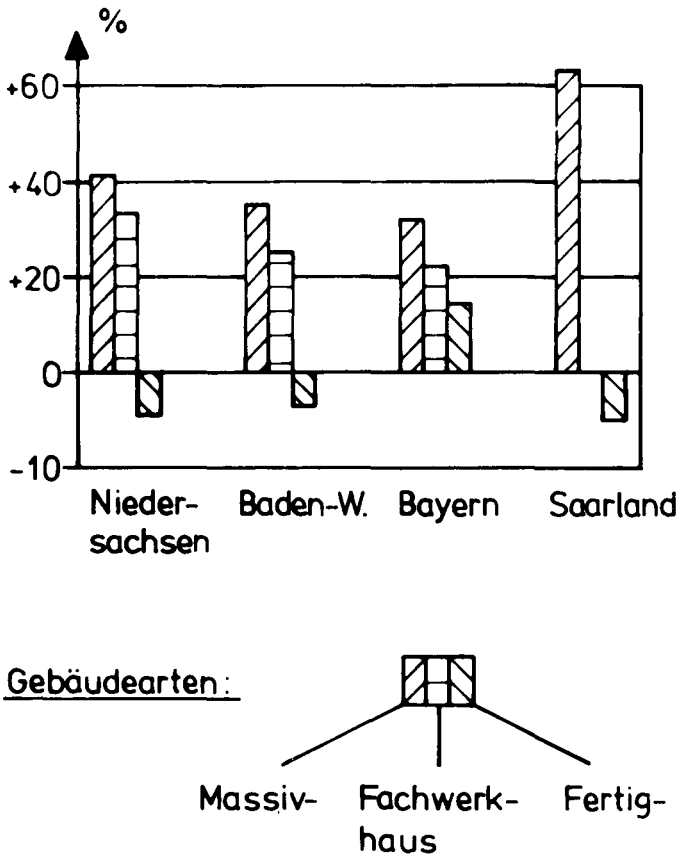

Abbildung III: Prozentuale Abweichung der Dosisleistung im Haus gegenüber der im Freien für verschiedene Gebäudearten (ohne kosmische Strahlung)

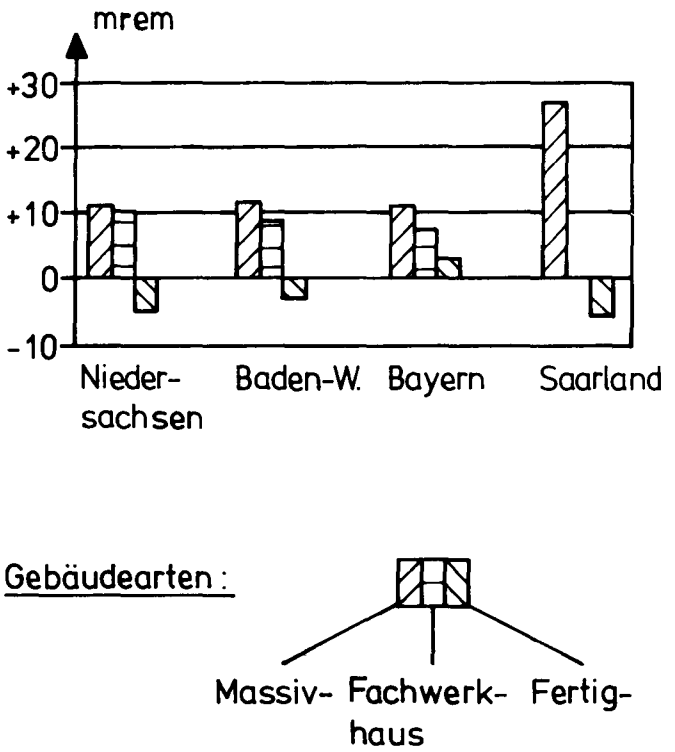

Abbildung IV: Abweichung der Jahresdosis im Haus von der im Freien für verschiedene Gebäudearten in mrem (ohne kosmische Strahlung)

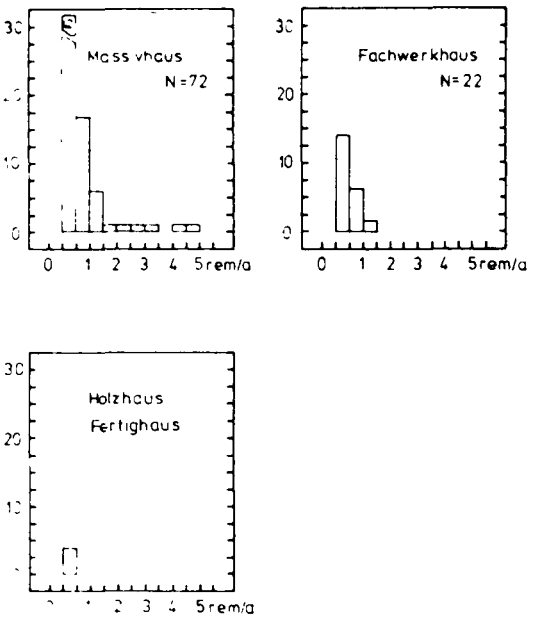

Abbildung V: Häufigkeitsverteilung der Inhalationsdosis in Wohnräumen verschiedener Gebäudearten

MESURE DE L'IRRADIATION EXTERNE A L'INTERIEUR DES HABITATIONS: PRESENTATION ET DISCUSSION DES RESULTATS OBTENUS EN FRANCE

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RESUME. Le programme de mesure de l'irradiation naturelle que le CEA a entrepris comprend la mise en place de dosimètres à l'intérieur des habitations en nombre sensiblement égal à ceux disposés à l'extérieur. Chacun de ces dosimètres est associé à un questionnaire demandant divers renseignements sur l'habitation dans laquelle il est placé. Les résultats obtenus, portant sur 5 départements et un millier de dosimètres, seront présentés et discutés en fonction des paramètres les plus importants (nature des matériaux de construction, âge et type d'habitation, etc...)

KURZFASSUNG. MESSUNGEN DER ÄUSSEREN STRAHLUNG IN WOHNUNGEN: ERGEBNISSE IN FRANKREICH. Im Rahmen des vom CEA durchgeführten Messprogramms zur Ermittlung der natürlichen Strahlung wurde in den Wohnungen nahezu die gleiche Anzahl Dosimeter aufgestellt wie im Freien. Jedem Dosimeter war ein Vordruck mit verschiedenen Fragen zu den Wohnungen beigegeben. Die in fünf Departements mit etwa 1 000 Dosimetern erhaltenen Ergebnisse werden als Funktion der wichtigsten Parameter (Art der Baustoffe, Baujahr, Wohnungstyp usw.) dargestellt und erörtert.

SUMMARY. INDOOR MEASUREMENTS A DISCUSSION OF THE RESULTS OBTAINED IN FRANCE. The programme of measurement of natural irradiation undertaken by the CEA includes the setting-up of dosimeters within dwellings in about the same number as those installed outdoors. Each of those dosimeters is associated with a form requesting information on the dwelling in question. The results obtained from about a thousand dwellings located in five départements will be presented and discussed as a function of the more significant parameters (nature of building materials, age and type of dwelling, etc.).

1 - INTRODUCTION

La connaissance des doses d'irradiation d'origine naturelle auxquelles sont soumises les populations, ainsi que leurs variations éventuelles d'une région à une autre, présente un intérêt scientifique certain et permet de juger de l'importance relative des diverses sources d'exposition.

En Europe, un certain nombre de pays, tels l'Allemagne [1], l'Autriche [2] et l'Italie [3] ont récemment procédé à des campagnes de mesure importantes.

En France, jusqu'à ces dernières années, seules des mesures très locales avaient été effectuées ; c'est pourquoi le Département de Protection (DPr) du Commissariat à l'Energie Atomique a entrepris, dès 1977, en collaboration avec le Service Central de Protection contre les Rayonnements Ionisants (SCPRI), une étude systématique sur l'ensemble du territoire national.

2 - METHODE ET MOYENS

Les dosimètres utilisés pour cette étude sont de type radiothermoluminescent au sulfate de calcium activé au dysprosium ; ils ont été fournis et développés par les laboratoires d'Instrumentation et de Dosimétrie du Service Technique d'Equipements de Protection et de Dosimétrie (DPr). La durée d'exposition de ces dosimètres est d'environ six mois.

Dans chacun des départements étudiés, 200 à 250 points de mesure sont répartis sur toute la surface du département en tenant compte de la distribution de la population. Chaque point de mesure comprend un dosimètre placé à l'intérieur d'une habitation et un dosimètre placé à l'extérieur. Chaque dosimètre est accompagné d'un questionnaire pour l'obtention de divers renseignements sur le lieu d'implantation, son environnement et les matériaux de construction des habitations (voir annexe).

Etant donné l'importance du nombre de mesures à effectuer dans chaque département, l'implantation des dosimètres ne pouvait se

faire sans avoir recours à un organisme représenté sur le plan local. La Direction Nationale de la Sécurité Civile a été sollicitée et par l'intermédiaire de ses Directeurs Départementaux nous a introduit auprès des autorités préfectorales.

Dès les premières prises de contact avec les préfectures intéressées, il est apparu que l'organisation, à la préfecture même, d'une réunion à laquelle assisteraient les responsables des différents services départementaux susceptibles d'apporter leur concours et au cours de laquelle seraient exposées les raisons de ces mesures, ne pouvait qu'en faciliter le déroulement et aider à vaincre les éventuelles réticences auprès du public.

Dans chaque département 200 à 250 personnes bénévoles acceptant "d'héberger" des dosimètres ont pu être trouvées. La majeure partie de ces bénévoles a été fournie par les Services d'Incendie et de Secours, le reste étant pris parmi d'autres organismes ou service locaux : Action Sanitaire et Sociale, Gendarmerie, Equipement, Croix-Rouge, etc...

Les dosimètres ont été expédiés par service postal puis par transporteur vers chaque département, la mise en place s'effectuant par les Services d'Incendie et de Secours. La récupération s'est déroulée de façon identique après une exposition de six mois.

La figure n° 1 montre les départements pour lesquels les résultats sont disponibles, ceux pour lesquels les mesures sont en cours et ceux pour lesquels les contacts avec les autorités préfectorales ont été pris ou seront pris avant la fin de l'année 1979.

Les résultats obtenus dans la région parisienne ont déjà fait l'objet de publications [4], [5] et seront rappelés ici pour mémoire.

III - RESULTATS OBTENUS

Les doses minimales, moyennes et maximales pour les départements étudiés sont indiquées dans le Tableau I ainsi que le nombre de mesures obtenues, compte tenu des dosimètres ou des questionnaires perdus (ou mal remplis). Dans les résultats indiqués les doses dues au rayonnement cosmique, supposées égales à 30 millirads par an, ont été déduites.

Les distributions des doses sont représentées par département dans la figure 2. Le rapport entre la dose moyenne absorbée à l'intérieur et celle absorbée à l'extérieur des habitations, pour chaque département ou région, est indiqué dans le Tableau II ; ce rapport est au moins égal à 1 et varie de 1,00 à 1,20, la valeur moyenne de ces rapports étant de 1,11.

IV - ANALYSE DES RESULTATS

Les résultats ont été analysés sous deux angles :

- variabilité des doses dans les habitations d'un département donné, ou d'un ensemble de départements,
- recherche des paramètres qui ont le plus d'influence sur les doses.

IV.1 - VARIABILITÉ DES DOSES

Le Tableau I montre qu'il existe des différences nettes entre les régions, l'irradiation externe d'origine naturelle dans les habitations des départements du Limousin (Creuse, Corrèze et Haute-Vienne) étant nettement plus élevée que dans l'Yonne, les Deux-Sèvres et la Région Parisienne.

Les histogrammes obtenus pour les départements de la Corrèze, la Creuse, les Deux-Sèvres et la Haute-Vienne sont de type plutôt symétrique, le dernier présentant à la fois des valeurs très basses et des valeurs très élevées. L'histogramme de l'Yonne présente quant à lui une dissymétrie assez nette.

Les distributions auxquelles nous avons tenté d'ajuster les histogrammes observés sont gaussiennes ou log-gaussiennes. On a utilisé pour cela des tests spécifiques de normalité (Geary, David, Agostino) en conjonction avec les tests d'ajustement classiques (Chi-2, Kolmogorov-Smirnov). Pour la Corrèze, la Creuse et les Deux-Sèvres, la distribution gaussienne est une bonne hypothèse de travail. Pour la Haute-Vienne, les hypothèses gaussienne et log-gaussienne sont rejetées ; pour obtenir une loi compatible avec les données il faudrait s'orienter vers des distributions dont les parties extrêmes ont des probabilités plus élevées que dans la loi de Laplace-Gauss. Dans le cas du département de l'Yonne, une hypothèse de log normalité est satisfaisante.

IV.2 - INFLUENCE DE DIVERS PARAMÈTRES SUR LA DOSE

L'influence sur la dose des principaux paramètres figurant dans le questionnaire donné en Annexe a été étudiée au moyen de méthodes statistiques. Ces principaux paramètres sont, d'une part, ceux qui caractérisent l'habitation :

- Nature du gros-oeuvre (bois, briques, béton, parpaings, granit, meulière, pierre de taille, pierre de pays).
- Support du plancher (dalle de béton, hourdis, planches sur bois).
- Revêtement du sol (carrelage, dallage, moquette, parquet).
- Nature de la cloison (béton, bois, brique, plâtre).
- Période de construction (avant 1900, de 1900 à 1945, depuis 1945).

et, d'autre part, ceux qui caractérisent l'emplacement du dosimètre dans l'habitation :

- Etage (sous-sol, rez-de-chaussée, étage supérieur).
- Position du meuble contenant le dosimètre (contre une

cloison, contre un mur de gros-oeuvre).

Dans chaque département et pour chaque paramètre, une analyse de variance à un facteur a été effectuée afin de juger s'il existe ou non des différences entre les valeurs moyennes de dose pour les diverses modalités du paramètre. Les résultats globaux de cette analyse (différence significative à 5 %) sont présentés au Tableau III. Pour tous les départements, la nature du gros-oeuvre a un effet net sur la dose tandis que l'étage n'en a pas. L'influence des autres paramètres est moins tranchée car elle est observée dans certains départements et pas dans d'autres.

IV.2.1 - Influence du gros-oeuvre

Les doses moyennes obtenues, par département et par type de matériau utilisé pour le gros-oeuvre, sont présentées au Tableau IV. Pour tous les départements, des doses relativement élevées ont été mesurées dans les habitations en granit tandis que l'influence des autres matériaux paraît être variable selon le département. Le test de STUDENT montre qu'il existe des différences significatives à 1 % dans la majorité des cas (Tableau V).

On peut en conclure que la dose d'irradiation externe à l'intérieur des habitations est essentiellement déterminée par le matériau de base utilisé pour la construction.

Les distributions des doses dues aux divers matériaux principalement utilisés dans le Limousin apparaissent dans la figure 3 ; les doses comprises entre 64 et 169 mrad par an sont réparties en 8 classes d'amplitude égale, les doses extérieures à ces limites formant les 2 classes extérieures.

IV.2.2 - Influence des autres paramètres

Il a déjà été indiqué que l'étage où est placé le dosimètre ne semble pas avoir d'influence sur la dose.

Pour les autres paramètres, des résultats contradictoires ont été obtenus pour les divers départements étudiés ; ils peuvent s'expliquer par le fait que les liaisons entre ces paramètres et la nature du gros-oeuvre sont plus ou moins fortes selon le département. Dans le Limousin, par exemple, le dallage semble être associé aux habitations anciennes construites en pierre du pays mais cela ne paraît pas être le cas dans l'Yonne.

CONCLUSION

Le Département de Protection du Commissariat à l'Energie Atomique a entrepris, en collaboration avec le Service Central de Protection contre les Rayonnements Ionisants, une étude systématique de l'irradiation externe d'origine naturelle sur l'ensemble du territoire national. Au 1er Août 1979, les mesures relatives à 5 départements ont été analysées. Pour ces départements, les doses absorbées à l'intérieur des habitations varient de 14 à 287 $mrad.an^{-1}$. Les moyennes obtenues pour chaque département varient de 43 à 120 $mrad.an^{-1}$. Une étude statistique fait ressortir que dans tous les départements la nature des matériaux de construction utilisés dans le gros-oeuvre a une influence sur les résultats obtenus. L'étude de l'influence des autres paramètres (âge, revêtement de sol, etc...) donne des résultats contradictoires suivant les départements.

La présentation de l'ensemble des résultats, portant sur les mesures effectuées à l'extérieur des habitations aussi bien qu'à l'intérieur de celles-ci, fait l'objet d'une publication séparée [6].

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		Nombre de dosimètres exploitables	Dose en mRAD.AN^{-1}			
			Minimale	Moyenne	Maximale	Ecart type
Région Parisienne		98	14	43	134	
Départements	Corrèze	107	48	114	189	34
	Creuse	186	68	120	208	26
	Deux-Sèvres	184	20	63	147	25
	Hte Vienne	202	25	116	287	33
	Yonne	243	21	61	151	23

TABLEAU I

DOSES ABSORBEES A L'INTERIEUR DES HABITATIONS
(RAYONNEMENT COSMIQUE 30 mRAD.AN^{-1} DÉDUIT)

		Intérieur / Extérieur
Région Parisienne		1,00
Départements	Corrèze	1,07
	Creuse	1,10
	Deux-Sèvres	1,17
	Hte Vienne	1,10
	Yonne	1,20

TABLEAU II

RAPPORTS ENTRE LES DOSES ABSORBÉES MOYENNES A L'INTÉRIEUR ET A L'EXTÉRIEUR DES HABITATIONS POUR LES DÉPARTEMENTS ET RÉGIONS ÉTUDIÉS

	Revêtement du sol	Support du plancher	Cloison	Année	Emplacement du meuble	Etage	Gros-Oeuvre
Corrèze	Non	Non	Non	Non	Non	Non	Oui
Creuse	Non	Oui	Non	Non	Non	Non	Oui
Deux-Sèvres	Oui	Oui	Oui	Non	Non	Non	Oui
Hte Vienne	Oui	Non	Non	Oui	Non	Non	Oui
Yonne	Non	Non	Non	Non	Non	Non	Oui
Limousin	Non	Oui	Oui	Oui	Non	Non	Oui

TABLEAU III

Effet des divers paramètres sur la dose
Oui : différences significatives à 5 %
Non : différences non significatives à 5 %

Nature du gros oeuvre	Corrèze		Creuse		Deux-Sèvres		Haute-Vienne		Yonne	
	Nombre de dosimètres	Dose moyenne	Nombre de dosimètres	Dose moyenne	Nombre de dosimètres	Dose moyenne	Nombre de dosimètres	Dose moyenne	Nombre de dosimètres	Dose moyenne
Bois	0	-	0	-	0	-	3	79	1	53
Briques	9	110	5	145	18	63	14	101	25	70
Béton	3	94	3	106	9	64	14	110	13	53
Parpaings	15	108	15	102	68	66	40	105	29	64
Granit	17	143	28	122	13	91	31	126	6	129
Meulière	0	-	7	111	5	41	1	129	5	52
Pierre de taille	4	107	11	122	7	41	4	112	66	57
Pierre de pays	37	115	61	118	57	56	62	129	67	56
Inconnue	1	65	0	-	2	69	2	133	3	66
Non réponse	2	82	0	-	0	-	6	128	9	58
Plusieurs réponses	19	102	56	125	5	72	25	104	19	71
Dose moyenne générale		114		120		63		116		61

TABLEAU IV

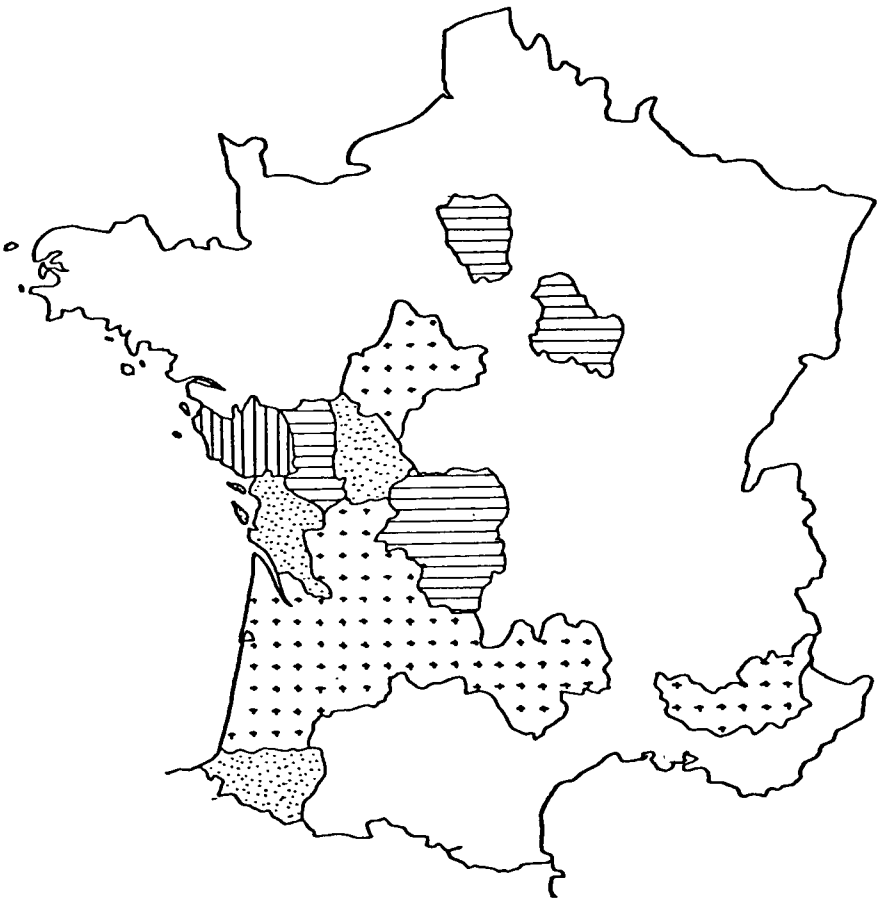
Moyennes des doses absorbées à l'intérieur des habitations en fonction de la nature des matériaux de construction utilisés pour le gros-oeuvre ($mRAD.AN^{-1}$)

Corrèze	Granit 143 ≠ Pierre du pays 115 ≠ Béton 106		Briques [9] 110 Meulière [4] 107
Creuse	Meulière 102 ≠ Granit 122 ≠ Béton 118 ≠ Pierre du pays 118		Briques [5] 145
Deux-Sèvres	Granit 91 ≠ Béton 66 ≠ Pierre du pays 56 ≠ Briques 63 ≠ Meulière 41	Meulière 41 ≠ Béton 66 ≠ Pierre du pays 56 ≠ Briques 63 ≠ Granit 91	
Haute-Vienne	Granit 126 ≠ Béton 106 ≠ Briques 101	Pierre du pays 129 ≠ Béton 106 ≠ Briques 101	Meulière [5] 115 Bois [3] 79
Yonne	Briques 70 ≠ Meulière 56 ≠ Pierre du pays 56 ≠ Béton 60		Granit [6] 129 Bois [1] 53
Limousin	Granit 129 ≠ Meulière 116 ≠ Pierre du pays 121 ≠ Briques 112 ≠ Béton 105	Béton 105 ≠ Pierre du pays 121	Bois [3] 79

TABLEAU V

Différences significatives de dose pour divers matériaux de construction utilisés dans le gros-oeuvre (seuil à 1 %)

FIGURE 1 - PROGRAMME D'ETUDE DE L'IRRADIATION EXTERNE DU CEA (SITUATION AU 1/8/1979)



DÉPARTEMENTS ÉTUDIÉS

DÉPARTEMENTS POUR LESQUELS LES DOSIMÈTRES SONT EN COURS DE DÉVELOPPEMENT

DÉPARTEMENTS POUR LESQUELS LES DOSIMÈTRES SONT EN COURS D'EXPOSITION

DÉPARTEMENTS POUR LESQUELS DES CONTACTS ONT ÉTÉ PRIS OU SERONT PRIS AVANT LA FIN DE 1979

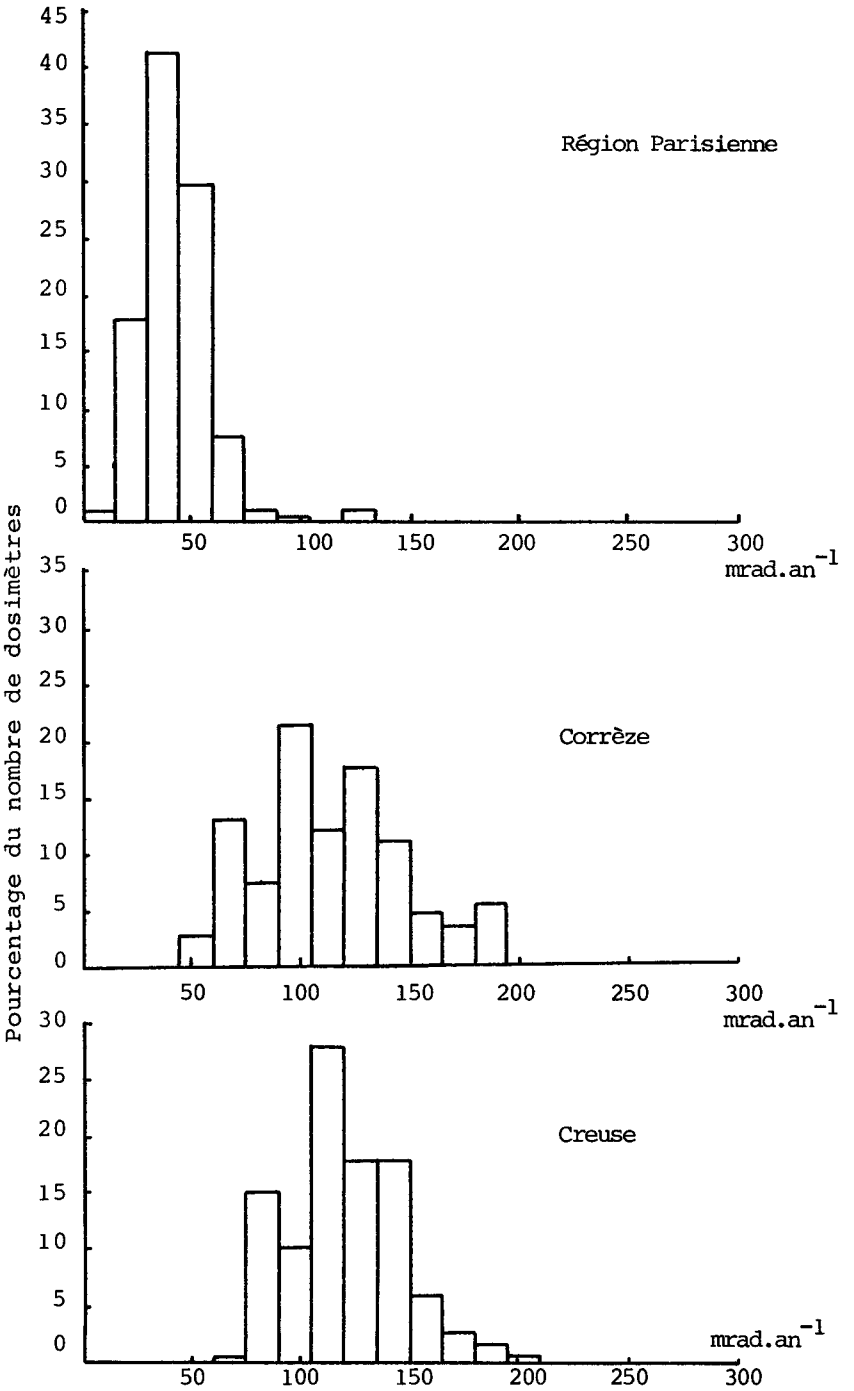


Figure 2 - Distribution des doses absorbées à l'intérieur des habitations (rayonnement cosmique exclus)

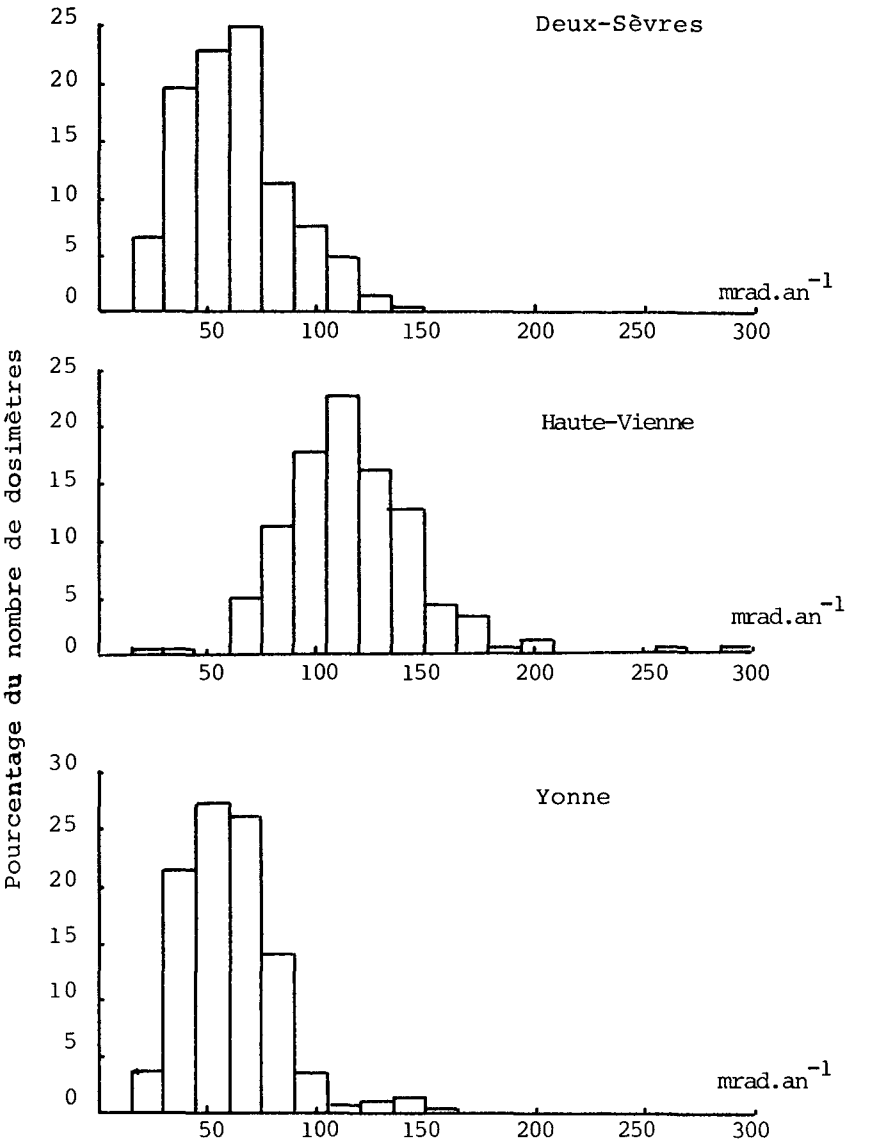


Figure 2 (suite) - Distribution des doses absorbées à l'intérieur des habitations (rayonnement cosmique exclus)

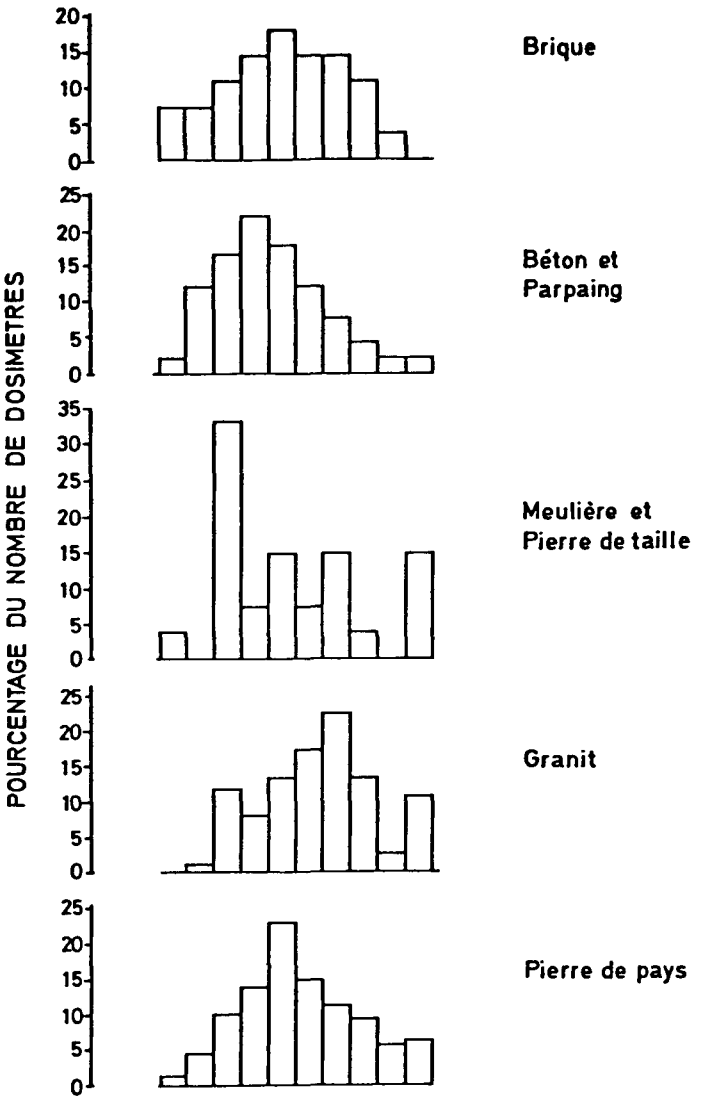


FIGURE 3 - DISTRIBUTION DES DOSES ABSORBÉES À L'INTÉRIEUR DES HABITATIONS DU LIMOUSIN SUIVANT LA NATURE DES MATÉRIAUX DE CONSTRUCTION UTILISÉS POUR LE GROS-OEUVRE

ANNEXE

MINISTERE DE L'INTERIEUR (Direction de la Sécurité Civile)
MINISTERE DE LA SANTE
MINISTERE DE L'INDUSTRIE ET DE LA RECHERCHE

QUESTIONNAIRE RELATIF AU DOSIMETRE PLACE A L'INTERIEUR DES HABITATIONS

Remplir le questionnaire en écrivant en lettres capitales et en mettant une croix dans les cases correspondant aux réponses.

Ne rien écrire dans cette colonne

Numéro du dosimètre : .....

	Jour	Mois	Année
Date de mise en place			
Date de relevé			

ND (1 2 3 4)
COM (6 10)
CX (12 17 21)
CY (23 28 32)
DS (34 38)
FS-EV (40 44)
MEP (46 50)
REL (52 56)
AR-DS (58 62)
DEV (64 68)

o NOM de la personne chez laquelle est placé le dosimètre

M., Mme, Melle : .....
.....

o Localisation géographique

Commune : .....
Lieu dit : .....
(éventuellement)
N° : ..... Rue : .....
Code postal : .....

EMPLACEMENT DU DOSIMETRE

I. Le dosimètre doit être placé dans une salle de séjour, dans un meuble en bois. Avez vous pu le placer :

- dans une salle de séjour *oui* [1] *non* [2] — SDS (70)
- dans un meuble en bois *oui* [1] *non* [2] — MEB (72)

Si vous avez répondu non à la dernière question, préciser la nature du support du dosimètre :
..... — AMEB (74)

II. Le meuble (ou support) où est placé le dosimètre est-il :

- contre un mur de gros-oeuvre [1]
- contre une cloison [2]
- dans une autre position [3]

PLS (76 77)

III. Indiquer l'étage où est placé le dosimètre :

- sous-sol [1]
- rez-de-chaussée [2]
- en étage [3]

lequel : .....

ND (1 4)
ET (6)
NUET (8 9)

T.S.V.P.

ANNEXE (SUITE)

Numéro du dosimètre (rappel) : .....

CONSTRUCTION (Répondre aux rubriques V ou VI selon l'appartenance de l'habitation à un type individuel "usage d'une famille" ou à un type collectif "plusieurs appartements dans un immeuble").

IV. Habitation individuelle oui [1]

Construite :

- sur vide sanitaire [1]
- sur sous sol ou cave [2]
- sur dalle [3]

Est-ce que sa construction est de type industriel ?

oui [1] *non* [2]

V. Votre habitation est un immeuble collectif : oui [2]

Précisez le nombre d'étages : .....

VI. Année de construction de votre habitation : .....

Si vous ne savez pas exactement, pouvez vous préciser si c'est :

- avant 1900 [1]
- de 1900 à 1945 [2]
- après 1945 [3]
- date inconnue [4]

MATERIAUX (Cette rubrique concerne la pièce où se trouve le dosimètre)

VII. Gros-oeuvre (murs)

- bois [1] - granit [5]
- briques [2] - meulière [6]
- béton [3] - pierre de taille (calcaire) [7]
- parpaings [4] - pierre du pays * [8]
- autre matériau : .....
- constitution inconnue [9]

VIII. Cloison

- bois [1] - plâtre [3] - béton [5]
- briques [2] - parpaings [4]
- autre matériau : .....
- constitution inconnue [6]

IX. Plancher

. Support :

- hourdis [1] - planches sur partie de bois [3]
- dalle de béton [2] - autre .....

. Revêtement :

- parquet [1] - moquette [4]
- carrelage [2] - plastique [5]
- dallage * [3]

autre matériau : .....

* Si vous pouvez préciser la nature de la pierre (schiste, grès, etc...) indiquer la à la rubrique autre matériau.

Ne rien écrire dans cette colonne

SSPAV [ ] 13

TYCO [ ] 11 PAVIND [ ] 15

NBET [ ][ ] 17 18

DCONS [ ][ ][ ] 20 22

GOEV [ ][ ][ ][ ][ ] 24 28

AMG [ ] 29

CLOIS [ ][ ][ ][ ][ ] 31 35

AMC [ ] 36

PLASU [ ][ ] 38 39

PLARE [ ][ ] 41 42

IRESB [ ][ ][ ][ ][ ][ ] 44 49

TENEURS EN RADIONUCLIDES NATURELS DES PHOSPHATES BRUTS, D'ENGRAIS PHOSPHATES ET DE MATERIAUX DE CONSTRUCTION UTILISES EN BELGIQUE

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RESUME. Un échantillonnage sélectif des phosphates bruts traités en Belgique a été effectué et les teneurs en radionuclides naturels ont été déterminées par mesure spectrométrique; des variations importantes de ces teneurs sont observées selon l'origine du minerai.
Un inventaire des principaux matériaux de construction à base de gypse, utilisés en Belgique, a été réalisé.
Les résultats des mesures de la radioactivité de ces matériaux sont présentés et discutés.

SUMMARY. NATURAL RADIONUCLIDE CONTENT OF RAW PHOSPHATES, PHOSPHATIC FERTILIZERS AND CONSTRUCTION MATERIALS USED IN BELGIUM. A selective sampling of raw phosphates processed in Belgium has been carried out and their natural radionuclide content has been measured by spectrometry. Depending on the origin of the ore, significant variation in the content has been detected.
An inventory of the main building materials containing gypsum and used in Belgium is presented and discussed.

KURZFASSUNG. GEHALT AN NATÜRLICHEN RADIONUKLIDEN DER IN BELGIEN VERWENDETEN ROHPHOSPHATE, PHOSPHATDÜNGEMITTEL UND BAUSTOFFE. Von den in Belgien verarbeiteten Rohphosphaten wurden selektive Proben genommen und durch spektrometrische Messung auf ihren Gehalt an natürlichen Radionukliden untersucht, der je nach der Herkunft des Erzes erhebliche Schwankungen aufweist.
Eine Zusammenstellung über die wichtigsten in Belgien verwendeten Baustoffe auf der Grundlage von Gips wird gebracht.
Die Ergebnisse der an diesen Baustoffen durchgeführten Radioaktivitätsmessungen werden zusammengestellt und erörtert.

INTRODUCTION

L'enquête avait initialement pour objectif de déterminer les concentrations en radionuclides naturels des phosphates bruts et de certains produits dérivés,notamment les engrais phosphatés et les matériaux de construction à base de gypse.

Au stade actuel, le bilan pour les phosphates bruts et les engrais a pu être réalisé alors que les résultats relatifs aux matériaux de construction sont fragmentaires; cependant l'enquête se poursuit et sera élargie à d'autres types de matériaux de construction.

TECHNIQUES DE MESURE

Les échantillons ont été conditionnés dans des flacons de 250 ml avec addition de charbon actif pour assurer la remise à l'équilibre des chaînes radio-actives ^{226}Ra ^{206}Pb et ^{228}Th ^{208}Pb.

Les mesures ont été exécutées par spectrométrie γ avec détecteur NaI dans une zone d'énergie de 0 à 2 MeV couvrant les principaux pics γ des descendants du ^{228}Th (essentiellement : 238 keV du ^{212}Pb, 510 et 583 keV du ^{208}Tl), du ^{232}Th (911 et 969 keV de l'^{228}Ac), du ^{226}Ra (242, 295 et 352 keV du ^{214}Pb, 609, 1112 et 1764 keV du ^{214}Bi) ainsi que du ^{40}K à 1460 keV.

Les échantillons sont introduits dans le puits d'un cristal NaI(Tl) de 17,8 cm Ø x 15,2 cm de haut connecté à des sélecteurs d'impulsion INTERTECHNIQUE à 400 canaux, reliés à un système informatique elaboré autour d'un mini-ordinateur MULTI-20 de 32 K octets.

Le traitement du spectre et le calcul des activités se fait sous contrôle d'un opérateur disposant d'une console de visualisation, par la méthode d'ajustage par moindres carrés, à partir de spectres standard obtenus dans des conditions identiques avec des solutions radioactives étalonnées.

La mesure directe de l'uranium est rendue délicate par le fait que l'émission γ de l'^{238}U est faible (seul le descendant ^{234m}Pa émet des γ à 767 keV-0,2% et 1001 keV 0,59% ainsi que le ^{234}Th à 63,3 keV - 3,8% et 92,6 keV - 5,4%) tandis que l'activité de l'^{235}U est 20 fois inférieure et que la mesure de son principal pic γ à 185 keV est perturbée par la présence du pic ^{226}Ra à la même énergie; de plus, par suite de la séparation de l'U et du Ra au cours de la fabrication des engrais, le rapport de ces 2 éléments varie très fortement et une forte activité en ^{226}Ra empêchera la mesure de l'U.

Celui-ci a été dosé par activation neutronique; environ 1 g de l'échantillon est mis en solution par une attaque en milieu HNO_3; après filtration, la solution ramenée à 10 ml est mise dans un flacon en polyéthylène et irradiée dans le convoyeur pneumatique de BR1 pendant 5 minutes dans un flux de 2.10^{11} neutrons/sec.cm^2 en même temps que des solutions étalons d'U.

La mesure est ensuite effectuée sur le pic γ de 75 keV de $1'^{239}U$, au moyen d'un détecteur Ge(Li) de 10% d'efficacité relative, couplé à un sélecteur d'impulsions à 4000 canaux. Le spectre est ensuite transféré dans un mini-ordinateur MULTI-20 et interprété au moyen d'un programme de traitement de spectres Ge(Li).

La présence d'U subsistant dans la fraction insoluble est mise en évidence par une activation et une mesure du dépôt sur filtre dans des conditions semblables à celles utilisées pour l'échantillon dissous.

TENEURS EN RADIONUCLIDES NATURELS

I. Phosphates bruts

Origine géographique du phosphate brut	µg U 238/g	pCi Ra 226/g	pCi Th 232/g	pCi K 40/g
Maroc	153,4	42 ± 0,25	< 0,8	4
Maroc	111,8	41 ± 0,3	< 0,8	< 4
Maroc	114,1	38 ± 0,25	< 0,8	< 4
Maroc	111,1	38 ± 0,25	< 0,8	5,2
Maroc	119,5	38 ± 0,3	∿ 0,86	< 4
Maroc	131	43 ± 0,25	⩽ 1	⩽ 5
Maroc	114,9	34 ± 0,25	∿ 1,7	< 4
Maroc	123,2	42 ± 0,4	⩽ 1,5	⩽ 8
Maroc	123,2	39 ± 0,3	∿ 0,94	⩽ 5
Maroc	122,3	40 ± 0,3	⩽ 1	⩽ 6
Maroc	120,7	39 ± 0,25	< 0,8	< 4
Maroc	138,8	42 ± 0,25	< 0,8	< 4
Maroc	121,4	40 ± 0,2	< 0,8	< 5
Maroc	120,2	42 ± 0,25	< 0,6	< 3
Togo	105,6	33 ± 0,25	2,9 ± 0,25	⩽ 3
Kola URSS	7,2	1,1 ± 0,05	2,5 ± 0,03	4,6
USA	154,2	51 ± 0,35	∿ 1,2 ± 0,4	< 5

TABLEAU 1

L'analyse s'est portée sur 17 échantillons pour lesquels les teneurs en U 238, Ra 226, Th 232 et K 40 ont été déterminées. Les données sont repris dans le tableau 1 dans lequel les échantillons sont groupés en fonction de l'origine géographique du minerai.
Les engrais du marché Belge sont principalement fabriqués à partir de phosphates provenant d'Afrique du Nord, c'est pourquoi 15 échantillons du lot proviennent de cette région.

U 238

Pour les 14 échantillons d'origine marocaine la valeur moyenne observée est de 123 µg par gramme de matière avec des valeurs extrêmes de 153,4 et 111,1 µg. Pour les échantillons provenants du Togo et des Etats-Unis les concentrations sont du même ordre de grandeur, alors que seul le minerai originaire de Kola (URSS) montre une teneur nettement inférieure (7,2 µg/g).

Ra 226

La teneur moyenne du minerai marocain est de 40 pCi/g de matière et varie entre 38 et 43 pCi/g, à comparer avec le phosphate brut américain contenant 51 pCi et d'URSS 1,1 pCi.

Th 232 et K 40

Pour le minerai marocain les teneurs en radionuclides sont égales ou inférieures à la limite de détection; deux échantillons d'autre provenance montrent des valeurs significatives en Th 232 : 2,9 pCi pour le minerai du Togo et 2,5 pCi pour celui de Kola.

II. Engrais phosphatés

Les tableaux 2, 3 et 4 donnent les teneurs en radionuclides naturels de plusieurs types d'engrais fabriqués à partir des phosphates bruts mentionnés dans le tableau 1.
Les engrais sont groupés en superphosphates 18% et complexes de composition diverse.
Deux cas de différents engrais du type "SUPERPHOSPHATE", fabriqués à partir du même minerai, sont rapportés.

U 238 (µg/g de matière)

Type d'engrais	Teneur en nuclide du minerai	Teneur en nuclide de l'engrais	Rapport U 238 $\frac{\text{engrais}}{\text{minerai}}$ en %
Superphosphate 18%	114,1	69,6	61
Superphosphate 18%	111,1	67,1	60
Superphosphate 18%	119,5	77,1	65
Superphosphate 18%	122,3	73,9	60
Superphosphate 18%	120,7	88,7	73
Superphosphate 18%	121,4	79,2	65
Superphosphate 18%	154,2	63,7	41
Superphosphate 18%	131	73,3	56
Superphosphate 40%	131	150,6	115
Superphosphate 45%	131	179,7	137
Superphosphate 18%	114,9	80,7	70
Superphosphate " enrichi	114,9	107,8	94
Complexe	120,45	34,7	29
Complexe	138,8	42,6	31
Phosphate double	153,4	44,6	29
Composé 11:11:22	130	43,3	33
Composé 18:8:20	120,2	33,6	28
Composé non déterminé	111,8	19,4	17
Binaire	7,2	17,2	238
Super simple	153,4	92,2	60

TABLEAU 2

Ra 226 (pCi/g de matière)

Type d'engrais	Teneur en nuclide du minerai	Teneur en nuclide de l'engrais	Rapport Ra 226 $\frac{\text{engrais}}{\text{minerai}}$ en %
Superphosphate 18%	38 ± 0,25	26 ± 0,2	68
Superphosphate 18%	38 ± 0,25	26 ± 0,15	68
Superphosphate 18%	38 ± 0,3	27 ± 0,2	71
Superphosphate 18%	40 ± 0,3	27 ± 0,15	67
Superphosphate 18%	39 ± 0,25	26 ± 0,2	67
Superphosphate 18%	40 ± 0,2	26 ± 0,15	65
Superphosphate 18%	51 ± 0,35	22 ± 0,15	43
Superphosphate 18%	43 ± 0,25	27 ± 0,3	63
Superphosphate 40%	43 ± 0,25	19 ± 0,15	44
Superphosphate 45%	43 ± 0,25	17 ± 0,2	40
Superphosphate 18%	34 ± 0,25	28 ± 0,15	82
Superphosphate " enrichi	34 ± 0,25	19 ± 0,2	56
Complexe	39 ± 0,3	9,7 ± 0,15	25
Complexe	42 ± 0,25	12 ± 0,15	29
Phosphate double	42 ± 0,25	20 ± 0,2	48
Composé 11:11:22	43 ± 0,25	$\leqslant$ 0,4	$\leqslant$ 0,9
Composé 18:8:20	42 ± 0,25	10 ± 0,25	24
Composé non déterminé	41 ± 0,3	7,8 ± 0,1	19
Composé binaire	1,1 ± 0,05	< 1,5	-
Compose super simple	42 ± 0,25	26 ± 0,2	62

TABLEAU 3

Dans les tableaux 2 et 3 les teneurs relatives en U 238 et Ra 226 des engrais par rapport au minerai sont indiquées.

Pour l'U 238 (tableau 2) on observe que dans les engrais du type "SUPERPHOSPHATE 18%" pour 9 échantillons analysés, en moyenne 61% du nuclide subsiste dans l'engrais avec des valeurs variant entre 41 et 73%.

Pour les engrais du type COMPLEXE cette teneur résiduelle varie fortement (entre 17 et 238%).

Dans les deux series du type SUPERPHOSPHATE pour lesquelles différentes qualités d'engrais sont produites à partir du même minerai on constate des quantités résiduelles croissantes en U 238 avec une augmentation du taux d'enrichissement en phosphate.

Pour le Ra 226 (tableau 3) la teneur résiduelle moyenne dans l'engrais SUPERPHOSPHATE est du même ordre de grandeur que pour l'U 238 (66%) avec des valeurs extrêmes de 43% et 82%.

Les COMPLEXES montrent également des grandes variations dans les quantités résiduelles de nuclides (entre 0,9 et 52%).
Contrairement aux observations pour U 238 un enrichissement croissant des engrais SUPERPHOSPHATES se traduit par une décroissance en teneur de Ra 226.

Th 232 et K 40 pCi/g de matière

Type d'engrais	Th 232	K 40
Superphosphate 18%	< 0,6	< 4
Superphosphate 18%	< 0,8	5,2
Superphosphate 18%	< 0,8	⩽ 5
Superphosphate 18%	< 0,8	⩽ 4
Superphosphate 18%	< 0,5	< 3
Superphosphate 18%	< 0,7	< 4
Superphosphate 18%	< 0,7	< 4
Superphosphate 18%	⩽ 0,5	∿ 5,9
Superphosphate 40%	⩽ 0,5	∿ 4,2
Superphosphate 45%	⩽ 0,6	∿ 3,5
Superphosphate 18%	⩽ 0,8	⩽ 4
Superphosphate enrichi	⩽ 1	⩽ 5
Complexe	⩽ 0,4	1
Complexe	< 0,4	130 ± 0,7
Phosphate double	∿ 0,97	∿ 4,7
Composé 11:11:22	⩽ 0,2	160
Composé 18:8:20	< 0,6	160 ± 1,5
Composé non déterminé	< 0,4	130
Binaire	7	44
Super simple	⩽ 0,8	3

TABLEAU 4

En ce qui concerne le Th 232 qui n'a pu être mis en évidence que dans les minérais provenants du Togo (2,9 pCi/g) et d'URSS (2,5 pCi/g) on observe une augmentation dans l'engrais dérivé (engrais binaire) du phosphate brut d'URSS (7 pCi/g).

Pour le K 40 seuls des engrais du type COMPLEXE montrent dans quelques cas une teneur supérieure à celle du minérai.

III. Matériaux de construction à base de gypse.

La production annuelle de ce type de matériaux est estimée, pour la Belgique, à environ 85.000 tonnes, la fabrication des panneaux GYPROC représentant de 40 à 50% du marché belge. Les échantillons ont été fournis par les Services du Ministère des Affaires Economiques.
Les premiers résultats sont mentionnés dans le tableau 5.
Il convient de signaler que cette enquête se poursuit et sera élargie ensuite aux autres types de matériaux de construction utilisés couramment en Belgique.

Nature du produit ou nom commercial	µg U238/g.mat.	K40 pCi/g	Ra226 pCi/g	Th232 pCi/g
A) Phosphate brut		$\leq$ 2,7	17,4	$\approx$ 0,3
Goldband		3,2	23,3	0,96
Rotband		$\leq$ 3,4	22,1	$\leq$ 0,4
MP 75		$\leq$ 2,4	24,7	0,9
B) A 40	0,73	$\leq$ 0,3	0,3	$\leq$ 0,03
Sprylith	4,6	$\leq$ 1,9	15,1	0,3
R 40	5,0	$\leq$ 2,7	16,5	$\leq$ 0,3
Sprytal	1,5	0,78	0,7	0,07
B 40	5,2	$\leq$ 2,5	16,5	$\leq$ 0,3
C) Plâtre du type Hemihydrate	1,2	$\leq$ 0,5	0,02	$\leq$ 0,05
D) Gypsin n°1		0,24	0,10	0,03
Gypsin n°2		0,40	0,28	0,04
Gypsin n°3		$\leq$ 0,27	0,38	$\leq$ 0,03
Gypsin n°12		$\leq$ 0,25	0,21	$\leq 3.10^{-3}$
E) Plaque Rygips		1,13	0,15	0,09

TABLEAU 5

NATURAL RADIOACTIVITY IN BUILDING MATERIALS IN DENMARK

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SUMMARY. Activity concentrations of K-40, Ra-226 and Th-232 in Danish building materials have been measured by use of a Ge(Li)-spectrometer. The measured samples have included materials such as concrete, concrete ballast, cement, clay brick, aerated concrete, sand-lime brick, clinker, light-weight aggregate, natural gypsum and fly ash with prime emphasis on samples of concrete ballast and clay brick. The results of the investigations have been compared with the NEA proposal concerning radiation protection standards for buiding materials.

RESUME. RADIOACTIVITE NATURELLE DANS LES MATERIAUX DE CONSTRUCTION AU DANEMARK. On a déterminé les concentrations d'activité de K-40, Ra-226 et Th-232 dans des matériaux de construction danois à l'aide d'un spectromètre Ge(Li). Les mesures ont porté sur des échantillons de matériaux tels le béton, le béton de cailloux, le ciment, la brique d'argile, le béton cellulaire, la brique silicocalcaire, le clinker, les agrégats légers, le plâtre naturel et la cendre volante, l'accent étant mis surtout sur le béton de cailloux et la brique d'argile. Les résultats des analyses ont été comparés avec la proposition de l'AEN relative aux normes de protection contre les rayonnements pour les matériaux de construction.

KURZFASSUNG. NATÜRLICHE RADIOAKTIVITÄT IN BAUSTOFFEN IN DÄNEMARK. Aktivitätskonzentrationen von K-40, Ra-226 und Th-232 in dänischen Baustoffen wurden mit einem Ge(Li)-Spektrometer gemessen. Zu den gemessenen Proben gehörten Stoffe wie Beton, Zuschlagstoffe, Zement, Tonziegel, Porenbeton, Kalksandstein, Klinker, Leichtzuschlagbeton, Naturgips und Flugasche; Schwerpunkt lag bei Betonzuschlag- und Tonziegelproben. Die Ergebnisse der Untersuchungen wurden mit dem Vorschlag der NEA über Strahlenschutznormen für Baustoffe verglichen.

Introduction

External radiation in buildings consists of cosmic radiation and radiation from ground and building materials. The radiation from the ground and building materials is caused by the presence of the primordial radionuclides K-40, Ra-226 and Th-232.

The contribution from building materials to the external irradiation of the population can be estimated by measuring the content of the primordial radionuclides in commonly used building materials.

The concentrations of K-40, Ra-226 and Th-232 in 248 samples of Danish building materials from 12 different material groups have been measured. The results have been evaluated by calculating the gammaindex m defined by (1):

$$m = \frac{C_K}{3000} + \frac{C_{Ra}}{300} + \frac{C_{Th}}{200}$$

where C_K, C_{Ra} and C_{Th} are the activity concentrations of K-40, Ra-226 and Th-232 given in Bq/kg (1 Bq/kg = 0,027 pCi/g).
In a building composed of building materials with m = 1, the increase in the annual effective dose-equivalent above the outdoor value (indoor occupancy factor of 0,8) will on an average be 0,5 mSv for flats and 0,3 mSv for single-family houses (1).

Experimental techniques

The measurements of the concentrations of K-40, Ra-226 and Th-232 were performed by a 90 cm^3 Ge(Li) detector and a Canberra Model 8100 multi channel analyser. The detector has a photopeak efficiency of 16% and an energy resolution of 2,1 keV for the gammatransition of 1332 keV from Co-60.

The samples were measured in a 1,8 liter can of Marinelli type. For K-40 the gammatransition of 1461 keV was used, for Ra-226 the gammatransitions of 352 keV (Bi-214), 609 keV (Bi-214) and 1120 keV (Bi-214), for Th-232 the gammatransitions of 583 keV (Tl-208) and 911 (Ac-228). A measured spectrum of a low-activity sample of sand is shown in figure 1.

Calibrations of the equipment were done by use of potassiumchloride dissolved in water and standard solutions of Ra-226 and Th-228 moulded in gypsum. The total error of the calibration is estimated to be less than 10%. Counting times 5.000-10.000 sec were used in order to get reasonable counting

statistics.

After measurement the samples were dried and the net weight determined.

If the minimum detectable activity is defined as three times the standard deviation of the background, the following values are obtained for a 1000 g sample and a counting time of 5000 sec:

K-40 20 Bq/kg, Ra-226 4 Bq/kg, Th-232 4 Bq/kg.

Results

The measured materials were commonly used building materials in Denmark with prime emphasis on samples of clay brick and concrete ballast. All results are given in tabel I.

107 samples of concrete ballast representatively selected from the whole country (see figure 2) were measured. The ballast included sand, gravel and macadam. The distributions of the measured values of the activity concentrations are given in figure 3. For K-40 a mean value of 360 Bq/kg (<20 - 1150) with a standard deviation of 240 Bq/kg was found, for Ra-226 a mean value of 19 Bq/kg (<4-95) with a standard deviation of 16 Bq/kg and for Th-232 a mean value of 13 Bq/kg (<4-56) with a standard deviation of 11 Bq/kg. The measured values for sand were generally among the lowest values and for macadam correspondingly among the highest measured values. F. ex. granite from Bornholm (island in the Baltic) has the highest measured values for all three radionuclides. The calculated values of the gammaindex varied from less than 0,04 to 0,98 with a mean of 0,25 and a standard deviation of 0,11.

For clay brick 79 samples representing the whole country (see figure 2) were measured. The distributions of the measured activities are given in figure 4. Here the results for K-40 varied between 340 and 900 Bq/kg with a mean of 630 Bq/kg and a standard deviation of 110 Bq/kg. For Ra-226 a mean of 42 Bq/kg (23-86) with a standard deviation of 11 Bq/kg and for Th-232 a mean of 34 Bq/kg (21-58) with a standard deviation of 8 Bq/kg were measured. The values of gammaindex varied between 0,37 and 0,78 with a mean of 0,52 and a standard deviation of 0,07.The standard deviation of the measured activity concentrations is relatively small ($\sim$ 20%) giving the small standard deviation of 0,07 (13%) of the gammaindex. The two distinctly highest values of Ra-226 (83 and 86 Bq/kg) were measured in two samples of light-weight "moler"brick (clay of vulcanic origin).

The measurements of cement, natural gypsum and fly ash are also representative of these material groups in Denmark. The mean values of the gammaindex are: cement 0,16, natural gypsum less than 0,05 and fly ash 1,19.

Samples of concrete, white bricks, aerated concrete, aerated concrete with alum shale, clinker and light-weight aggregate have also been measured and the results are also given in tabel I. These values may not be representative for these material groups.

Discussion

All measured samples except samples of fly ash and aerated concrete based on alum shale of Swedish origin have values of gammaindex of less than 1 and radium concentrations of less than 100 Bq/kg.

The calculated mean values of the gammaindex for concrete ballast and cement imply a value of the gammaindex of an average Danish concrete of 0,22, when the proportion between ballast, cement and water of 82:11:7 is assumed. This will correspond to an average increase in the annual effective dose-equivalent above the outdoor value (indoor occupancy factor of 0,8) in flats built of concrete of 0,11 mSv. The calculated mean values of the gammaindex of clay bricks of 0,52 imply correspondingly an average annual effective dose-equivalent in single-family houses built of bricks of 0,16 mSv.

The measured mean and maximum values of the activity concentrations in Danish concrete and clay bricks are low compared with reported values from most other countries. A comparison with the other Nordic countries is given in tabel II for clay bricks and in tabel III for concrete ballast.

The measured materials with higher activity concentrations such as aerated concrete based on alum shale are not used any more in Denmark and have only been used very little in the past. Fly ash from coalfired powerplants is today used in the production of cement and may replace a part of the cement in concrete in the future. If 20% of the cement is replaced by fly ash this will on average increase the gammaindex for the finished concrete from 0,22 to 0,25.

Acknowledgement

I wish to thank K. Bækmark, CtO, Aalborg Portland for his great help in collecting all samples of concrete ballast, cement and fly ash.

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Tabel I. The content of radioactive nuclides and the calculated gammaindex m defined as $m = C_K/3000 + C_{Ra}/300 + C_{Th}/200$ of Danish building materials.

Material	No. of samples	Activity concentrations, Bq/kg	K-40	Ra-226	Th-232	m	Comment
concrete ballast	107	min. mean max.	240 360 1150	<4 19 95	<4 13 56	<0.04 0.25 0.98	adequate sampling
cement	6	min. mean max.	<20 90 140	9 20 30	<4 12 21	<0.06 0.16 0.29	adequate sampling
concrete	6	min. mean max.	280 360 420	13 16 24	9 13 17	0.24	not adequate sampling
clay bricks	79	min. mean max.	340 630 900	23 42 86	21 34 58	0.37 0.52 0.78	adequate sampling
white bricks	3	min. mean max.	160 260 340	6 9 11	4 8 11	0.16	not adequate sampling
aerated concrete	2	min. mean max.	280	18	10	0.20	not adequate sampling
aerated concrete with alum shale (Swedish origin)	2	mean	1190	670	53	2.90	not adequate sampling
clinker (from different countries)	13	min. mean max.	230 710 1080	22 66 108	22 55 73	0.73	not adequate sampling
light-weight aggregate	3	min. mean max.	860 910 1000	36 40 43	37 45 51	0.66	not adequate sampling
natural gypsum	12	min. mean max.	<20 <20 35	<4 8 13	<4 <4 6	<0.04 <0.05 0.09	adequate sampling
fly ash	10	min. mean max.	190 730 1030	110 150 210	74 90 160	0.96 1.19 1.58	adequate sampling
insulating material	5	mean	<190	<40	<40	<0.40	not adequate sampling

Tabel II The mean content of radioactive nuclides in clay bricks in the Nordic countries, Bq/kg (1 Bq/kg = 0,027 pCi/g)

Country	No. of samples	C_K	C_{Ra}	C_{Th}	m	ref	comment
Denmark	79	630	42	34	0,52		adequate sampling
Finland	37	984	80	62	0,90	(2)	adequate sampling
Norway	18	1140	63	74	0,96	(3)	
Sweden	12	960	96	127	1,28	(4)	not adequate sampling

Tabel III The mean content of radioactive nuclides in concrete ballast in the Nordic countries, Bq/kg (1 Bq/kg = 0,027 pCi/g)

Country	No. of samples	C_K	C_{Ra}	C_{Th}	m	ref	comment
Denmark	107	360	19	13	0,25		adequate sampling
Finland	266	967	34	39	0,63	(2)	adequate sampling
Norway	137	650	28	36	0,49	(3)	concrete, adequate sampling
Sweden	306	814	48	72	0,79	(5)	adequate sampling

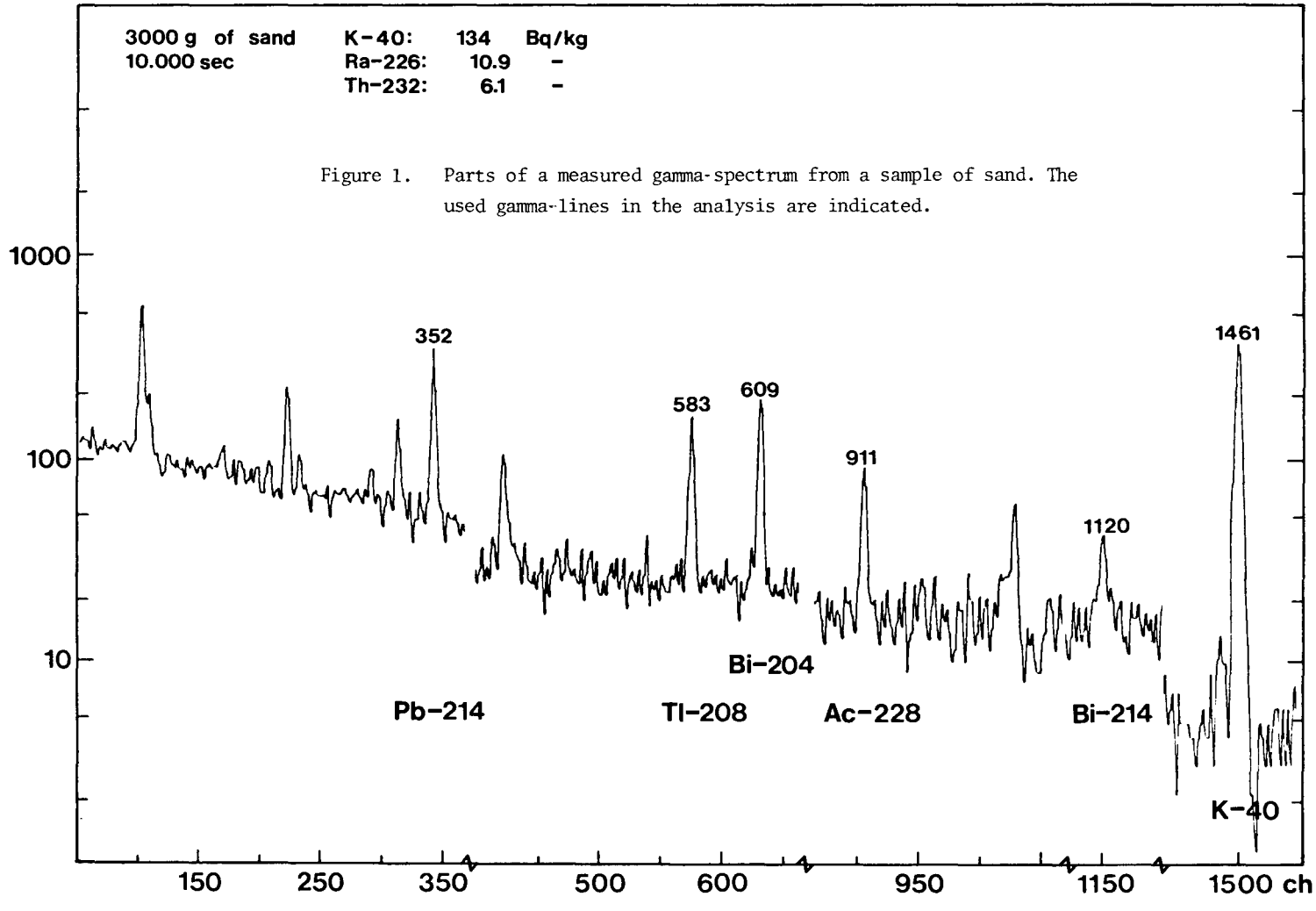
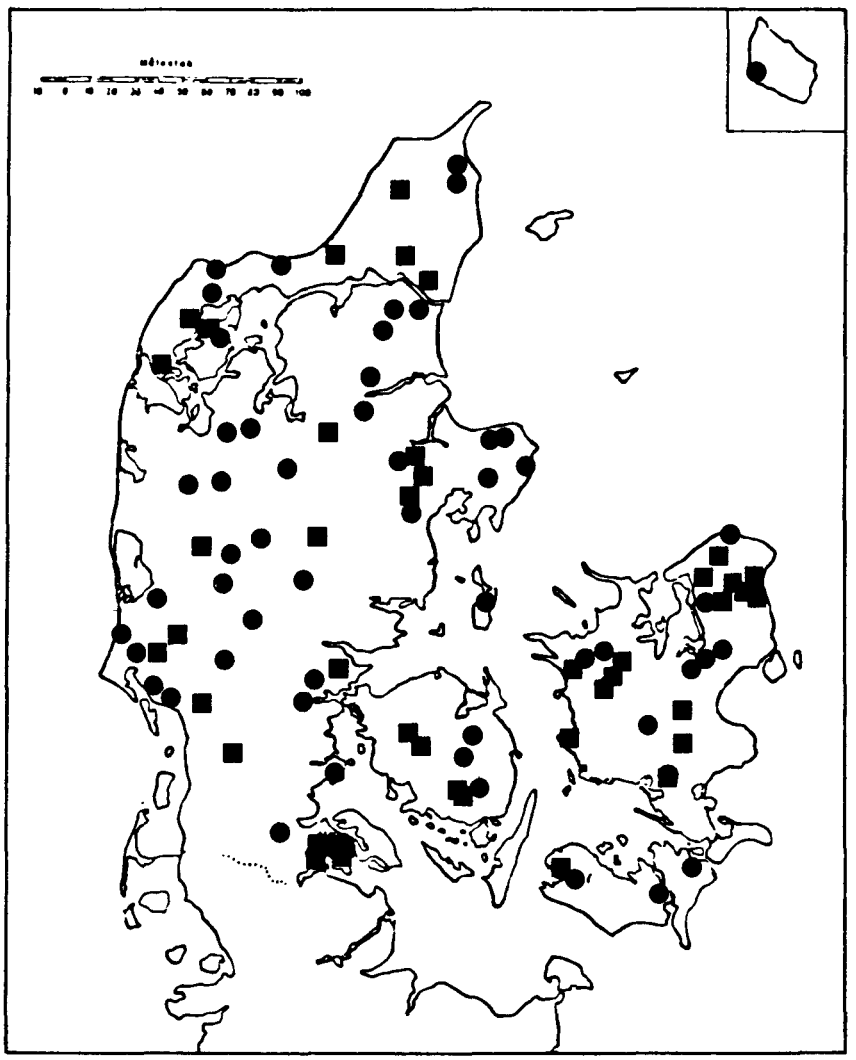


Figure 1. Parts of a measured gamma-spectrum from a sample of sand. The used gamma-lines in the analysis are indicated.

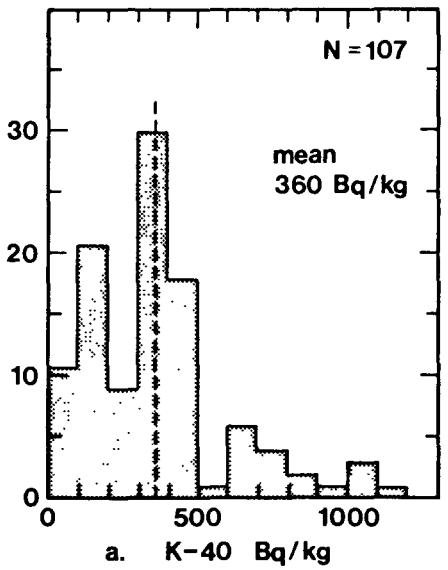
Figure 2. Sampling places for concrete ballast and clay bricks.

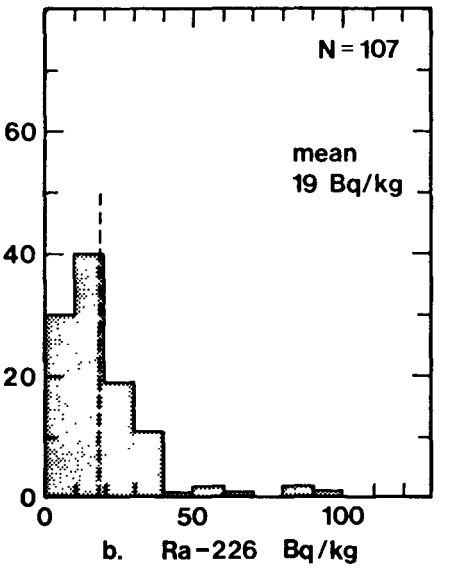


■ Clay brick

● Concrete ballast

Figure 3. Number of measurement in each activity interval for concrete ballast.





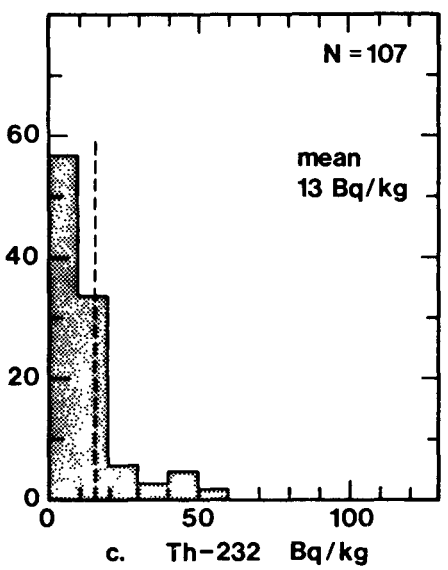
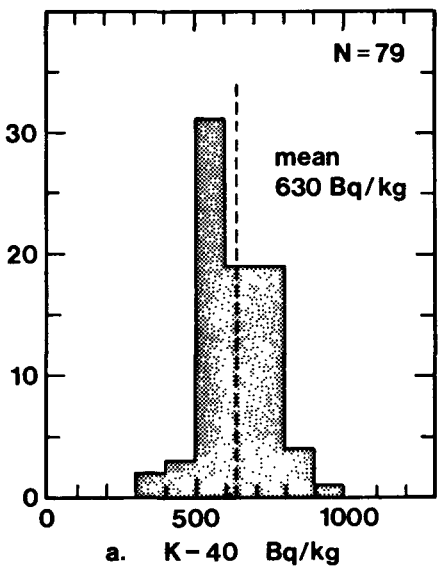
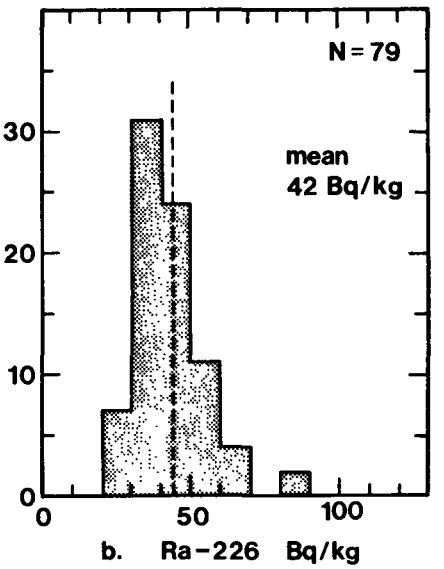
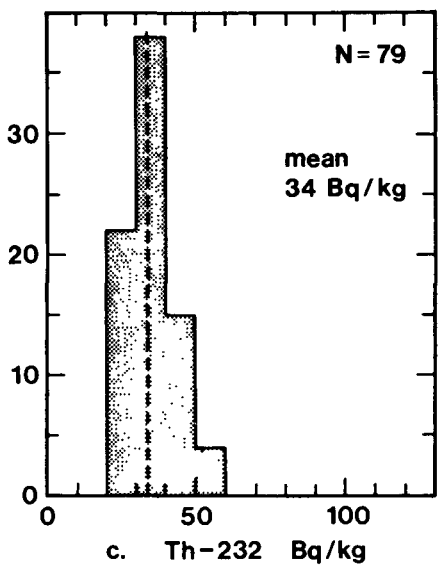


Figure 4. Number of measurement in each activity interval for clay bricks.







EXHALATION MEASUREMENTS AND INDOOR RADON LEVELS IN DENMARK

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SUMMARY. The exhalation of radon from a series of building materials has been measured by placing samples of the materials in closed containers and following the build up of activity in the containers.
The values measured range from 0.3 to 4000 $\frac{atoms}{m^2 \cdot s}$

(1.6×10^{-5} to 0.22 $\frac{pCi}{m^2 \cdot s}$)

In a single case the exhalation from the walls of a room was measured directly using exhalation cans sealed onto the walls, and it was shown that the measured exhalation values correspond reasonably well with the long term average room air activity under the given ventilation conditions.
Approximately 700 air samples were taken from about 100 houses and apartments under normal living conditions and analyzed for radon content. The overall average concentration was 0.24 pCi/l. Houses made of brick usually had concentrations about one half of those built of concrete.
Winter concentrations (with low ventilation rates) were often higher than summer concentrations by a factor of up to four.
The highest concentration encountered was about 10 pCi/l. A few houses showed concentrations consistently higher (above 2-3 pCi/l) than neighbouring houses built of similar materials.
An investigation of the effect of improved insulation (lowered ventilation rates) on radon concentrations in apartments is under way.

RESUME. MESURE DES EMANATIONS DE RADON ET NIVEAUX ATTEINTS DANS LES BATIMENTS AU DANEMARK. Le radon émanant d'une série de matériaux de construction a été mesuré sur des échantillons de matériaux placés en conteneurs fermés dans lesquels on a suivi l'évolution de l'activité.
Les valeurs mesurées vont de 0,3 à 4000 $\frac{atomes}{m^2 \cdot s}$

($1,6 \cdot 10^{-5}$ à 0,22 $\frac{pCi}{m^2 \cdot s}$)

Dans un seul cas, l'émanation par les murs d'une pièce a été mesurée directement à l'aide de boîtes scellées sur les murs; on a montré que les valeurs mesurées correspondent assez bien à l'activité moyenne à long terme de l'air de la pièce dans les conditions de ventilation données.

Dans une centaine de maisons ou appartements, on a prélevé dans des conditions de vie normales quelque 700 échantillons d'air dont on a déterminé la teneur en radon. La concentration moyenne globale était de 0,24 pCi/l. Les concentrations dans les maisons en briques, étaient en général moitié de celles dans des maisons en béton. En hiver (faible taux de ventilation), les concentrations étaient souvent plus fortes qu'en été jusque d'un facteur 4.
La concentration maximale trouvée était d'environ 10 pCi/l. Quelques maisons avaient des concentrations régulièrement supérieures (plus de 2-3 pCi/l) à celles des maisons voisines construites de matériaux similaires. Une étude de l'effet d'une meilleure isolation (taux de ventilation réduit) sur les concentrations de radon dans les appartements est en cours.

KURZFASSUNG. EMISSIONSMESSUNGEN UND RADONPEGEL IN WOHNUNGEN IN DÄNEMARK.
Die Radonemission gewisser Baustoffe wurde gemessen, indem Proben dieser Stoffe in geschlossenen Behältern untergebracht und der Aktivitätsaufbau in den Behältern festgestellt wurde.
Die gemessenen Werte liegen zwischen 0,3 und 4000 $\frac{\text{Atome}}{m^2 \cdot s}$

($1{,}6 \cdot 10^{-5}$ bis 0,22 $\frac{pCi}{m^2 \cdot s}$)

In einem Fall wurde die von den Wänden eines Zimmers emittierte Radioaktivität direkt mit einem hermetisch an den Wänden befestigten Emissionsmessbehälter gemessen, und es wurde nachgewiesen, dass die gemessenen Emissionswerte gut mit der über lange Zeiträume ermittelten durchschnittlichen Aktivität der Raumluft unter den gegebenen Belüftungsverhältnissen übereinstimmen.
Rund 700 Luftproben wurden in ca. 100 normal bewohnten Häusern und Wohnungen entnommen und auf ihren Radongehalt analysiert. Die mittlere Gesamtkonzentration betrug 0,24 pCi/l. In mit Mauerziegeln erbauten Häusern waren die Konzentrationen fast durchweg etwa halb so hoch wie in Betonhäusern. Im Winter (bei geringer Belüftung) waren die Konzentrationen häufig bis zu viermal höher als im Sommer.
Die höchste festgestellte Konzentration lag bei 10 pCi/l. In einigen Häusern waren die Konzentrationen merklich höher (über 2-3 pCi/l) als in benachbarten Häusern aus ähnlichen Baustoffen.
Eine Untersuchung über die Auswirkungen einer besseren Isolierung (geringere Frischluftzufuhr) auf die Radonkonzentrationen in Wohnungen ist im Gange.

The levels of radon and radon daughters in an indoor environment are under given ventilation conditions governed by the feed of radon from the room surfaces. In many cases most of the radon is supplied from the soil, and this is reflected in the fact, that the air activities of radon usually are higher in basements and ground floor rooms than at higher floors.

When, however, building materials with a high radium concentration are being used, the resulting radon exhalation may substantially contribute to unacceptably high levels of airborne radioactivity.

Measurement of exhalation rate.

The exhalation properties of building materials can be studied by enclosing samples of the materials in closed containers and follow the growth of radon activity in the container by analysis of air samples.

If the radon concentration in the container (atoms per unit volume) at equilibrium is A_0, the dead space volume of the container V_d, the radon exhaling surface S, and the decay constant of radon λ, then the net exhalation rate E' at equilibrium can be written |1|

$$E' = \frac{\lambda A_0 V_d}{S} \qquad (1)$$

This figure, however, will be lower than the free exhalation rate E corresponding to exhalation into an empty space, or into an ordinary room.

The difference is caused by the so-called back diffusion, which will slow down the exhalation rate from the material as the activity (concentration) in the container grows.

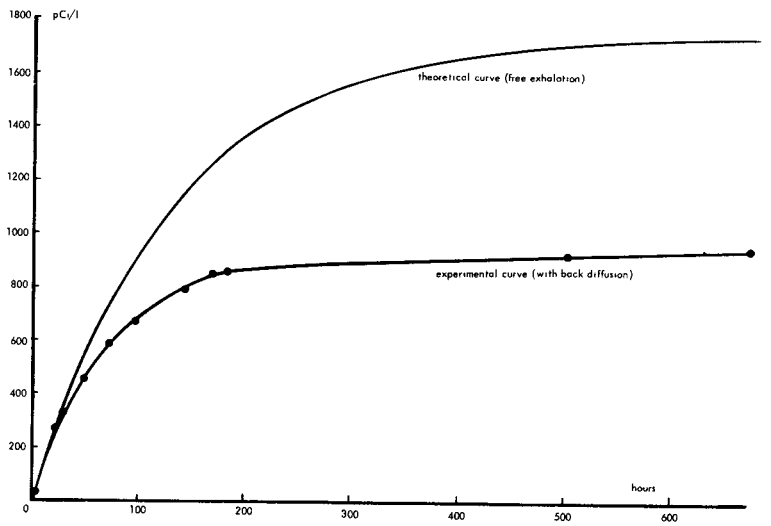

Fig. 1. Radon activity in a closed container as a function of time.

This effect is shown in Fig. 1, where the activity in a closed container is plotted as a function of time for a sample of a rather active (and porous) exhaling material (a phosphate fertilizer). It appears that the experimental curve lies well below the (theoretical) curve extrapolated from the initial value of the activity growth rate, corresponding to free exhalation, the difference between the equilibrium values being approximately 50 % of the free exhalation value.

This difference depends upon a) the dimensions of the sample relative to the diffusion length of the material, and b) the ratio between the dead volume of the container and the void volume of the sample, i.e. the product of the porosity and the sample volume. If the dead space is chosen as approximately twice the sample volume, then for most building materials with porosities below 0.5 the exhalation value determined from eq. (1) will not differ from the free exhalation value by more than 10 - 15 % at the most.

A series of building materials have been tested in the way described yielding exhalation rates as shown in the table.

material	exhalation rate		radon concentration	
	$\frac{atoms}{m^2 s}$	$\frac{pCi}{m^2 s}$	R_0	$R_{0.5}$
			$\frac{pCi}{l}$	
chipboard fiberboard gypsum board (nat. gypsum)	< 1	$< 5 \cdot 10^{-5}$	$< 5 \cdot 10^{-2}$	$< 8 \cdot 10^{-4}$
rockwool bricks	2	$1.1 \cdot 10^{-4}$	$1 \cdot 10^{-1}$	$1.6 \cdot 10^{-3}$
light weight concretes (dan. origin)	20-30	$1.1\text{-}1.7 \cdot 10^{-3}$	1-1.5	$1.6\text{-}2.4 \cdot 10^{-2}$
ordinary concretes (danish dep.)	130-180	$7\text{-}10 \cdot 10^{-3}$	7-10	$1.1\text{-}1.5 \cdot 10^{-1}$
gypsum tiles (chemogyps.)	800	$4.5 \cdot 10^{-2}$	43	$6.4 \cdot 10^{-1}$
alum shale light weight concrete	1400-4000	$8\text{-}22 \cdot 10^{-2}$	76-200	1.1-3.2

Exhalation rates and contributions to radon concentrations for various building materials.

If the material is being used in a room the contribution to the radon concentration from the material is given by

$$R_n = \frac{\lambda}{\lambda+n} \cdot \frac{S}{V} \cdot E \qquad (2)$$

where E is the (free) exhalation rate, S the exhaling area, V the volume of the room, n the ventilation rate and λ the decay constant of radon.

If all surfaces of a room are covered with the exhaling material, then $\frac{S}{V} \sim 2\ m^{-1}$. The corresponding radon concentrations R_0 for an unventilated room and $R_{0.5}$ for a room with a ventilation rate of 0.5 h^{-1} are also shown in the table. These concentrations are the maximum contributions from the materials under the given ventilation conditions.

Radon levels in Danish dwellings.

It appears from the figures in the table that some commonly used building materials at low ventilation rates can contribute significantly to indoor radon levels.

In order to see to what extent this actually is the case a field investigation was undertaken involving approximately 100 Danish houses and apartments according to the following schedule: twenty sampling sites were selected, and from each of these a one liter air sample was taken once a week for five weeks. Then another series of twenty locations were chosen a.s.o. Measurements were taken in brick buildings as well as in buildings made of various kinds of concrete. No effort was done to ensure a certain condition of the sampling locations, since it was the intention to get air samples which were as representative as possible of the normal condition of the room.

For three of the concrete buildings (or building complexes) measurements were taken in the winter as well as in the summer.

The main results of the measurements were the following: The all-over average of the measurements were 0.24 pCi/l with the lowest value close to zero (lower than the detection limit of about 0.05 pCi/l) and a maximum value of about 10 pCi/l. 59 (or 8 %) out of a total of 718 measurements gave concentrations above 1 pCi/l. In ordinary houses made of bricks only in badly ventilated basement rooms concentrations above this value (1 pCi/l) were found. In houses made of concrete the average radon concentration was about the double of that in brick houses, measured at the same time of the year. In concrete houses with mechanical ventilation (or open windows) concentrations above 1 pCi/l were seldom found, while in unventilated houses higher concentrations were commonly encountered.

In the three locations where measurements were taken at two seasons the ratio between the winter and summer concentrations were 1.3, 2.9 and 5.1. The higher winter concentrations are undoubtedly due to a lower natural ventilation rate during this season. The figure of 1.3 was found for a high-rise building with mechanical ventilation.

As a part of a project on energy saving efforts, it is planned to investigate the effect of improved insulation and reduced ventilation on radon levels in a group of dwellings. These measurements have been started, but results are not yet available.

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A TECHNIQUE FOR MEASURING THE RELATIVE EXHALATION RATES OF RADON AND THORON FROM BUILDING MATERIALS

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SUMMARY. In the technique described Radon and Thoron simultaneously exhaling from the same portion of surface area of a sample of material diffuse into two identical cylindrical detection chambers mounted above the sample. Detection of the exhaled gases and their respective daughter products in the two chambers is by means of their alpha activities recorded on cellulose nitrate dielectric track detectors. One of the detection chambers is preceded by a porous barrier through which the gases must diffuse to enter this chamber. Because of its short half life relative to that of Radon the Thoron concentration is considerably reduced by decay in its transit by diffusion through the barrier. From measurements of the time integrated alpha track densities on the dielectric track detectors in the two detection chambers the relative exhalation rates of Radon and Thoron from a sample of material may be obtained. A description of the state of development of this technique and some preliminary results obtained are presented.

RESUME. TECHNIQUE DE MESURE DES TAUX RELATIFS D'EMANATION DU RADON ET DU THORON DE MATERIAUX DE CONSTRUCTION. Dans la technique décrite, le radon et le thoron qui se dégagent simultanément de la même portion de surface spécifique d'un échantillon de matériau diffusent dans deux chambres de détection cylindriques identiques montées au-dessus de l'échantillon. La détection des gaz émanés et de leurs descendants respectifs dans les deux chambres s'effectue par enregistrement de leur activité alpha au moyen de détecteurs de traces diélectriques sur le nitrate de cellulose. L'une des chambres de détection est précédée d'une barrière poreuse à travers laquelle les gaz doivent diffuser pour entrer dans cette chambre. En raison de sa courte période par rapport à celle du radon, la concentration du thoron est considérablement réduite par décroissance lors de sa diffusion à travers la barrière. La mesure des densités de trace alpha en fonction du temps par les détecteurs de traces diélectriques installés dans les deux chambres de détection permet de déterminer les taux relatifs d'émanation du radon et du thoron à partir d'un échantillon de matériau. Le niveau de développement de cette technique fait l'objet d'une description, complétée de quelques résultats déjà obtenus.

KURZFASSUNG. VERFAHREN ZUR MESSUNG DER RELATIVEN EXHALATIONSRATEN VON RADON UND THORON AUS BAUMATERIALPROBEN. Bei dem beschriebenen Verfahren diffundieren das gleichzeitig aus demselben Oberflächenbereich einer Materialprobe austretende Radon und Thoron in zwei identische, über der Probe angebrachte zylindrische Nachweiskammern. Der Nachweis der exhalierten Gase und ihrer Folgeprodukte in den beiden Kammern erfolgt mittels ihrer Alpha-Aktivitäten auf dielektrischen Zellulosenitrat-Spurendetektoren. Vor einer dieser Detektionskammern befindet sich eine poröse Trennwand, durch die die Gase diffundieren müssen, um in die Kammer zu gelangen. Wegen der im Vergleich zum Radon kurzen Halbwertszeit wird die Thoronkonzentration auf ihrem Diffusionsweg durch die Trennwand weitgehend durch Zerfall reduziert. Aus Messungen der über die Zeit integrierten Alpha-Spurendichten auf den dielektrischen Spurendetektoren in den beiden Detektionskammern können die relativen Exhalationsraten von Radon und Thoron aus einer Materialprobe bestimmt werden. Der Entwicklungsstand dieses Verfahrens wird beschrieben, und es werden einige vorläufige Ergebnisse gebracht.

Introduction.

Most building materials in common use contain small amounts of Radium-226 and Thorium 232 which are responsible for the production within the materials of the radioactive gases Radon-222 ($T_{\frac{1}{2}}$=3.82 days) and Radon-220 ($T_{\frac{1}{2}}$=54.5sec) respectively. Because of its short half life the migration of Thoron (Radon-220) through a material is considerably restricted in comparison with that of Radon (Radon-222). Thoron exhalation from building materials can thus be considered as taking place essentially from the outer surface layers. The radiological health hazard associated with the breathing of air containing Thoron and its daughters is generally considered to be of less importance than that due to the inhalation of Radon and its daughters. One reason for this is because of the relatively short half lives of radon daughters compared to the 10.6 hour half life of Pb-212 (ThB). With deep lung clearance times of approximately 10 hours and those from the upper bronchial tree of about 1 hour it would appear that lung deposited thoron daughters are substantially cleared before they decay (1), (2). Other comparisons with radon and its daughters (i.e. effect of ventilation etc.) generally indicate that thoron and its daughters in the air are of lesser radiological significance. It should however be stated that the hazard from inhaled thoron and its daughters is not well understood their being in addition an absence of any epidemiological studies on this matter. For these and other reasons the exhalation of thoron from building materials has received little attention. Radon exhalation from such materials has on the other hand been the subject of a number of studies. (3), (4).

The building industry in recent years has shown an increasing requirement for low cost raw materials. This has given rise to the incorporation of industrial waste products into building materials. This development in many ways has proved to be economically and ecologically sensible. Some examples of this approach have however created radiological difficulties. One notable example of this has been the use of by-product gypsum of high radium content arising as a waste in the fertilizer industry. The resulting elevated radon exhalation rate from plasterboard made from by-product gypsum and its radiological implications are well documented (5). The possibility also exists that industrial waste (or natural material) with a high specific activity of thorium may at some time become incorporated

into extensively used building materials. Mean specific activities of thorium in excess of 200 Bq/kg (5.4 pCi/g) have been reported for some building materials at present in use in Europe (6). It should be noted that in most techniques where the exhalation of radon from building materials is studied the nature of the technique used in usually such that the presence of a high exhalation rate of thoron would not normally be observed. It is felt by the author in the light of the above considerations that a need exists for the development of techniques which are simple, inexpensive and reliable to measure the exhalation of both thoron and radon from building materials. One such technique is presented below.

Description of Method.

Due mainly to its short half life reliable methods for the direct measurement of thoron at atmospheric concentrations have only been developed within the last 20 years (7). In the case of thoron exhalation from construction material the static and dynamic techniques of Wicke and Porstendörfer are worthy of note (8). In the static method the exhalation rate of both radon and thoron may be inferred from two gamma spectroscopic measurements made on a piece of material sealed in an airtight PVC box. The first gamma measurement is made 3 hours after sealing and the second after 30 days which is close to equilibrium. In the dynamic method a direct measurement of thoron flux is made by using a special scintillation chamber of the type designed by Bogen, Sappok and Schumann where thoron is determined from delayed alpha particle coincidences from the decay of Thoron and ThA (9).

In the present work the Radon-Thoron exhalation measuring technique presented is based on the approach used by Ward, Fleischer and Mogro-Campero to eliminate thoron "noise" in soil gas radon surveys employed in this discovery of subsurface uranium (10). The principle used is that radon may be separated from thoron by the use of either a gas-permeable or a diffusional type barrier. Due to its very short half-life relative to that of radon the thoron concentration is considerably reduced by decay in transit through the barrier.

In the interests of simplicity and reproducibility it was decided, in the technique described here, to use a diffusional barrier composed of a layer

of small glass spheres of mean radius 300 microns in preference to a gas-permeable polymeric membrane barrier. A number of measurements on the diffusion of thoron through such a barrier was made using the simple apparatus shown in Figure 1. Thoron is produced by Thorium Oxide in the base of the apparatus. The alpha particles emitted by thoron and its daughters when they have reached equilibrium in the cylindrical detection volumes are recorded on discs of Kodak Pathé LR115 alpha sensitive cellulose nitrate plastic mounted as shown. The alpha tracks were made visible by etching the exposed plastic detectors in 2.5 N Na OH at 60^{o} C. A series of exposures using diffusional barriers ranging in thickness from 0 to 7.5 cm was made.

It is convenient at this stage to define a ratio R as that between the concentration of thoron in a detection volume with a barrier present and the thoron concentration in that volume with no barrier. It can be seen that a value of R may be obtained by taking the ratio between the track densities recorded in volume A and volume B. The results obtained are presented in Figure 2 as a plot of barrier thickness against R. It can be seen for example that a barrier of thickness 2 cm is sufficient to produce a reduction of approximately 90% in the thoron concentration.

It can be shown in general (10) for apparatus of the type depicted in Figure 1 that:-

$$R = \frac{DA(V_1+V_2)}{DA(V_1+V_2)+\lambda h V_1 V_2} \qquad \text{Equation (1)}$$

Here D is the diffusion constant of thoron in the barrier of cross-sectional area A and thickness h. V_1 and V_2 are the detection and source volumes respectively and λ is the decay constant of the radioactive gas used. For the experiments described using the apparatus in Figure 1 a mean value of $D_{thoron} = 0.026$ cm^2 sec^{-1} was obtained which is comparable to its reported values in loose sediments and sands (11), (12).

The type of apparatus used to simultaneously measure both radon and thoron exhalation from the same portion of surface area of a sample of material is shown in Figure 3. It is made of mild steel and is hermetically sealed to the surface of a building material using epoxy resin as the principal

sealant. In Figure 4 are shown details of the top of the detecting volumes in which the plastic detectors are mounted. The significance of this diagram will be described below. In the apparatus a diffusional barrier of thickness 8 cm and composed of the small glass spheres is used. Using equation(1)taking $D_{radon} = D_{thoron}$, the appropriate values of V_1 and V_2 and h = 8 cm yields $R_{radon} = 0.995$ and $R_{thoron} = 0.034$. A plastic detector mounted in volume A therefore records almost exclusively alpha particles from radon and its daughters. A detector mounted in volume B on the other hand records alpha particles from both radon and thoron and their respective daughters. Information on the relative exhalation rates of radon and thoron from a material may thus in principle be obtained from the alpha densities recorded in the two detection volumes.

Experimental Procedure.

When an exhalation apparatus is attached to a surface initially the enclosed air will only contain amounts of radon and thoron appropriate to the levels in the ambient air. After sealing the concentrations of the gases will increase with time due to exhalation from the material. In addition the respective daughter products of the gases will grow towards their appropriate equilibrium values. When the concentrations of either thoron or radon become sizeable fractions of the corresponding concentrations in the pore spaces of the exhaling material the effective exhalation rate of each gas will be reduced (4). Eventually final equilibrium concentrations of the gases are reached within the apparatus.

A direct comparison between the radon and thoron exhalation rates can only properly be made if measurements of the alpha activities in the detection volumes are made when all states of equilibrium of both the gases and their respective daughters are reached in the closed air of the apparatus. Here this is accomplished by sealing the exhalation apparatus to the surface of a material and by waiting 30 days before commencing to record alpha activity in the detection volumes. In practice the plastic detectors are not introduced into the detecting volumes until a minimum of 30 days has elapsed since the sealing of the apparatus to the surface of a material. The method of introducing the plastic detectors into the equilibrium air of the apparatus is indicated in Figure 4. At the top of each detection volume is an airtight window consisting of a thin glass microscope slide cover attac-

hed by means of epoxy resin. A metal washer of thickness 1.5 mm is left resting on top of the glass window. To commence the exposure period of a plastic detector the detector carrier is slowly screwed into position. Near the end of its travel the front surface of the carrier comes into contact with the loose metal washer. Further screwing of the carrier transmits force to the glass window which is shattered thereby commencing the exposure of the plastic detector to the alpha activity. The washer and shattered glass fall into the detection volume. The dimensions involved are such that in its final position the plastic detector is flush with the inner surface of the tip of the detection volume. Minimal disturbance of the active air occurs during the emplacement of the detectors.

Geometrical and other considerations.

Disregarding insignificant branches in the decay chains an equilibrium atmosphere of Radon, Thoron and their respective short-lived daughters emit alpha particles of seven different energies E ranging from 5.49 MeV in the case of Radon-222 to 8.78 MeV in the case of ThC' (Po-212). These alpha particles have a spread in mean range in air at atmospheric pressure from approximately 4 to 8.5 cm. For the LR 115 plastic detector used and the chemical etching conditions employed only alpha particles in the approximat -e energy range $1<E_{\alpha}<4$MeV produce tracks which can be counted visually. This means in effect that for each alpha emitting species only alpha particles emitted by atoms of this species at a distance in air of $r_1 < r < r_2$ from a plastic detector surface will produce tracks which can be counted. Here r_1 is the distance in air required to reduce the energy of the alpha particle in question to 4 MeV and r_2 is the distance in air required to reduce its energy to 1 MeV. For each of the seven alpha emitters under consideration there is a different set of r_1 and r_2 values. This gives rise to a different geometrical detection efficiency for each emitter. Detection geometry calculations are further complicated by the fact that the daughter products of thoron and radon may become attached to the walls of the cylindrical detection volumes.

Geometrical detection efficiencies were calculated for different possible and likely distributions of the various daughter products in the aerosol free air of the detection volumes. Consideration was given in these calculations to the dimensions of the detection volumes, r_1 and r_2 values,

alpha recoil effects, the various half lives involved, and information on the diffusion constants of unattached radon and thoron daughter. On these bases it was considered not unreasonable to assume that the alpha emitting species would be distributed in a manner somewhat similar to one of the following two distributions:-

Distribution 1: Radon and Thoron in the airborne phase with all of the alpha emitting daughter products deposited on the walls.

Distribution 2: Radon, Thoron and ThA in the airborne phase with the remainder of the alpha emitting daughter products deposited on the walls.

When comparing the alpha track densities (tracks/mm^2) arising from the exposure of the plastic detectors to the two gases and their respective daughters it is convenient to define a ratio k_e.

Here $k_e = \frac{\text{Track density due to radon and daughters}}{\text{Track density due to thoron and daughters.}}$

This definition of k_e is taken to refer to equal time exposures to equal specific air activities of radon and thoron in equilibrium with their respective daughters within a detection volume.

The values of k_e calculated were as follows:-

For distribution 1: $k_e = 1.18$

For distribution 2: $k_e = 1.11$

For the dimensions of the detection volumes used the value of k_e does not appear to be very sensitive to variations on the daughter product distributions. Until satisfactory experimental determinations of k_e are made a mean value of $k_e = 1.14$ will be used.

At the end of an exposure period the plastic detectors are removed from the detection volumes and are etched as described. Due to the small internal dimensions of the exhalation apparatus shown in figure 3 the effects of

back diffusion on the exhalation rates of both radon and thoron may be considerable. No attempt was therefore made to determine the value of the exhalation rates of the individual gases in the present work. Because of their almost identical masses it is reasonable to expect that exhalation rates of these two radon isotopes from the same portion of material surface will be equally affected by back diffusion. The ratio between the equilibrium radon and thoron concentrations in volume B of the apparatus is thus in principle equal to the ratio between the exhalation rates of these two gases from the material. This ratio is the property of the material which is determined in the method described here. It is called the exhalation ratio E.R. and defined as follows:-

$$E.R. = \frac{\text{Radon Activity Exhalation Rate}}{\text{Thoron Activity Exhalation Rate}}$$

$$\equiv \frac{\text{Equilibrium Air Activity of Radon in Volume B}}{\text{Equilibrium Air Activity of Thoron in Volume B}}$$

It is to be noted that because of radiological considerations E.R. is defined in terms of activities of the two gases instead of in terms of concentrations of atoms of the gases.

If the track densities measured for the plastic detectors exposed to the alpha activities in volume A and in volume B are T_A and T_B respectively it can be shown that:-

$$E.R = \left[\frac{T_A - R_{thoron}\, T_B}{R_{radon}\, T_B - T_A}\right] \cdot \frac{1}{K_e} \qquad \text{Equation (2)}$$

Equation (2) has been used in the experimental determinations of E.R.

Initial Results and Conclusions.

At present only some preliminary measurements of E.R. from three types of building materials have been obtained using the present technique. The three materials investigated to date have been concrete slabs, by-product gypsum plasterboard and granite. Each experiment has lasted approximately two months consisting of a 30 day pre-exposure period followed by a one month exposure period. In the case of materials of low exhalation rates

exposure periods of many months may be required. This is in order to achieve a high enough alpha track density on the plastic detectors to facil-itate rapid counting and good counting statistics. The preliminary results obtained are presented in the table below.

Type of Material	Alpha track production rate Tracks mm^{-2} day^{-1} $metres^{-2}$		$(E.R)^{-1}$	(E.R)
	Detector from Volume A	Detector from Volume B		
Concrete Slabs	33.8	45.34	0.4	2.48
Granite	32.2	35.4	0.1	9.7
By-product gypsum plasterboard	67.0	77.7	0.18	5.5

For comparison purposes the track density information is expressed as an alpha track production rate as shown in the table. It can be seen from this table that the exhalation of thoron activity takes place from all three materials. In the case of the by-product gypsum plasterboard indepenent confirmation of thoron exhalation was obtained. This was achieved by means of a crude variation of the present method in which ZnS(Ag) scintillation counters were used as alpha detectors instead of the plastic detectors. An approximate E.R value of 3.3 was indicated which compares reasonably well with the 5.5 E.R value in the table.

An improvement in the sensitivity and accuracy of the technique presented here may be obtained if a plastic detector known as CR-39 is used instead of the cellulose nitrate plastic currently used. The unique alpha particle recording properties of CR-39 have only been recently recognised (13), (14). Unlike cellulose nitrate where only energies in the approximate range $1<E_{\alpha}<4$ MeV produce visible tracks with the most recently developed form of the CR-39 plastic alpha particles greater in energy than 10 MeV can be "seen"(15).

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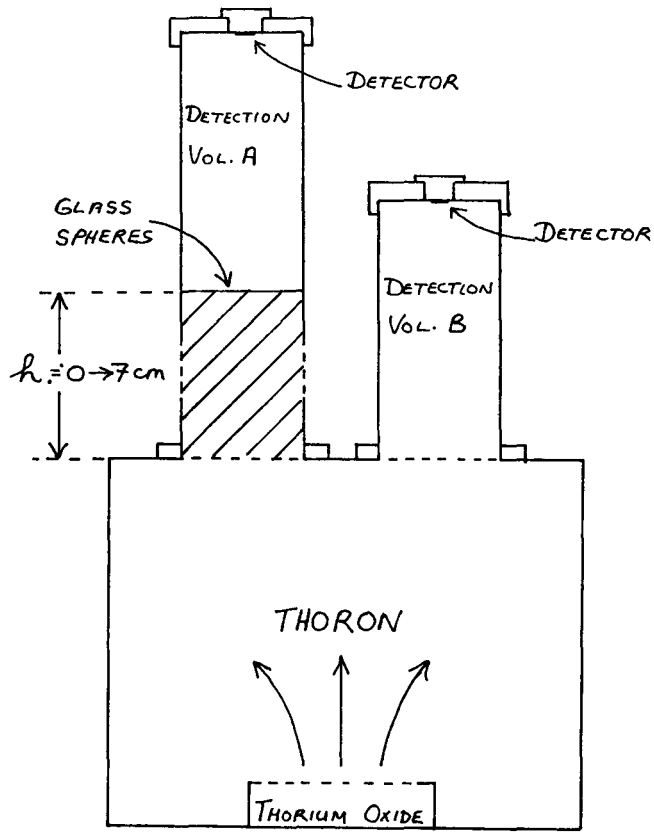

Figure 1. Apparatus used to determine the Diffusion Constant of Thoron.

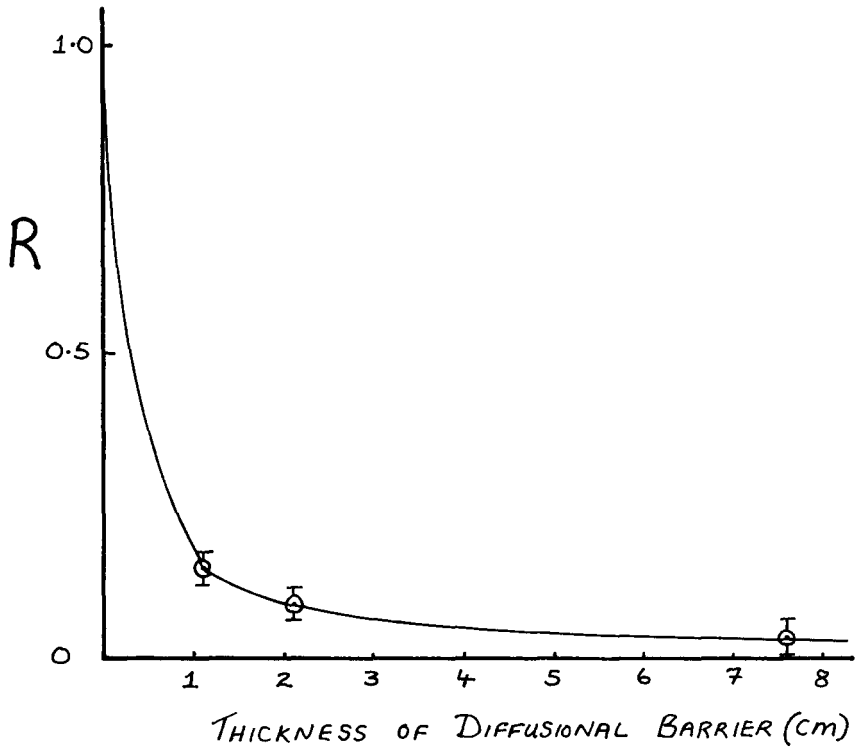

Figure 2. Dependence of R on thickness of Diffusional Barrier.

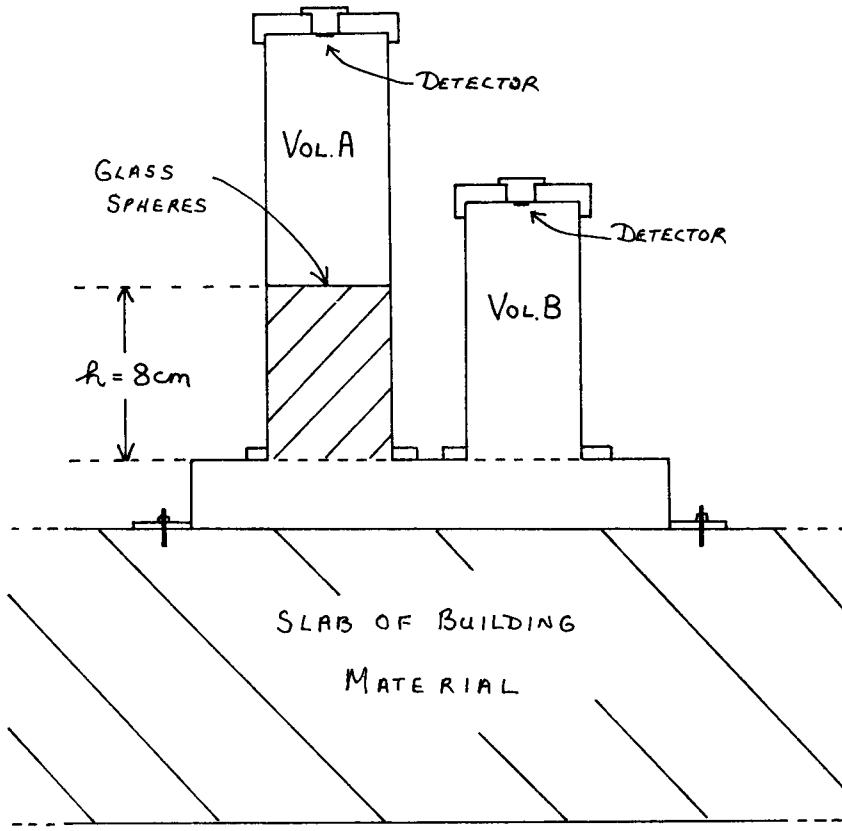

Figure 3. Schematic diagram of Exhalation Apparatus in use.

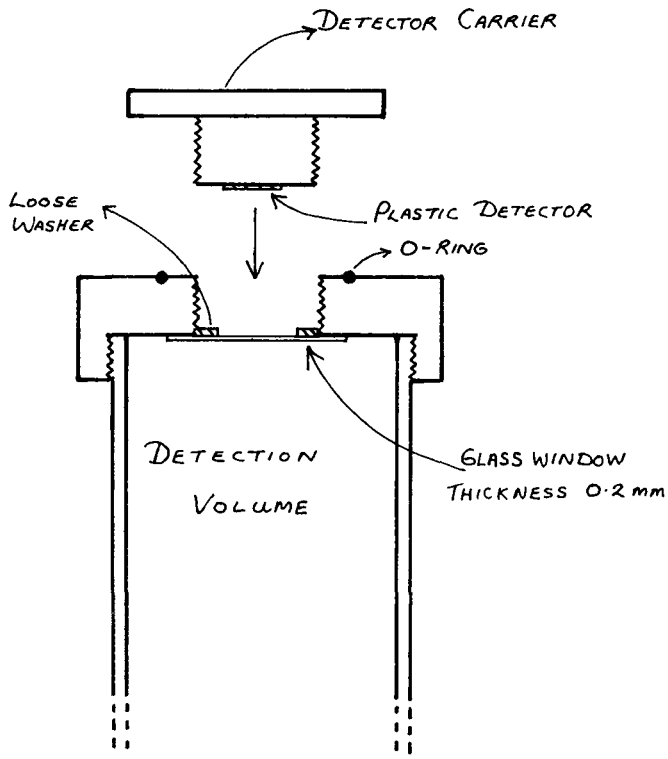

Figure 4. Diagram of top of a Detection Volume.

INDOOR MEASUREMENTS OF NATURAL RADIOACTIVITY IN SWEDEN

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SUMMARY. A country-wide investigation of the gamma radiation levels in Swedish dwellings has been made. A preliminary value for the average absorbed dose to the gonads was found to be 0.6 mGy/a. Several parameters have been studied e.g. types of building material. Studies have been made of specific activities of naturally occurring radionuclides in building materials, concentrations of radon and radon daughters in air indoors, in some cases with simultaneous measurements of ventilation rates, and long-term variations of radon. On the basis of the above studies, of the results of research projects on ventilation rates in various types of Swedish houses and of the investigation by HULTQVIST from the 1950s, an attempt has been made to show how the radon concentrations have changed or may be changed by human activities. The calculations indicate that the average concentration of radon has increased from 23 Bq/m^3 for dwellings existing in 1950 to 57 Bq/m^3 in dwellings existing in 1975.

Houses with unusually high concentrations of radon are reviewed. They are built entirely of aerated concrete based on alum shale. High radon concentrations have also been found in houses built of other building materials on sites covered with old waste tailings from alum shale workings.

RESUME. MESURES DE LA RADIOACTIVITE NATURELLE DANS LES BÂTIMENTS EN SUEDE. Une enquête sur les niveaux de rayonnement gamma dans les habitations a été mené dans toute la Suéde. On a trouvé 0,6 mGy/a comme valeur préliminaire de la dose moyenne absorbée aux gonades. Plusieurs paramètres ont été étudiés, dont le type de matériau de construction. Les mesures ont porté sur l'activité spécifique des radionucléides naturels présents dans ces matériaux, les concentrations de radon et de ses descendants dans l'air des bâtiments avec parfois la mesure simultanée des débits de ventilation et les variations à long terme du radon. Sur base de ces études, des résultats des projets de recherche sur les débits de ventilation dans différents types de maisons suédoises et de l'enquête de HULTQVIST des années 1950, on a tenté de montrer comment les concentrations

de radon changent ou peuvent changer par les activités humaines. Les calculs indiquent que la concentration moyenne due au radon dans les habitations est passé de 23 Bq/m^3 en 1950 à 57 Bq/m^3 en 1975.
Les maisons à concentrations anormales de radon sont examinées. Elles sont bâties tout de béton cellulaire sur des schistes alunifères. On a relevé aussi de hautes concentrations de radon dans des maisons bâties d'autres matériaux sur des remblais de vieux résidus d'exploitation des schistes alunifères.

KURZFASSUNG. MESSUNGEN DER NATÜRLICHEN RADIOAKTIVITÄT IN WOHNUNGEN IN SCHWEDEN. Eine landesweite Untersuchung der Gamma-Strahlenpegel in schwedischen Wohnungen wurde durchgeführt. Für die mittlere absorbierte Gonadendosis wurde ein vorläufiger Wert von 0,6 mGy/a ermittelt. Die Untersuchung bezog sich auf verschiedene Parameter und umfasste u.a. mehrere Baustoffsorten. Untersucht wurden die spezifischen Aktivitäten natürlicher Radionuklide in Baustoffen, die Konzentrationen von in der Raumluft enthaltenem Radon und dessen Folgeprodukten, wobei in einigen Fällen gleichzeitig die Belüftungsraten gemessen wurden sowie die Schwankungen des Radongehalts über lange Zeiträume. Auf der Grundlage dieser Untersuchungen, anhand der Ergebnisse von Untersuchungen über die Belüftungsverhältnisse in verschiedenen Arten schwedischer Häuser und mit Hilfe der von HULTQVIST in den 50er Jahren durchgeführten Untersuchung wird zu zeigen versucht, wie die Radonkonzentrationen durch menschliche Tätigkeiten verändert wurden bzw. verändert werden können. Die Berechnungen erweisen, dass die aus Radon-Folgeprodukten stammende mittlere Konzentration von 23 Bq/m^3 bei den im Jahre 1950 bestehenden Wohnungen auf 57 Bq/m^3, bei den bis 1975 errichteten Wohnungen angestiegen ist.
Es wird ein Überblick über die Häuser mit ungewöhnlich hohen Radonkonzentrationen gegeben. Sie sind ganz aus Porenbeton auf der Grundlage von Alaunschiefer (alum shale) erbaut. Hohe Radonkonzentrationen fand man auch in Häusern aus anderen Baustoffen auf Grundstücken, die mit alten Rückständen aus der Alaunschieferverarbeitung bedeckt waren.

In the beginning of the 1950s, Hultqvist (1) studied the gamma radiation level and the concentration of radon indoors in houses built before 1946 in Sweden. He also determined the content of radionuclides in building materials.

In the middle of the 1970s, extensive new studies were started on the natural radioactivity including country-wide investigations of the contents of radionuclides in building materials, of the gamma radiation levels indoors and of the concentrations of radon and radium in tap water. In various types of houses the concentration of radon and radon daughters are being studied.

Building materials

Table I shows the contents of the naturally occurring radionuclides potassium-40, radium-226 and thorium-232 in Swedish building materials (2, 3). The measurements have been performed by gamma spectrometry. For various reasons not all the given values are representative of the situation today. The building materials market changes with time and some of the measurements were made some years ago. Not all the values are representative for the production in the whole country.

In Sweden aerated concrete has been produced in several material mixes with different properties regarding the resultant activity levels. The bulk density is about 0.5 g/cm^3 and the material is porous with differences in colour depending on the ballast used in the particular case. When alum shale is used as ballast, the colour is greyish blue in different shades. If sand is used the material is white or light grey. The aerated concrete based on alum shale has not been produced since 1975.

It can be seen from Table I that the highest activities in commonly used Swedish building materials are found in aerated concrete based on alum shale. The enhanced activity level is due to the high content of radium-226. The average activity concentration of radium-226 varies between different factories from 700 Bq/kg (20 pCi/g) to 2 400 Bq/kg (65 pCi/g) according to Table II. Houses built in the same way with the same amounts of aerated concrete can therefore have very different gamma levels and very different concentrations of radon in the air with the same air exchange rate.

The white aerated concrete based on sand of limestone shows the lowest content of radionuclides together with bricks of the same material.

Gamma radiation indoors

A country-wide investigation of gamma radiation in Swedish houses has been made with $CaSO_4$-Dy dosimeters (4). Three such detectors were sent to the occupants of about 1 500 dwellings picked out at random. The final results are not yet available but some preliminary results are shown in Table III and Fig 1. Table III shows the absorbed dose rates from gamma radiation in Swedish houses related to the main building materials in the outer walls. The high content of radionuclides in alum shale is seen in the higher gamma radiation levels in the dwellings.

The dwellings have been selected at random from each county. The lowest dose rates in air, 3 nGy/h (0.3 μrad/h), have been measured in the Gotland County where the average value was 21 nGy/h (2.1 μrad/h). The highest dose rates have been measured in the Stockholm County, where the average value was 127 nGy/h (12.7 μrad/h), the lowest value was 15 nGy/h (1.5 μrad/h) and the highest was 530 nGy/h (53 μrad/h). This is illustrated in Fig 1, where the values have been given as annual absorbed tissue dose rates in mGy/a.

From Table I it can be seen that the most radioactive building material, the aerated concrete based on alum shale, has an activity concentration of the gamma radiating nuclides 5 to 6 times as high as that in normal bricks or concrete. If the latter materials are replaced by the same amount of weight of aerated concrete, absorbed dose rates between five and six times as large might be expected.

As has been pointed out, the aerated concrete has a low bulk density, and therefore the weight of a house is lower than for bricks or concrete. However, in light materials the gamma radiation penetrates the material more easily. Field measurements have shown absorbed dose rates in air of about 100 nGy/h (10 μrad/h) in houses built of concrete or bricks. In houses where all the material consists of aerated concrete, therefore, about 500 to 600 nGy/h (50 - 60 μrad/h) might be expected from the measurements of building materials. In investigations of various types of houses, measurements of the dose rates in air have shown values of 700 nGy/h (70 μrad/h) as an

average for a group of houses. This value is higher than was found in the country-wide investigation.

Measurements of radon and radon daughters

The concentrations of radon and radon daughters depend on the ventilation rates during a period prior to as well as during a measurement. The ventilation rate varies with several parameters such as the meteorological conditions and the behaviour of the occupants, especially when natural draught ventilation is used. This kind of ventilation system is the most common in multi-family houses built before 1950 and in one-family houses. When a measurement is to be made, therefore, the occupants were asked to keep the windows shut from the evening prior to the measurements.

Air samples were taken with evacuated containers and measured with regard to the radon concentration in ionization chambers (5). Filter samples have been taken for determining the equilibrium equivalent concentration of radon (EEC). The total alpha activity of the filters was measured with a zinc sulphide detector. The EEC was calculated using Kusnetz´method (6). The measurements of the daughter products are based on measurement of energy. In this report the corresponding activity concentrations have been given.

Continuous recording of radon has been performed with an ionization chamber through which the air passes continuously (7). The radon daughters are filtered before the passage through the chamber.

The measurements of the air exchange rates have been made by the National Swedish Insitute for Building Research with a tracer gas method (8). N_2O is released in the dwelling and an analyzer records the decrease of the concentration of the gas.

Radon concentrations indoors

The radon concentration indoors depends on several parameters. The effects of some of them are illustrated by continuous recordings of the radon concentrations in some types of houses.

Fig 2 shows an example of how the radon concentration varies in a typical bedroom in a detached house with a natural draught ventilation system.

As is common in Swedish homes, the bedroom had no direct ventilation ducts.

The door to the room was closed at the beginning of the measurement and the radon concentration then increased. From the beginning of the measurement, the wind direction was towards the opposite side of the house. After some hours the wind direction changed so that it blew towards the side of the house where the measured room had its window. This caused a decrease in the radon level to one third of the previous value.

Bedrooms without direct ducts for exhaust air present special problems not only for houses with natural draught ventilation systems. When the doors and windows to a bedroom are closed, the concentration of radon and radon daughters may increase as in Fig 3 which shows a continuous recording in a house with a mechanical exhaust air ventilation system. When the door to the bedroom was closed the radon level increased by a factor of five.

The type of house is also a factor of importance for the time variations. Fig 4 shows continuous recordings from two houses built on abandoned waste from alum shale mining. One of the houses, No 121, had a cellar where the radon measurement was performed. The other house was built on a crawl space with four ventilation ducts. The radon concentration in the house with a cellar varied with wind strength, but much less than in a room in the other house. Both houses had natural draught ventilation systems. The large variations of the radon concentrations in the houses on the crawl space are due to the effects of the meteorological conditions on the radon concentration in the foundations as well as in the rooms.

It is very common at present to seal windows and doors in order to conserve energy. The next figure, No 5, shows the effect of sealing two opposite windows in a room in a single-family house with natural draught ventilation. In this room the radon concentration is strongly dependent on the wind strength but not as much on the wind direction. The figure shows that the radon concentration increased by about 50 per cent after the sealing of the windows when the wind strength was only somewhat higher than during the time before the tighting. Then the wind strength increased and the radon concentration decreased but not as much as would be expected from the radon concentration found before sealing the windows.

Another common way to save energy, not officially allowed, is to stop the

mechanical fans during the night. Fig 6 shows how the radon concentration increased in a room in a multi-family house when the fans were stopped each night at midnight and then decreased when the fans were started again at six o´clock in the morning.

The average concentration of radon in Swedish dwellings

In Houston in 1978 I presented a paper (9) in which the average concentration of radon in Swedish dwellings had been calculated. Those calculations were based on studies of the concentrations of radionuclides in building materials, the gamma radiation levels in dwellings, the concentration of radon, radon daughters and ventilation rates indoors in groups of houses, continuous recording of radon in some types of houses and the concentrations of radon in tap water. The calculations were also based on the investigation of Hultqvist from the beginning of the 1950s (1).

A summary of the results is presented in Table IV which shows that the average radon concentration may have increased from 23 Bq/m^3 (0.6 pCi/l) in houses existing in 1950 to 57 Bq/m^3 (1.5 pCi/l) in houses existing in 1975. An occupancy factor of 100 per cent has been used.

Investigations made during 1978 and 1979 have shown lower air exchange rates and higher radon concentrations than were assumed in the calculations. The average concentration in Swedish dwellings might therefore be as much as 50 per cent higher today.

The radon concentration in some groups of houses

The major problem in Sweden is the increasing collective dose equivalent. This is partly due both to the more frequent use of building materials based on stone or clay and to the decreasing air exchange rates.

The decreasing air exchange rates have also resulted in such high radon concentrations indoors that the total risk situations for the individuals have increased in houses with high concentration of radium in the building material or in the ground or with high concentration of radon in the tap water.

Of these radon sources the most difficult problem to solve is the limitation

of dose burdens to people who live in houses built of aerated concrete based on alum shale. This is because the combination of the high concentration of radium in the material, the large amount of the material in each of many houses and the large number of houses in which the material has been used.

A house may be built entirely with aerated concrete or it can have only one wall or a part of a wall of this material. Since the radium contents also vary, as already pointed out, the radon concentrations vary within a large range between the houses.

The highest radon concentrations have been found in houses built entirely of aerated concrete based on alum shale. A group of 9 houses with natural draught ventilation systems was investigated. The averages for the group were 780 Bq/m^3 (21 pCi/l) for the concentration of radon and 410 Bq/m^3 (11 pCi/l) for the equilibrium equivalent concentration of radon, EEC. The lowest average value for a house was 540 Bq/m^3 (15 pCi/l) of radon. In this house the EEC was 290 Bq/m^3 (7.8 pCi/l) and the air exchange rate 0.43 h^{-1} which was the highest air exchange rate found in the group. The highest radon concentration found was 1 160 Bq/m^3 (31 pCi/l). The EEC in this house was 770 Bq/m^3 (21 pCi/l) and the air exchange rate was 0.21 h^{-1} which was the lowest value measured in the group. The measurements were carried out in the autumn with normal meteorological conditions. The results should therefore be reasonably representative of the normal conditions in these houses.

The gamma dose rate in the air was about 650 nGy/h (65 µrad/h) in the centre of the rooms. Local authorities, in their seach for houses with similar conditions, have found higher gamma levels.

It has been estimated that 10 - 20 per cent of the Swedish dwellings (400 - 800 thousand) are built of aerated concrete based on alum shale to a major extent. Of these dwellings 3 to 15 thousand might be built entirely of this material.

Another problem is that presented by houses built on ground which contains high concentrations of radium. Only two years ago we became aware of the fact that houses were built on abandoned waste from alum shale mining as described in my other presented paper. In one of these houses have been found the highest concentrations of radon and radon daughters in Sweden

1000 Bq/m^3 (27 pCi/l) of EEC as an estimated average for a house (10). The concentration of radon in this house varied very much with time as is shown in Fig 4. Measured air exchange rates were between 0.17 h^{-1} and 0.28 h^{-1}.

This problem is not as difficult to manage as that for aerated concrete based on alum shale because the number of dwellings with enhanced concentration of radon is not so large. It has been estimated that 200 to 2 000 houses may be built on ground which contains concentrations of radium which might give enhanced concentration of radon indoors compared with houses on normal ground.

A third problem is the occurrence of radon in tap water. In Sweden about half of the population get their tap water from ground water. Most of the population get their water from centralized water supplies. An investigation of all water supplies in Sweden using ground water and which produce more than 0.2 million cubic metres of water per year have been carried out with regard to the concentration of radon and radium-226. The report will be published in the spring of 1980. The highest value for radon was found to be 150 Bq/l (4 nCi/l) (11).

Higher values of radon are to be expected in private deep-bored wells. Such wells have been investigated to some extent and some of the results have been published (12, 13, 14). The highest value found hitherto in Sweden is 4 000 Bq/l (100 nCi/l) of radon. The next task will be to investigate smaller water supplies and private deep-bored wells selected on a geological basis.

If the conversion factor from radon in the water to radon in the air is taken as $2 \cdot 10^{-4}$ (15) for an air exchange rate of 0.5 h^{-1}, the average radon concentration in the air in houses using the most active tap water in Sweden, can be estimated to be 800 Bq/m^3 (20 pCi/l) of radon and about half that value for radon daughters. If the air exchange rates are lower, as is most usual, the radon concentration indoors might be still higher.

The radiation dose from naturally occurring gamma radiation indoors and outdoors and the estimated risk for cancer

The average dose rates in air are about 100 nGy/h (10 µrad/h) in Swedish

dwellings. The average outdoor dose rate is estimated to be 80 nGy/h (8 μrad/h). With estimated occupancy factors of 20 per cent outdoors and 80 per cent indoors the average dose equivalent for the Swedish population from the terrestrial gamma radiation is estimated to be 0.61 mSv/a (61 mrem/a). To this value must be added the cosmic radiation. For the conversion from absorbed dose rates in air to dose equivalents the factors $7.2 \cdot 10^{-3}$ nGy mSv^{-1} for the outdoor geometry and $6.0 \cdot 10^{-3}$ nGy mSv^{-1} for the indoor geometry have been used (16). The collective dose equivalent is estimated to be about 7 500 manSv/a (750 000 manrem/a) for the Swedish population from the gamma radiation in the houses and the ground.

ICRP (17) has estimated the mortality risk factor for radiation induced cancer to be about 10^{-2} Sv^{-1}. Others have given other risk factors. For example the Swedish Energy Commission (18) has estimated the risk for cancer to be about $2 \cdot 10^{-2}$ Sv^{-1}.

If the ICRP risk factor is used, it may be estimated that about 100 cancer deaths will occur annually in Sweden due to the gamma radiation from the houses and the ground.

The estimated risk from exposure to radon and radon daughters in houses

Present-day knowledge indicates that the major risk from the inhalation of radon and radon daughters is lung cancer. The risk factors for radon and daughters in houses are at present based on epidemiological studies of miners.

For the estimation of the risk for lung cancer from inhalation of radon and radon daughters in houses, the risk factors for miners given by the 1977 UNSCEAR report (15), $200 - 450 \cdot 10^{-6}$ WLM^{-1}, have been used. After correction for a different breathing rate and a different occupancy time for a person in a dwelling in comparison with a worker in a mine, the risk factor obtained is $2.1 - 4.7 \cdot 10^{-6}$ per Bq a/m^3 air of the equilibrium equivalent concentration of radon.

Using this risk factor and an occupancy factor of 80 per cent indoors, between 500 and 1 100 lung cancers may be expected from the inhalation of radon and radon daughters in Swedish houses existing in 1975 for a popula-

tion of 8.4 millions. This is a much higher value than the estimated number of cancers from gamma radiation in houses.

For persons living in the group of houses built entirely of aerated concrete based on alum shale the risk for lung cancer is calculated to be between 0.7 and 1.5 · 10^{-3}. The corresponding risk for cancer from the gamma radiation in the same group of houses is about 0.04 · 10^{-3}. Thus, the major problem for Swedish houses is the concentration of radon and radon daughters.

Countermeasures

These results indicated exposures of such magnitude that they could not be neglected and the National Institute of Radiation Protection therefore passed this information to the authorities responsible for dwellings and to the government. A Committee on measures against radiation hazards in buildings etc was formed. It has been working since March 1979 and a preliminary report was presented two months later (19). This report recommends that local authorities endeavour to identify houses with enhanced concentrations of radon. For guidance in deciding priorities, the committee also proposed provisional action levels while awaiting the permanent limits which the committee has been instructed to work out.

The search for houses with enhanced radon concentrations is carried out in many ways by the local authorities; reports from the owners, studies of archives, interviews with builders, measurements from a car of the gamma levels etc. The houses which this screening indicated might contain high levels of radon are measured with regard to radon daughters.

The measures now recommended are improvements to or reconstruction of the ventilation systems so that the air exchange rate does not fall below 0.5 h^{-1}. The Swedish Building Code of 1975 requires that the air exchange rates must not be below 0.5 h^{-1} for hygienic reasons but this value should not be exceeded to avoid energy wastage. This requirement concerns only dwellings with mechanical ventilation systems. In the coming Swedish Building Code of 1980 natural draught ventilation systems will not be allowed and therefore the requirement will concern all newly built dwellings.

Studies are in progress to determine the effectiveness of possible counter-

measures. These include improvements to ventilation systems, covering of radon-emanating surfaces with barrier-layers such as aluminium foil, removal or replacement of inner walls or wall fillers of unsuitable material and so on. The studies take into account the economic aspects and the practicability of the countermeasures as well as the improvements in radon and radon daughter levels.

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Table I. The content of radioactive nuclides in building materials, Bq/kg

Building materials	No of sampl.	C_{Ra}			C_{Th}			C_K			Ref	Comment
		Min	Aver	Max	Min	Aver	Max	Min	Aver	Max		
Brick	12	41	96	152	100	127	178	560	960	1090	SSI	Not adequate sampling
Bricks of limestone	3	7	10	15	4	8	10	22	160	440	SSI	Not adequate sampling
Concrete	14	32	47	58	56	80	105	577	723	932	Ha 78	Not adequate sampling
Cement	16	20	41	168	24	40	81	21	233	378	Li 64 SSI	Adequate sampling
Concrete ballast (gravel, shingle macadam)	306	7	48	167	3	72	463	137	814	3120	Ha 78	Adequate sampling
Aerated concrete based on sand	24	7	35	130	4	42	155	26	205	490	SSI	Adequate sampling
Aerated concrete based on alum shale (in production 1929-1975)	70	620	1300	2620	30	67	115	618	770	1062	SSI	Adequate sampling
Aerated concrete based on alum shale (in production 1974-1979)	12	320	466	560	24	30	37	488	508	548	SSI	Adequate sampling
Gypsum plasterboards												
Natural gypsum	8	<1	4	9	<1	<1	12	<3	31	68	SSI	Adequate sampling
Phosphogypsum	1		27			65			38		SSI	Not adequate sampling

Table I (cont.)

Building materials	No of sampl.	C_{Ra}			C_{Th}			C_K			Ref	Comment
		Min	Aver	Max	Min	Aver	Max	Min	Aver	Max		
Light-weight aggregate	6	135	170	195	153	164	186	929	1021	1110	SSI	Not adequate sampling
Slag aggregate	2	84	118	151	114	148	182	121	141	160	SSI	Not adequate sampling
Insulation materials wool of stone or glass	3	11	13	15	15	15	15	160	230	300	SSI	Not adequate sampling

Table II. The concentration of radium-226 in samples of aerated concrete based on alum shale in various factories and the amounts of such concrete produced until 1970.

Factory	Operating period	Produced amount m^3	Type of product	No of samples	Radium-226 Bq/kg[1]		
						1953	1973
Borensberg	1936 - 1968	$0.90 \cdot 10^6$	inner-walls	6	aver	1200	-
					min	1100	-
					max	1350	-
Yxhult N:a	1929 - 1959	0.57	reinforced, floor structures	5	aver	1600	-
					min	1500	-
					max	1750	-
Yxhult S:a	1947 - 1975	3.7	reinforced, floor structures	5 + 15	aver	1250	1300
					min	1100	1100
					max	1600	1950
Falköping	1930 - 1974	0.43	unreinforced products	5 + 17	aver	1900	2200
					min	1800	2100
					max	2100	2500
Uddagården	1955 - 1974	3.3	- " -	11	aver	-	2400
					min	-	2200
					max	-	2600
Grönhögen (Öland)	1943 - 1972	2.7		6	aver	650	-
					min	620	-
					max	700	-
Skövde/Durox	1929 - 1968	2.1	all types	1		-	1500

[1] Can be converted to pCi/g by multiplying by the factor 0.0270

Table III. Absorbed dose-rates from gamma radiation in Swedish houses. Averaged over all the 24 counties. Preliminary values.

Building material in outer walls	Number of houses measured	Absorbed dose rate in air mean, nGy/h[2)]
Wood	405	53
Brick	382	92
Concrete	221	115
Aerated concrete[1)]	181	170
Overall average	1 189	96

[1)] Mostly containing alum shale.

[2)] Can be converted to μrad/h by multiplying by the factor 0.1.

Table IV. The average radon concentrations in dwellings existing in 1950 and 1975 for an occupancy factor of 100 per cent.

Houses existing in	Apartment houses Bq/m^3	Detached houses Bq/m^3	Averages Bq/m^3
1950	43	18	29
1975	89	48	71

1 Bq/m^3 $\approx$ 0.027 pCi/litre

Fig 1. Map of Sweden. The mean absorbed tissue dose-rates from gamma radiation in the 24 counties of Sweden are shown in mGy per year. The estimated contribution from cosmic radiation has been subtracted. Preliminary values (4).

Fig 2. The dependence on the wind direction of the radon concentration in a detached house with a natural draught ventilation system. The door to the room was closed at the beginning of the measurement.

Fig 3. The variation of the radon concentration in a dwelling in an apartment house built of elements of both ordinary concrete and aerated concrete based on alum shale and with forced ventilation system for the exhaust air. The arrow shows when a secondary door from an adjacent room was opened after having been closed for 9 hours.

Fig 4. The variation of the radon concentration in two single-family houses built in different ways. No 121 had a cellar where the detector was placed. No 123 was built on a crawl space ventilated by four small ducts. The detector was placed in a room.

Fig 5. The variation of the radon concentration in a single-family house with a natural draught ventilation system. Sealing of two windows in a room was carried out on the 30th October. In this case the ventilation rate has little dependence on the wind direction.

Fig 6. The variation of the radon concentration in a room in a multi-family house, in which the fans were stopped each night from 12 pm to 6 am. The house had a mechanical exhaust ventilation system.

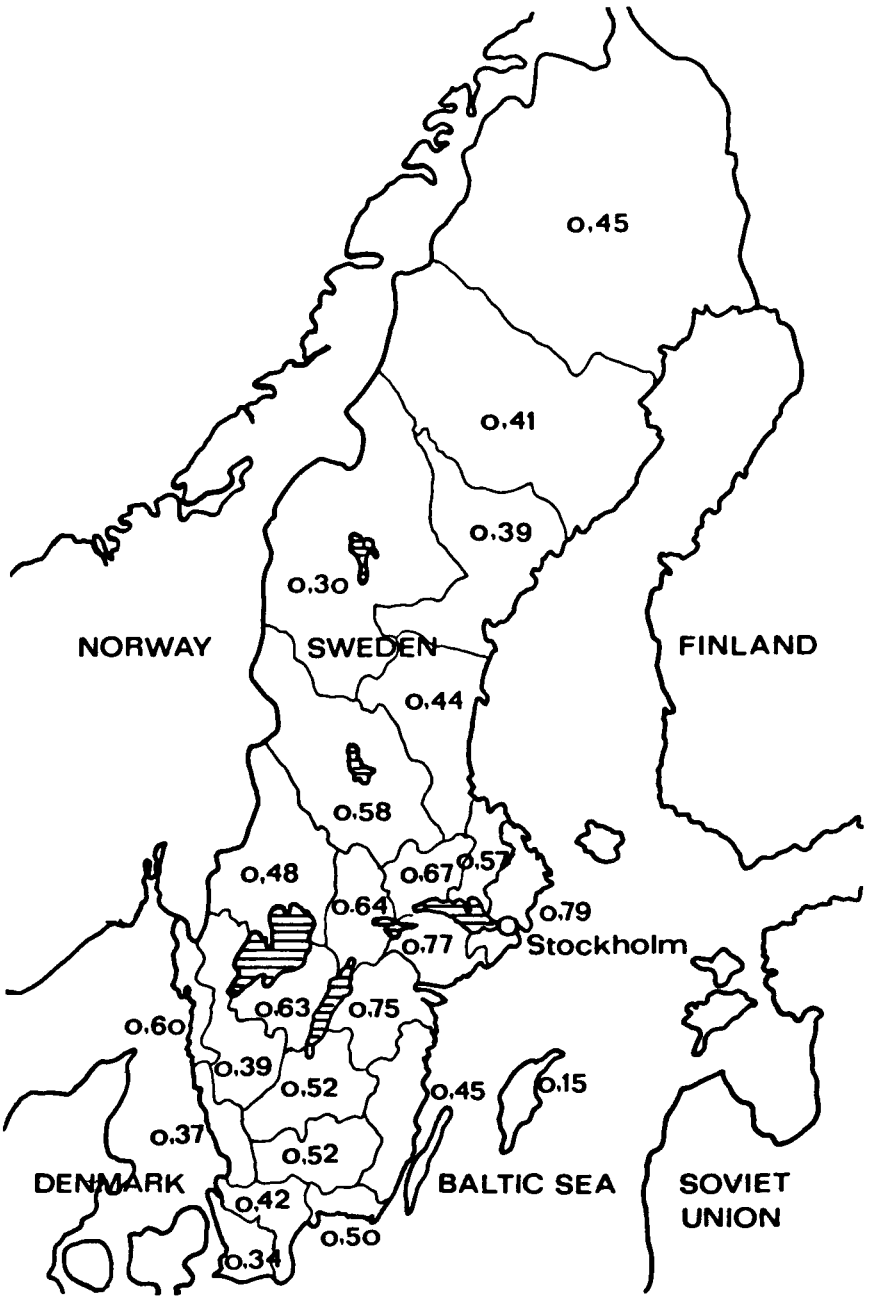
0.45
0.41
0.39
0.30
NORWAY
SWEDEN
FINLAND
0.44
0.58
0.48
0.67
0.57
0.64
0.79
0.77
Stockholm
0.63
0.75
0.60
0.39
0.52
0.45
0.15
0.37
0.52
DENMARK
BALTIC SEA
SOVIET
UNION
0.42
0.50
0.34

FIG. 1

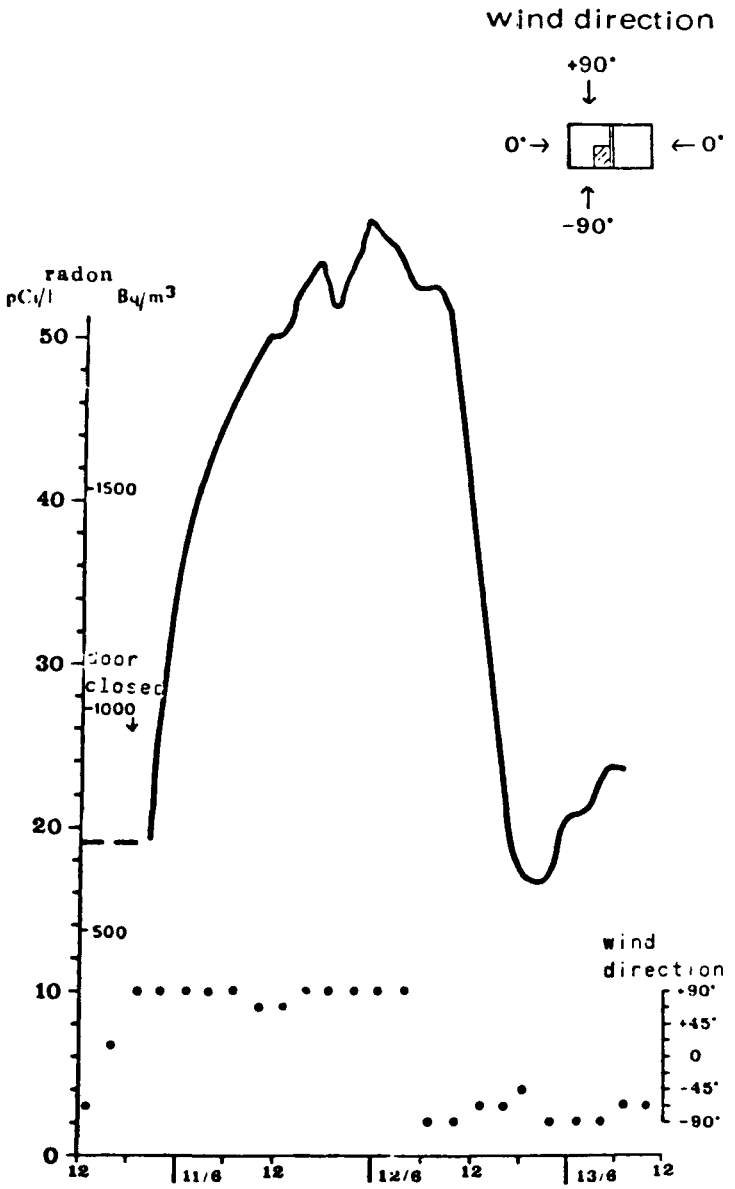
wind direction
+90°
0°→
←0°
-90°
radon
pCi/l
Bq/m³
50
40
30
20
10
0
1500
door
closed
1000
500
wind
direction
+90°
+45°
0
-45°
-90°
12
11/6
12
12/6
12
13/6
12

FIG. 2

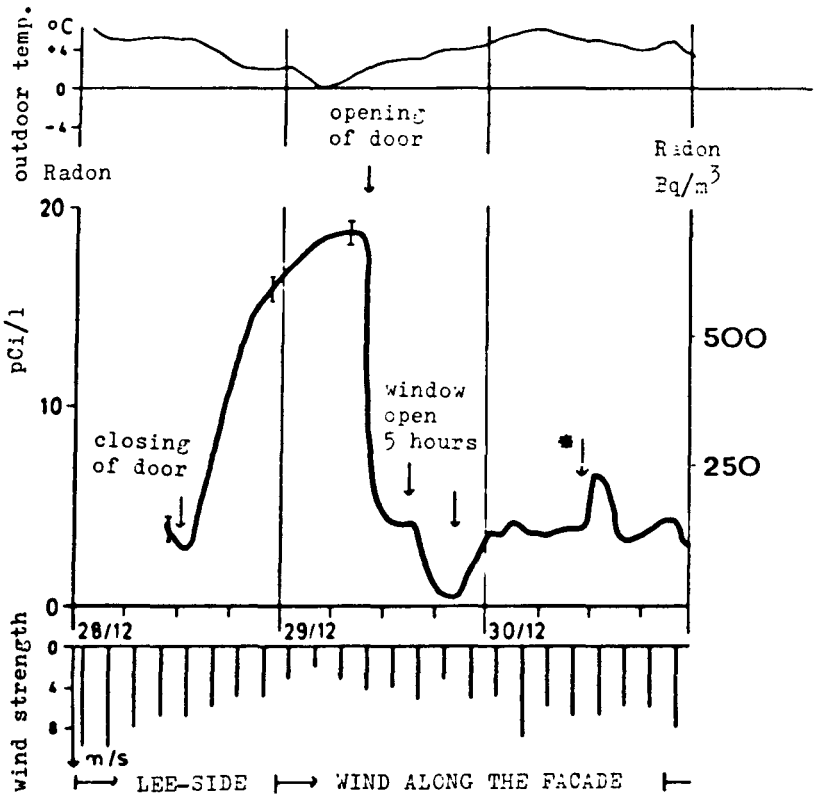
outdoor temp.
°C
4
0
-4
opening
of door
Radon
Radon
Bq/m3
20
pCi/l
10
0
500
250
closing
of door
window
open
5 hours
28/12
29/12
30/12
wind strength
0
4
8
m/s
LEE-SIDE
WIND ALONG THE FACADE

FIG. 3

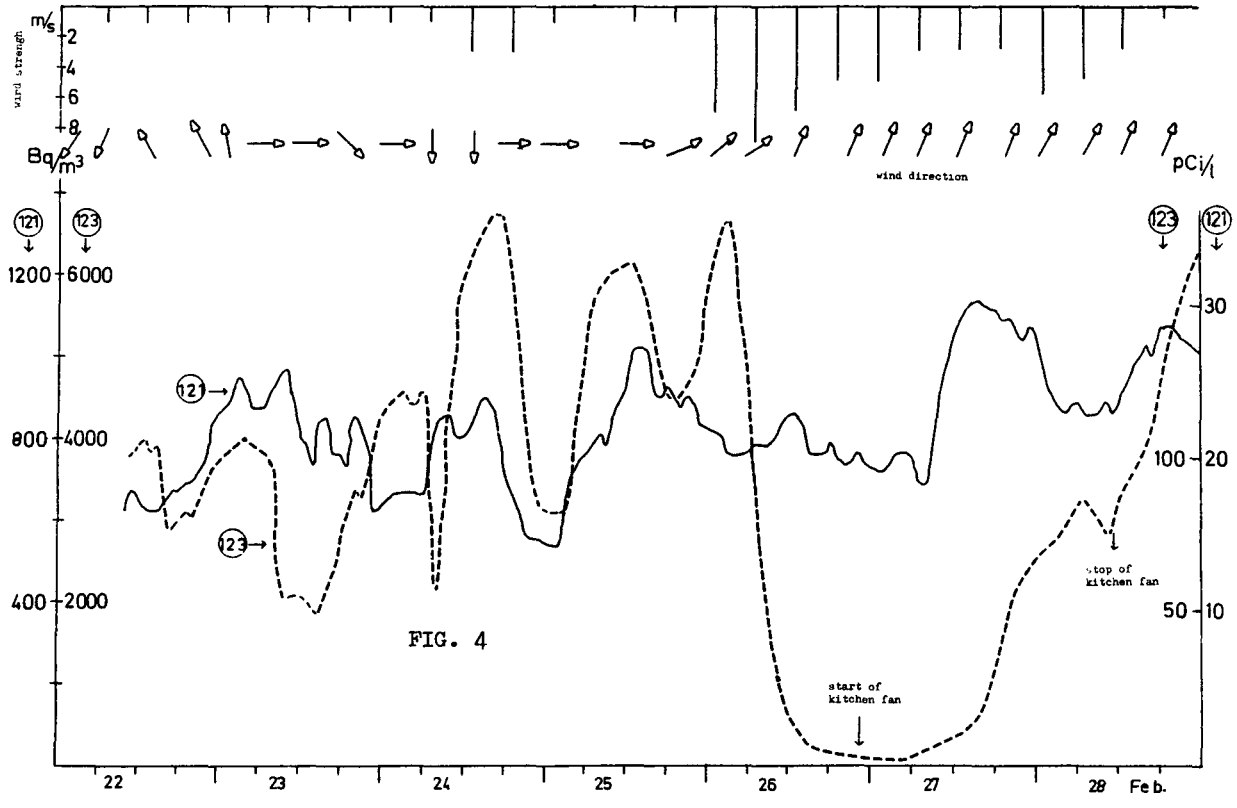
wind strength
m/s
2
4
6
8
wind direction
Bq/m3
pCi/l
121
123
1200
6000
800
4000
400
2000
30
20
100
10
50
top of kitchen fan
start of kitchen fan
22
23
24
25
26
27
28
Feb.

FIG. 4

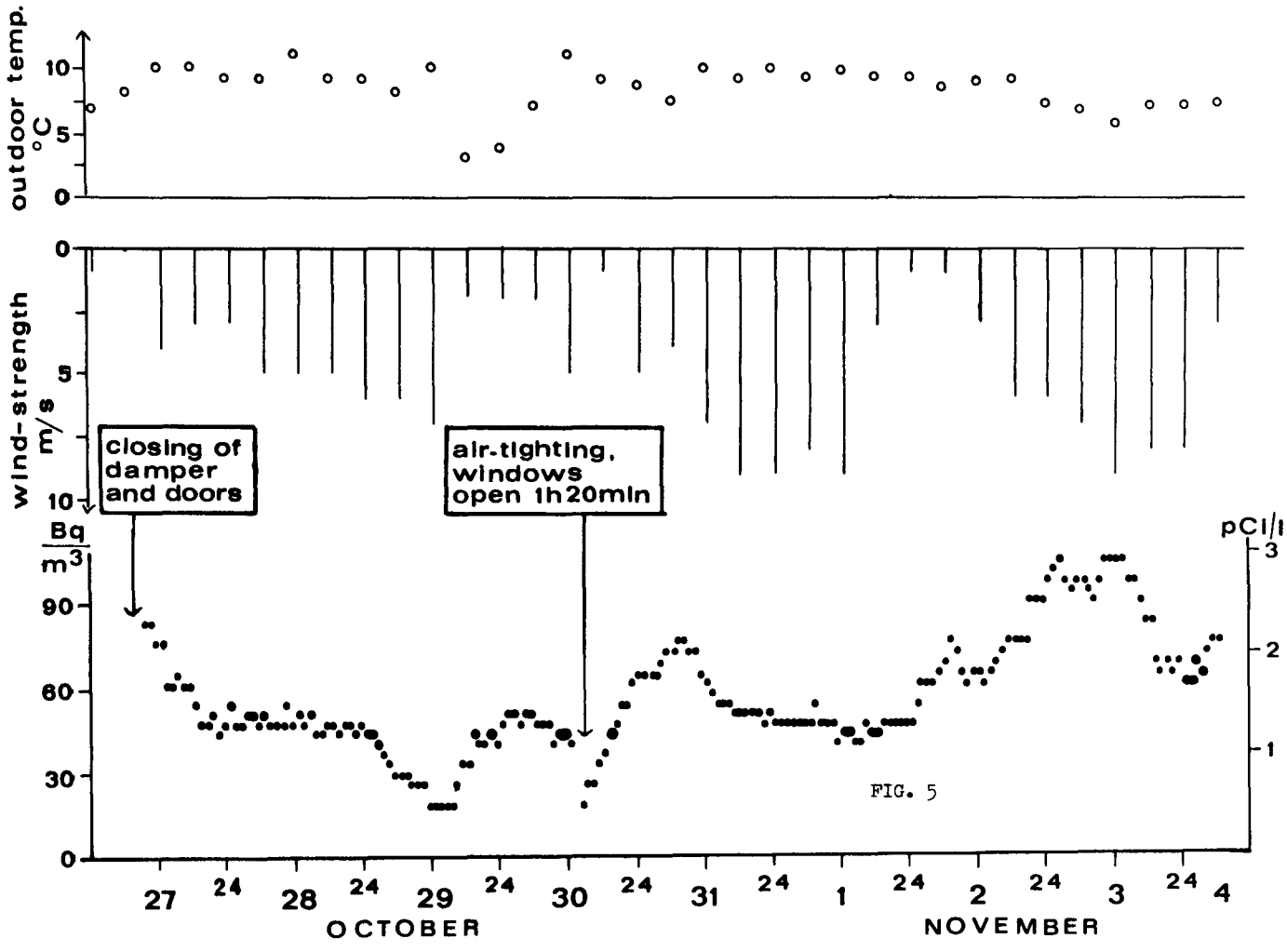
outdoor temp.
°C
10
5
0
wind-strength
m/s
0
5
10
closing of
damper
and doors
air-tighting,
windows
open 1h20min
Bq
m3
pCi/l
90
60
30
0
3
2
1
27
24
28
24
29
24
30
24
31
24
1
24
2
24
3
24
4
OCTOBER
NOVEMBER

FIG. 5

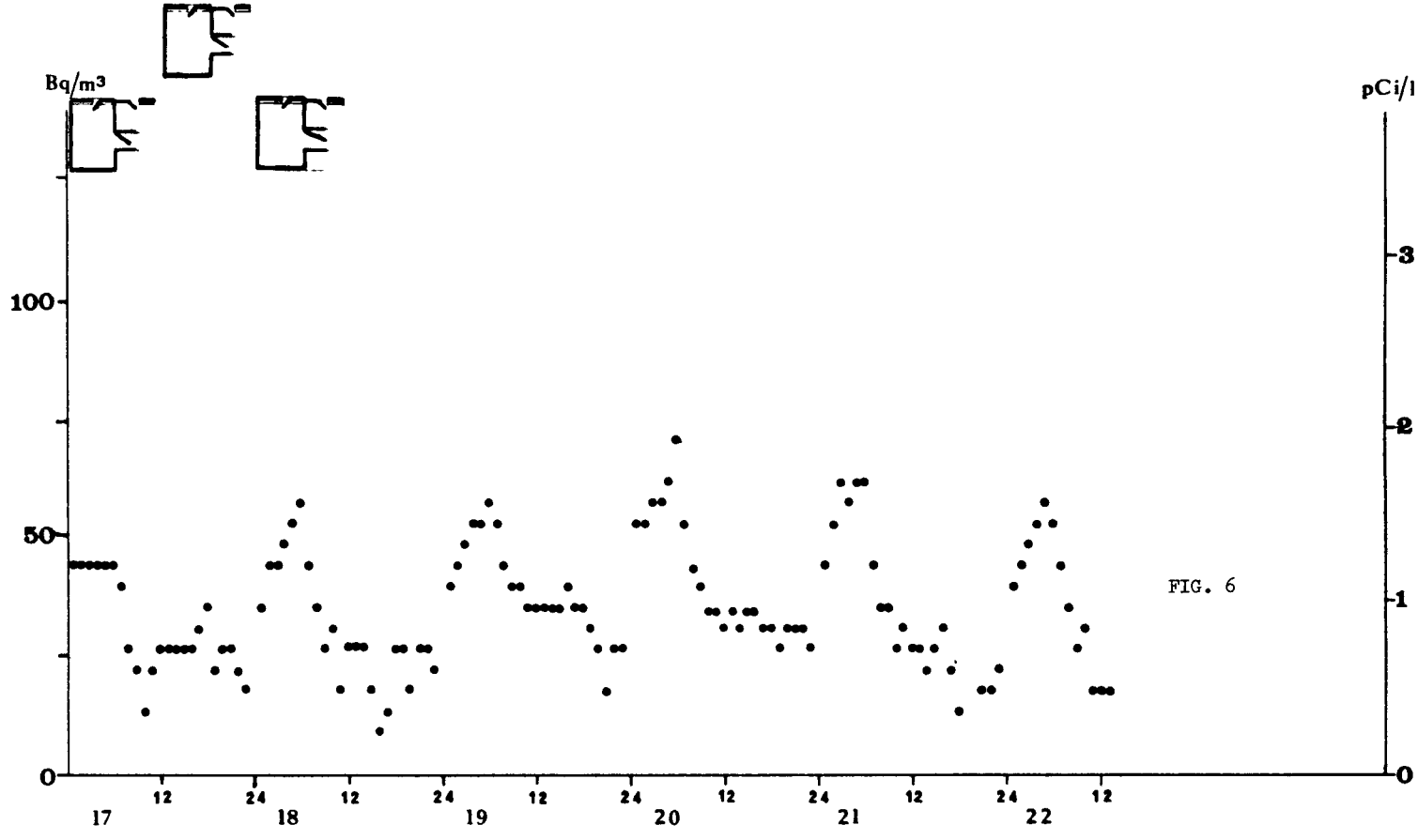

FIG. 6

INDOOR LEVELS OF NATURAL RADIATION IN THE UNITED KINGDOM

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SUMMARY. Of all body tissues that receiving the highest radiation dose from natural radioactivity is undoubtedly the bronchial epithelium. This dose is due predominantly to the short-lived daugthers of radon-222 with a lesser contribution from the daughters of radon-220. Details of measured concentrations of the daughters of radon-222 and radon-220 in domestic premises in the United Kingdom will be presented together with an assessment of current population exposure. The implications of the current exposure to radon daughter products and the effect of reduced ventilation in increasing this exposure will be discussed. Effective methods of reducing radon daughter concentrations within buildings other than by increased ventilation will also be outlined.

RESUME. NIVEAUX DE RAYONNEMENT NATUREL DANS LES BATIMENTS AU ROYAUME-UNI. De tous les tissus du corps l'épithélium bronchique est sans conteste celui qui reçoit la plus haute dose d'irradiation liée à la radioactivité naturelle. Cette dose est due surtout aux descendants à vie courte du radon-222, dans une moindre mesure, à ceux du radon-220. Le rapport donne des détails sur les concentrations des descendants du radon-222 et du radon-220 mesurées dans les locaux domestiques au Royaume-Uni ainsi qu'une évaluation de l'exposition actuelle de la population. On examine les conséquences de cette exposition aux descendants du radon ainsi que l'accroissement de cette exposition lié à la réduction des débits de ventilation. Des méthodes efficaces pour réduire les concentrations des descendants du radon dans les bâtiments, sans augmenter les débits de ventilation, sont aussi exposées.

KURZFASSUNG. DER PEGEL DER NATÜRLICHEN STRAHLUNG IN WOHNUNGEN IM VEREINIGTEN KÖNIGREICH. Von allen Körpergeweben empfängt zweifellos das Bronchialepithel die höchste Strahlendosis aus natürlicher Radioaktivität. Zu dieser Dosis tragen vor allem die kurzlebigen Folgeprodukte des Radon-222 und in geringerem Umfang auch die Folgeprodukte des Radon-220 bei. Eine detaillierte Aufstellung der im Vereinigten Königreich in Wohnhäusern gemessenen Konzentrationen der Folgeprodukte des Radon-222 und Radon-220 sowie eine Beurteilung der gegenwärtigen Exposition der

Bevölkerung werden vorgelegt. Es wird erörtert, wie sich die derzeitige Strahlenbelastung durch die Folgeprodukte des Radons auswirkt und inwieweit eine verringerte Belüftung zu einer Erhöhung dieser Exposition beiträgt. Wirksame Methoden, die ausser einer intensiveren Belüftung angewandt werden können, um die Konzentrationen der Radon-Folgeprodukte in Gebäuden zu verringern, werden ebenfalls behandelt.

Introduction

Although no recent extensive surveys of indoor γ-ray and cosmic-ray exposures have been undertaken in the United Kingdom the highest radiation dose to the body tissues from natural radioactivity is undoubtedly that to the bronchial epithelium. This arises predominantly from the short-lived daughters of ^{222}Rn with a lesser contribution from the daughters of ^{220}Rn. This paper will discuss the estimation of current United Kingdom population exposure to the short-lived daughters of ^{222}Rn and the daughters of ^{220}Rn (mainly ^{212}Pb).

Following common usage in the epidemiological studies of ^{222}Rn daughter exposures in miners the unit of exposure used will be the Working Level Month (WLM) and exposure rate the Working Level (WL). The WL is defined as any combination of the short-lived daughters of ^{222}Rn (^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po) in one metre^3 of air which will result in the ultimate release of 1.3×10^8 MeV of alpha energy in decaying to ^{210}Pb. The Working Month is taken as 170 hours. Exposure is then found as $\frac{\text{WL} \times \text{period of exposure in hours}}{170}$. Although the Working Level concept was introduced for the short-lived daughters of ^{222}Rn it can be extended to the daughter products of ^{220}Rn. However, the usefulness of the WL in the context of ^{220}Rn daughter exposure is questionable.

Method of measurement of radon daughters

In a survey of radon daughter concentrations in dwellings in England and Scotland an instrument based on the Radon Daughter Monitor (RDM) design of James and Strong[1] was used with the counting regime described by Cliff[2], [3] which permitted assessment of environmental ^{218}Po concentrations with adequate precision using readily portable equipment. In that survey measurements were made in living rooms of dwellings which had been closed for a period of at least 2½ hours. Measurements were made of ^{218}Po concentrations in the room air and in the open air outside the room and the ventilation rate of the room was measured using a tracer gas technique[4]. In this survey it was assumed that the production rate of ^{222}Rn into the room in terms of Bq per metre^3 of room air per hour is temporally invariant and hence the ^{222}Rn production rate could be determined from the measurements outlined above.

The ^{222}Rn production rate, K, of the room is given by[5]:-

$$K = j\ (1 + 0.0734j)\ (C_A - C^1_A)\ \text{Bq m}^{-3}\ \text{h}^{-1}$$

where j is the ventilation rate in room changes per hour, h^{-1}

C_A is the ^{218}Po concentration in room air Bq m^{-3}

C^1_A is the ^{218}Po concentration in outside air Bq m^{-3}

To ensure that ventilation occurred predominantly between the room under investigation and outside air (rather than between adjoining rooms) rooms on the windward side of dwellings were chosen for study.

Subsequent to the original survey a limited number of measurements of ^{222}Rn and ^{220}Rn daughter concentrations have been carried out. The instrument used was again the RDM but using a five count regime[6] similar to that of Harley and Pasternak[7]. The assessment of ^{222}Rn and ^{220}Rn daughter concentrations were made from the same sample thus allowing the ratio of the relative Working Levels from ^{222}Rn daughters (WL_{Rn}) and ^{220}Rn daughters (WL_{Th}) to be calculated.

Results

Figure I shows the distribution of ^{222}Rn production rates found in the original survey; full details of which were given by Cliff[5]. A constant production rate of ^{222}Rn was assumed as this allows a simple model for the behaviour of ^{222}Rn daughters in room air to be used. However the ^{222}Rn production rate is influenced by changes in meteorological parameters such as atmospheric pressure but the variations found in ^{222}Rn production rate are much smaller than are the variations found in intermittent measurements of the ^{222}Rn concentration in room air. The mean production rate calculated from the results of all the dwellings surveyed is believed to be representative of the true mean production rate. The range of ^{222}Rn production rates varied from 0.44 to 204 Bq m^{-3} h^{-1} and the distribution was normal under a logarithmic transformation, i.e. a log-normal distribution. The highest value of 204 Bq m^{-3} h^{-1} was a factor of 2 higher than any other measured value and occurred in a dwelling constructed from local granite with a high ^{226}Ra concentration (> 100 Bq Kg^{-1}) and in an area of low population density. For the purpose of estimating population exposure this highest value was excluded and with this result eliminated the arithmetic mean production rate was found to be 20.0 Bq m^{-3} h^{-1}.

To assess the population exposure to the short-lived daughters of ^{222}Rn from the mean ^{222}Rn production rate a value had to be assumed for the yearly average ventilation rate. A mean ventilation rate of one room change per hour was chosen as representative of present conditions in the United Kingdom after consultation with the Building Research Station of the

UK Department of the Environment. (A limited survey of ventilation rates in closed living rooms by Warren[8] found a mean value of $0.8h^{-1}$ but when the smallest window in the room was opened to its first fixable position the ventilation rate increased typically by a factor of 4.) Assuming a ventilation rate of $1.0h^{-1}$, a mean ^{222}Rn production rate of 20.0 Bq $m^{-3}h^{-1}$ and that ^{222}Rn daughter products are lost from room air solely by radioactive decay and ventilation to outside air containing ^{222}Rn at the mean concentration found in the survey of 2.6 Bq m^{-3} then the mean Working Levels in living rooms is 0.0035 WL. Assuming an occupancy factor of 0.8 and including the exposure received in the 20% spent in outside air the average UK population exposure rate is 0.15 WLM y^{-1}.

To determine the ^{222}Rn output of the room tested, doors and windows of the room were shut after installation of the equipment. A period of 2½ hours was then allowed before measurements of ^{222}Rn daughter concentrations were taken that were used in the ^{222}Rn output assessment. However, measurements of ^{222}Rn daughter concentrations were made immediately following the installation of the equipment to give some indication of the concentrations likely to be measured later in the day. These measurements taken just after the equipment was installed might be taken as representative of actual conditions in dwellings in the early morning (these measurements were made between 0830 and 1030 hours). The distribution of Working Levels found in living rooms upon arrival (or shortly afterwards) is shown in Figure II. In this case the mean value of WL found was 0.00296 with a range of 1.9×10^{-5} to 0.0296 WL and the distribution again is approximately normal under a logarithmic transformation. It should be noted that the extreme value of these measurements (0.0296 WL) was not found in the same dwelling as the highest ^{222}Rn output of 204 Bq m^{-3} h^{-1}. Thus the mean WL found in the early morning is some 16% lower than that calculated from ^{222}Rn output assessment. This lower figure can be explained in part by the propensity of British housewives to open windows during the morning and that 50% of the measurements were made during the summer of 1976 which was an unusually dry and warm year. It is nevertheless gratifying that by either approach a similar mean WL value for living rooms is obtained. This leads to the conclusion that the assessed average population exposure to the short-lived daughters of ^{222}Rn of 0.15 WLM y^{-1} for the population of the UK is a reasonable working figure and agrees with theoretical predictions for single-family masonry houses[9].

In determining the representative exposure in single-family masonry dwellings a substantial contribution to the ^{222}Rn concentrations in dwell-

ings is shown to be the ^{226}Ra concentration of the subsoil[9]. This point has been confirmed by the difficulty in accounting for the ^{222}Rn daughter concentrations in rooms on the basis of the ^{226}Ra concentration of the building elements - in ground floor rooms a much higher ^{222}Rn daughter concentration is generally measured than can be supported by the ^{226}Ra concentration in the construction elements[10]. It might appear from the foregoing that in multi-storey flats occupants on higher floors will receive a lower ^{222}Rn daughter exposure than that received by occupants of the ground floor or single-family dwellings. In the original survey[5] the only first floor flats studied were in converted single-family dwellings and were not purpose built as flats. Recently ^{222}Rn daughter concentrations were carried out in the basement and then identical rooms on different floors of an eighteen storey hall of residence at a British University. These measurements were made during the summer vacation when the rooms had been closed and unoccupied for several weeks. The results of these measurements are given in Table I and with the exception of the room on the third floor the measured ventilation rate is given. Table I demonstrates that the expected reduction in ^{222}Rn daughter concentration in rooms remote from the ground is not observed (at least in this particular building). The variation in WL values in the various rooms is partly explained by the variation in ventilation rates but this can not explain the high values found on floors 6 and 14. In this building, common services (water pipes) were carried up the building in a service duct which linked all rooms. This duct was enclosed by hard-board panelling which was not efficiently sealed. It is postulated that a source of radon in each room was this communal service duct allowing radon from the lower floors to enter rooms at higher elevations. Further work is in hand to measure the ^{222}Rn flux from walls within the rooms and from cracks in the service ducting. The form of construction of this building in which service ducts link each floor is not uncommon in other multi-storey dwellings and it is unlikely that the exposure of occupants of flats on the higher floors of multi-family dwellings in the UK to the daughters of ^{222}Rn is significantly less than that of occupants of single family dwellings.

In a very limited number (8) of rooms measurements of ^{220}Rn and ^{222}Rn daughters were made from the same sample. The results of these measurements are given in Table II. The relative WL contributions from ^{222}Rn and ^{220}Rn daughters varies widely and no firm conclusion can be drawn from such a limited survey. However, in general the WL contribution from ^{220}Rn daughters is less than that from the daughters of ^{222}Rn. It should be noted

that 1 WL is given by 3.7 kBq m^{-3} of ^{222}Rn daughters in equilibrium compared with only 0.28 kBq m^{-3} of ^{220}Rn daughters (^{212}Pb, ^{212}Bi). Obviously more work is required to determine the ^{220}Rn daughter concentrations in rooms but if one assumes that the ^{220}Rn daughter WL is 20% of that due to ^{222}Rn then the mean annual exposure rate to ^{220}Rn daughters for the UK population is about 0.03 WLM/y. Various authors have derived conversion factors for dose per WLM from radon daughters [11,12,13,14,15]. The conversion factors so derived have varied by nearly two orders of magnitude for the daughters of ^{222}Rn depending upon the lung model used, assumed aerosol concentration in the atomosphere and assumed breathing rates. As the epidemiological data relating excess incidence of lung cancer to ^{222}Rn daughter exposures is based on cummulative exposures expressed in WLM it is preferable to give population exposures from environmental ^{222}Rn daughters in the same units. However, Jacobi[16] has calculated doses per WLM for both ^{222}Rn daughters and ^{220}Rn daughters. In the tracheo-bronchial region he finds tne dose per WLM from ^{222}Rn daughters to be between 15 and 40 times that per WLM from ^{220}Rn daughters. The concentration of ^{222}Rn daughters in room air is more influenced by ventilation rate (increasing with decreasing rate) than is the concentration of ^{220}Rn daughters[17]. In view of these considerations it is apparent that the daughters of ^{222}Rn in the environment within buildings are the source of most concern with regard to the radiation dose to the bronchial epithelium of members of the general population.

Possible consequences of changes in ventilation rate

The dramatic increase in energy costs over the past few years has encouraged energy conservation in buildings and one effective measure is to reduce ventilation rates. The reduction of ventilation rate, in the absence of any compensating action, will increase the ^{222}Rn daughter concentration within buildings and hence increase the population exposure from this source. In the UK, energy conservation is unlikely to affect ventilation rates during the summer months and assuming that the mean ventilation rate during the five summer months is 2 air changes per hour and does not change it is possible to calculate the increased exposure rate as the mean winter (7 month) ventilation rate is reduced. Also in reviewing the epidemiological data from the uranium and fluorspar miner studies on excess lung cancer incidence and exposure to ^{222}Rn daughters Jacobi[18] has arrived at a risk estimate of 200 excess lung cancers per 10^6 miners per WLM. Hofman et al[19] and James[20] have indicated that the dose per

WLM to a member of the adult population is about 0.5 of that to a working miner. Apart from relative dose per WLM to miners and members of the general population other factors such as different aerosol size distributions in the differing environments will influence the relative risk to the two groups. The National Radiological Protection Board sought the advice of the Medical Research Council regarding the level of risk for members of the population exposed to environmental ^{222}Rn daughters and was advised that based on existing evidence an appropriate value for the induction of fatal lung cancer is 10^{-4} per WLM. Using this risk estimate, the implied increase in lung cancer incidence as the mean winter ventilation rate is reduced can be calculated and these results are shown in Table III. Although houses have been designed with ventilation rates as low as 0.1 h^{-1} it is doubtful whether in practice such a low ventilation rate would be maintained for long periods during the winter months. A realistic lower limit to the mean winter ventilation rate is 0.2 h^{-1} and at this value the predicted incidence of lung cancer due to environmental ^{222}Rn daughter exposure of about 60 per million population per year is approaching 10% of the current total incidence of lung cancer in the UK (650 per million per year), assuming that the whole population adopted decreased ventilation rate.

Method of removing radon daughters

As the above assumptions, which may or may not be well founded, lead to the prediction of a possible substantial increase in the lung cancer incidence as ventilation rates are reduced methods of removal of ^{222}Rn daughters from room air are being investigated. A recent investigation by Miles et al[21] tested the effect of various air treatment devices on the ^{222}Rn daughter concentrations within a room. Measurements were carried out in an environmental chamber of volume 33 m^3 with a ventilation rate of 0.05 h^{-1}. The radon concentration within the chamber was enhanced by removing the ^{222}Rn from a ^{226}Ra salt solution by aeration. Initially the ^{222}Rn concentration was increased and measurements made periodically over a period of 4 hours of WL value and condensation nucleus concentration. The air was continuously mixed using domestic air fans. The results of these measurements are shown in Figure III. The condensation nucleus concentration decreased with time due to wall attachment and sedimentation. The WL values also decreased due partly to the loss of ^{222}Rn daughters by ventilation and partly because the low condensation nucleus concentration resulted in a higher unattached fraction of ^{222}Rn daughters which were re-

moved by plate-out to the chamber surfaces. The removal of radon daughters was probably enhanced by plate-out on the air fan blades (a recently reported removal mechanism [22]). In subsequent experiments with the environmental chamber similar measurements were made but the condensation nucleus concentration was periodically increased by allowing a cigarette to smoulder. Three air treatment devices were used; a humidifier, a dehumidifier and an electrostatic precipitator. Neither the humidifier nor the dehumidifier had any significant effect on the WL value. The electrostatic precipitator, however, produced a dramatic reduction in both WL and condensation nucleus concentration as is shown in Figure IV. Under favourable conditions the precipitator produced a reduction in WL by a factor of 20. This reduction was due to direct removal of ^{222}Rn daughters by the precipitator and by increased removal by plate-out as the condensation nucleus concentration was reduced resulting in a higher fraction of unattached ^{222}Rn daughters.

Discussion

At present the exposure to the short-lived daughters of ^{222}Rn for an average member of the population of the UK is 0.15 WLM y^{-1} for whom the winter ventilation rate is 0.8 h^{-1}. This will increase to 0.58 WLM y^{-1} if the winter ventilation rate is reduced to 0.2 h^{-1}. One possible method of effectively reducing the ^{222}Rn daughter concentration and hence the exposure is to use an electrostatic precipitator air treatment unit. However, the use of such a device would increase the unattached fraction of ^{222}Rn daughter products which might result in an increased absorbed dose per WLM. The alpha-dose to the tracheo-bronchial region has been shown to be dependent upon the total unattached fraction, f_p, of ^{222}Rn daughters according to the relationship Absorbed Dose $\alpha(1 + 6\ f_p)$ [23].

If risk of lung cancer is proportional to absorbed dose then the use of the electrostatic precipitator would reduce population exposure in WLM y^{-1} but would also increase the risk per WLM compared with the risk derived from the miner studies. Further study is required before the balance of risk and benefit can be fully quantified.

In the light of present, admittedly scant, data on ^{220}Rn daughter products in room air these nuclides do not pose such a problem as the decay products of ^{222}Rn. Further work in this area is necessary to confirm this belief. Methods of reducing ^{222}Rn daughter concentration in buildings are being investigated. These methods include prevention of the ingress of ^{222}Rn from the subsoil and the coating of construction materials with

a "radon barrier" to prevent or reduce the emanation of radon from building materials, whether such practices could be justified by cost-benefit analysis remains to be seen. The reduction in emanation of radon from building materials would lead to a higher γ-ray exposure from retained daughters to the inhabitants of rooms and this effect needs to be studied.

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Table I

Concentration of ^{222}Rn daughter products in similar rooms on different floors of a multi-storey residential building.

Floor*	WL X10^{-3}	Ventilation Rate, h^{-1}
Basement	14.3	0.23
1	0.98	0.79
2	1.13	0.58
3	2.13	-
4	3.70	0.43
5	3.58	0.40
6	7.41	0.41
8	3.58	0.36
9	4.67	0.21
10	2.47	0.58
12	3.09	0.40
14	5.70	0.65

*The floor number is for floors above ground level, i.e. ground level would be floor 0.

Table II

^{222}Rn daughter and ^{220}Rn daughter concentration, measured from the same sample, in different rooms.

WL x 10^{-3}		$\frac{^{222}Rn\text{-}WL}{^{220}Rn\text{-}WL}$
^{222}Rn	^{220}Rn	
3.79	1.20	3.16
0.20	0.30	0.67
11.1	4.38	2.53
3.61	0.45	8.02
14.7	1.20	12.3
24.5	1.50	16.3
24.0	1.60	15.0
1.73	1.55	1.12

Table III

Predicted lung cancer incidence in the UK due to environmental ^{222}Rn daughter concentrations as the mean winter (7 months) ventilation rate is reduced: the summer (5 months) mean ventilation rate is assumed to be constant at 2 air changes per hour.

Winter Ventilation Rate h^{-1}	Mean population exposure WLM y^{-1}	Lung cancer incidence predicted per 10^6 population per year
0.8	0.15	15
0.5	0.22	22
0.4	0.28	28
0.3	0.38	38
0.2	0.58	58
0.1	1.15	115

CAPTIONS TO FIGURES

Figure I — Distribution of ^{222}Rn production rates in living rooms of dwellings in the UK.

Figure II — Distribution of WL values recorded in living rooms of dwellings in the UK under normal occupational conditions between 0830 and 1030 hours.

Figure III — Variation of WL and condensation nucleus concentrations in a sealed room without the operation of the electrostatic precipitator.

Figure IV — Variation of WL and condensation nucleus concentration in a sealed room demonstrating the effect of enhanced condensation nucleus concentrations by means of cigarette smoke and the effects of the electrostatic precipitator.

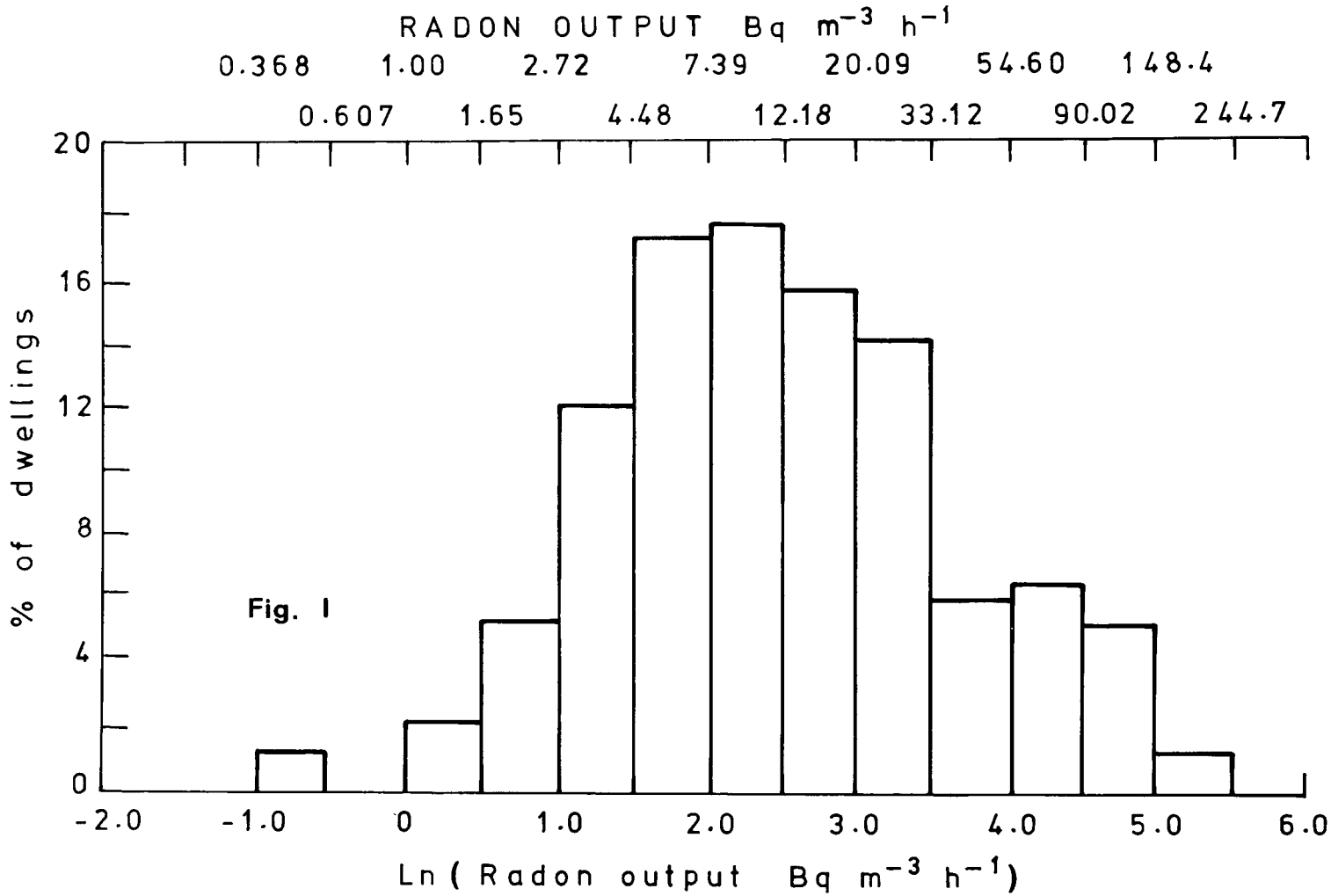
RADON OUTPUT Bq m^{-3} h^{-1}
0.368
1.00
2.72
7.39
20.09
54.60
148.4
0.607
1.65
4.48
12.18
33.12
90.02
244.7
20
16
12
8
4
0
% of dwellings
-2.0
-1.0
0
1.0
2.0
3.0
4.0
5.0
6.0
Ln (Radon output Bq m^{-3} h^{-1})

Fig. 1

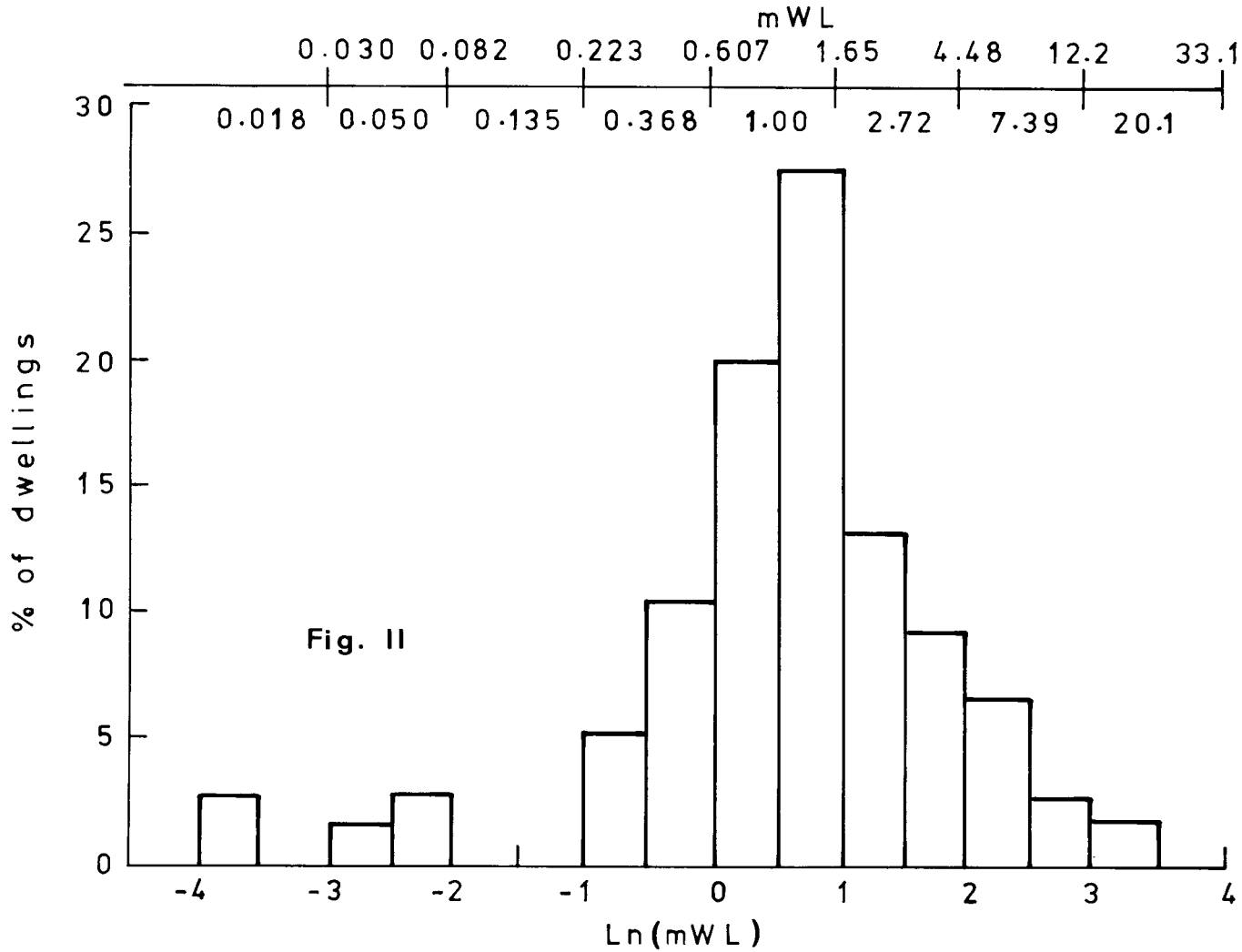
mWL
0.030 0.082 0.223 0.607 1.65 4.48 12.2 33.1
0.018 0.050 0.135 0.368 1.00 2.72 7.39 20.1
% of dwellings
0 5 10 15 20 25 30
-4 -3 -2 -1 0 1 2 3 4
Ln(mWL)

Fig. II

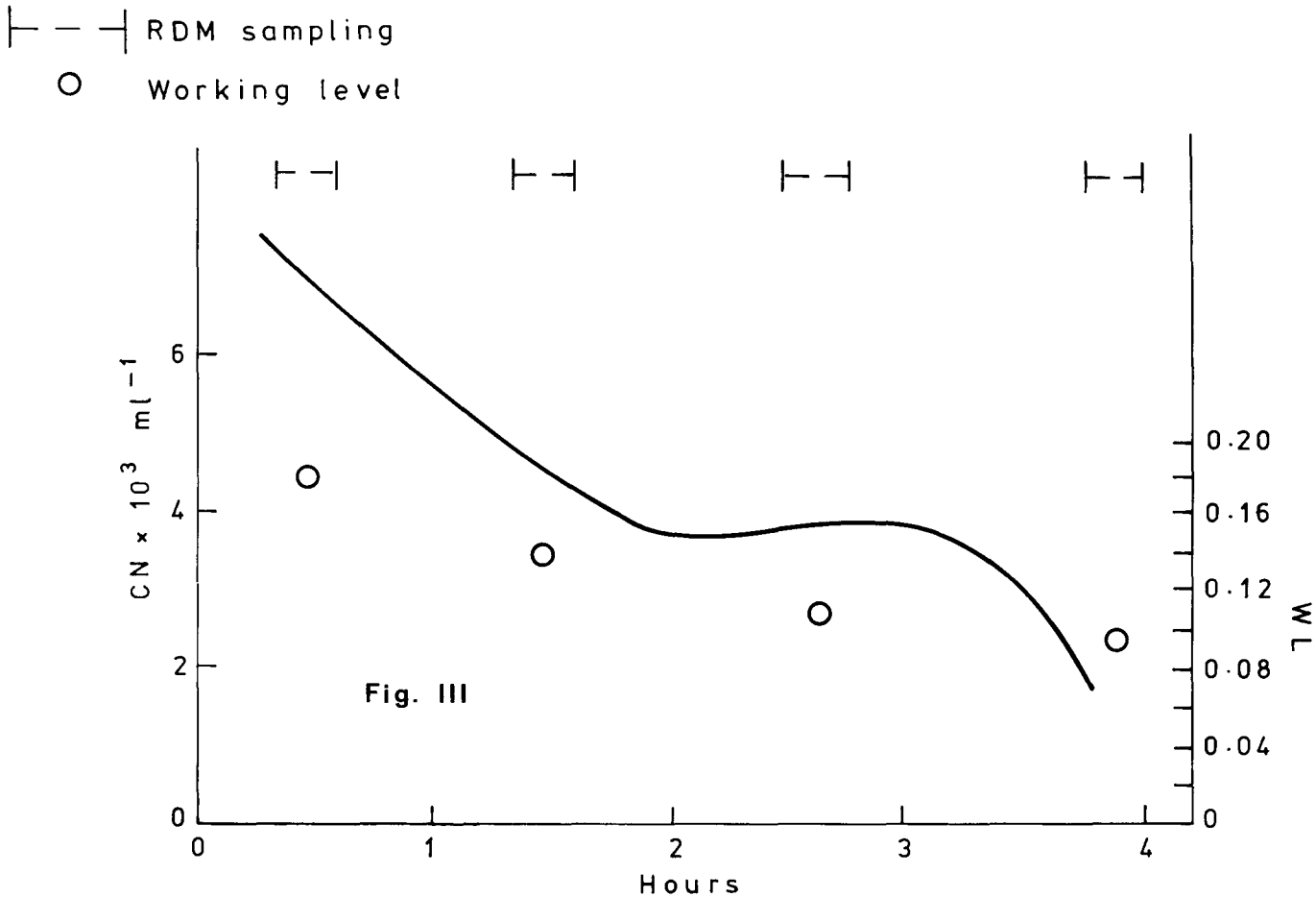
RDM sampling
Working level
CN × 10³ ml⁻¹
0
2
4
6
0
1
2
3
4
Hours
0·20
0·16
0·12
0·08
0·04
0
WL

Fig. III

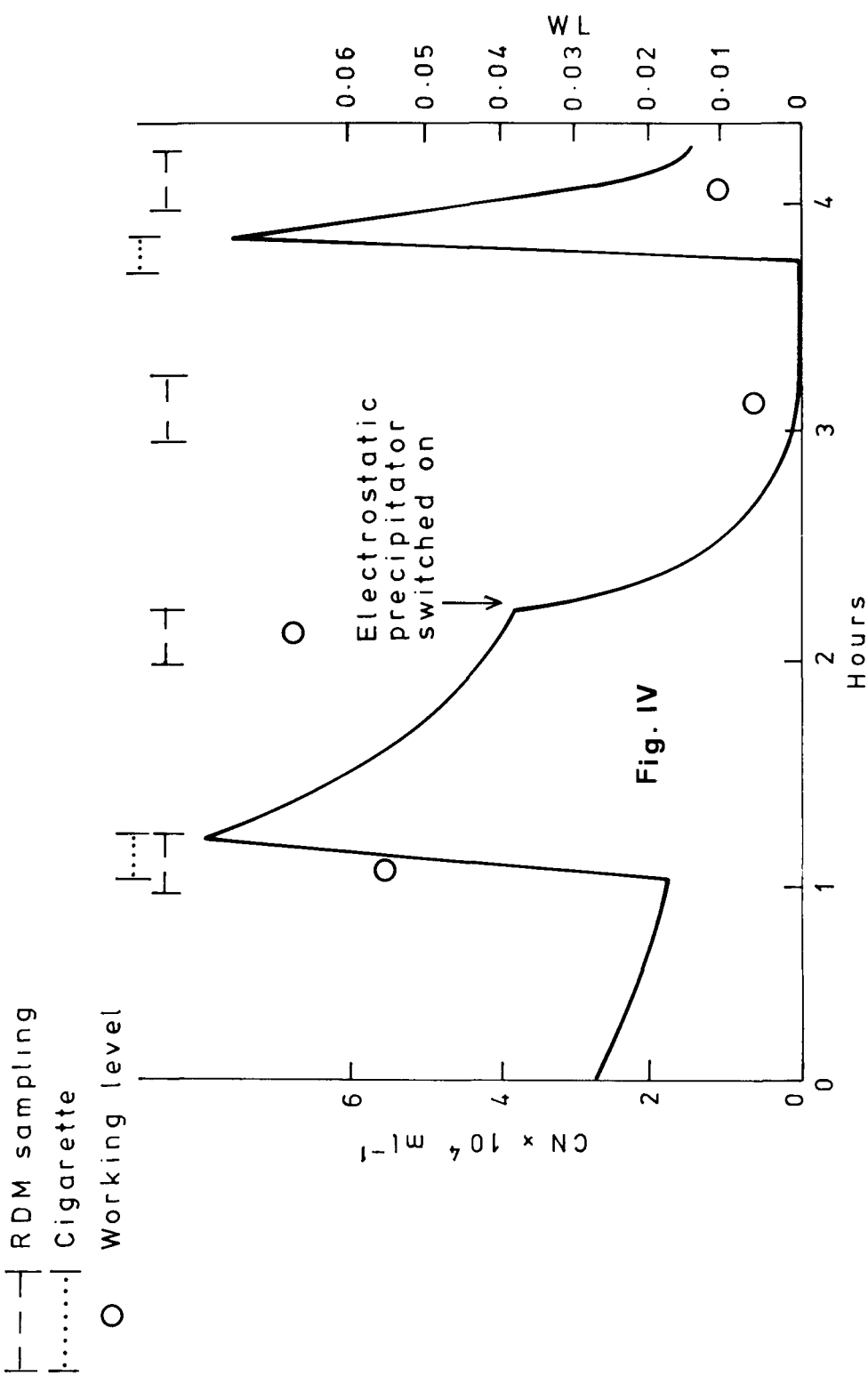
RDM sampling
Cigarette
Working level
CN × 10⁴ m⁻³
WL
Hours
Electrostatic precipitator switched on

Fig. IV

ON THE EVALUATION OF INGESTION DOSE FACTORS FOR NATURALLY OCCURRING RADIONUCLIDES

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SUMMARY. The ICRP has recently published its new recommendation on limits for the activity intake of radionuclides by workers (ICRP Publ. 30, 1979), which replaces ICRP Publication 2. These new metabolic and dosimetric models of ICRP, especially for the lung, the GI-tract and bone are briefly described with respect to their application to natural occurring radionuclides. As an alternative the dose factors for ingestion of natural radionuclides by members of the public can be directly estimated on the basis of the measured values of natural activity in food and body tissues. The resulting dose factors for adults, derived from this natural exposure data, are in some cases somewhat higher than those which are given by the ICRP for workers. This difference is probably mainly caused by the higher uptake factor from the GI-tract to blood for radioactive atoms which are bound to the organic material in foodstuffs. As examples dose factors for U-238, Ra-226, Pb-210 and Po-210 are given.
A special problem is the evaluation of the dose distribution within the respiratory tract by the inhalation of short-lived Rn-daughters. The range in which the resulting α-dose to the basal cells in the bronchial epithelium may lie and how this range depends on the size of the carrier particles and the velocity of ciliary transport is discussed. Mean values for the dose to these cells at risk are proposed.

RESUME. DETERMINATION DE FACTEURS DE DOSE POUR L'INGESTION DE RADIONUCLIDES NATURELS. Récemment la CIPR a publié de nouvelles recommandations de limites applicables à l'incorporation d'activité de radionucléides par les travailleurs exposés (CIPR Publ. 30, 1979) qui remplacent celles de la publication 2 de la CIPR. Les nouveaux modèles métaboliques et méthodes dosimétriques, notamment pour le poumon, le tractus gastro-intestinal et l'os, y sont décrits brièvement en vue de leur application aux radionucléides naturels.
Les facteurs de dose relatifs à l'ingestion de radionucléides naturels par la population peuvent aussi se déduire directement des mesures disponibles de la radioactivité naturelle présente dans les aliments et les tissus de l'organisme. Certains des facteurs de dose déduits de ces données sont légèrement plus élevés que ceux dérivés des modèles de la CIPR pour les travailleurs. Cette différence tient sans doute surtout au facteur

d'absorption plus élevé des atomes radioactifs mêlés à la matière organique des aliments lors de leur passage du tractus gastro-intestinal dans le sang. Les facteurs de dose de U-238, Ra-226, Pb-210 et Po-210 sont indiqués à titre d'exemple.
Le calcul de la distribution de la dose dans les voies respiratoires par suite de l'inhalation de produits de filiation à vie courte du radon pose un problème particulier. Le domaine de variation de la dose alpha qui en résulte dans les cellules basales de l'épithélium bronchique et son rapport avec la taille des particules porteuses et la vitesse d'entraînement ciliaire font l'objet d'un exposé. On propose des valeurs moyennes de facteurs de dose pour cette couche critique de cellules basales.

KURZFASSUNG. ERMITTLUNG VON DOSISFAKTOREN FÜR DIE AUFNAHME NATÜRLICHER RADIONUKLIDE MIT DER NAHRUNG. Die ICRP hat kürzlich ihre neuen Empfehlungen über die Grenzwerte der Aktivitätszufuhr von Radionukliden bei beruflich strahlenexponierten Personen publiziert (ICRP Publ. 30, 1979). Diese Empfehlungen lösen die ICRP-Publikation 2 ab. - Die neuen Stoffwechselmodelle und Methoden der Dosisberechnung, insbesondere für die Lunge, den Magen-Darm-Trakt und den Knochen werden, im Hinblick auf ihre Anwendung für natürliche Radionuklide, kurz beschrieben.

Alternativ können die Dosisfaktoren für die Ingestion natürlicher Radionuklide durch die Bevölkerung direkt aus den vorliegenden Messungen der natürlichen Radioaktivität in der Nahrung und in Körpergeweben abgeschätzt werden. Die aus diesen Daten ermittelten Dosisfaktoren sind teilweise etwas höher als diejenigen, die auf der Basis von der der ICRP-Modelle für Beschäftigte resultieren. Dieser Unterschied wird wahrscheinlich in erster Linie verursacht durch den höheren Aufnahmefaktor aus dem Magen-Darm-Trakt in das Blut für radioaktive Atome, die an das organische Material der Nahrung gebunden sind. Als Beispiele werden Dosisfaktoren für U-238, Ra-226, Pb-210 und Po-210 angegeben.
Ein spezielles Problem ist die Ermittlung der Dosisverteilung im Atemtrakt infolge der Inhalation kurzlebiger Radon-Zerfallsprodukte. Der Variationsbereich der resultierenden α-Dosis in den Basalzellen des Bronchialepithels und seine Abhängigkeit von der Grösse der Trägerpartikel und der Ciliartransportgeschwindigkeit werden diskutiert. Mittelwerte der Dosisfaktoren für diese kritische Basalzellschicht werden vorgeschlagen.

THE CONCEPT OF COMMITTED AND EFFECTIVE DOSE

The concepts for internal dosimetry and for the assessment of incorporation limits, which are applied in the old and new ICRP-recommendations, are principally different. This is outlined schematically in table 1.

Table 1: Comparison of concepts for the evaluation of dose factors for incorporated radionuclides

ICRP 2 (1959)	ICRP 30 (1979)
Constant Intake Rate over 50 years	Single or Protracted Intake
↓	↓
Equilibrium Activity in Tissues per Unit Intake Rate	50 y-Time Integral over Tissue Activities per Unit Intake
↓	↓
Dose Rate in Tissues per Unit Intake Rate (Chronical Exposure)	50 y-Committed Dose to Tissues per Unit Intake
↓	↓
Selection of a Critical Tissue for each Radionuclide	Effective Dose $H_E = \Sigma w_i H_i$ per Unit Intake

The old recommendations on internal dosimetry [1] started from the presumption of a chronical intake at a constant intake rate. Then the activity in body tissues was calculated, which is reached after an intake period of 50 years. In the next step the tissue dose rates at the end of this intake period were determined, which are proportional to the tissue activities at this time.

On the basis of this old ICRP-concept the dose factor for a tissue T is defined as the tissue dose rate $\dot{H}_T$ after 50 years of chronical intake divided through the assumed constant intake rate $\dot{I}$ during this 50 y-period:

$$h_T\ (\text{ICRP}-2) = \frac{\dot{H}_T\ (\text{after 50 years})}{\dot{I}\ (\text{chronical intake})} \qquad (1)$$

For the assessment of incorporation limits in ICRP 2 the reverse way was followed. For this purpose the basic values of the maximum permissible dose equivalent per year (MPH per y) were interpreted as maximum permissible

dose equivalent rates ($\dot{MPH}$) in these tissues:

(2) $$MPH \text{ per year} = \dot{MPH}$$

This means for example that the dose rate after 50 years of chronical intake should not exceed 5 rem/y in the gonads and red bone marrow, 15 rem/y in the lungs and 30 rem/y in bone (mean bone dose).

After selection of a "critical tissue" from the $\dot{MPH}$-value of this tissue the maximum permissible activity burden in the critical tissue ($MPTB_{\text{critical tissue}}$), the corresponding maximum permissible total body burden (MPBB) and finally the maximum permissible concentrations (MPC) in air and water were derived.

This "rate concept" of ICRP 2 has been often misunderstood and misused. It was often wrongly interpreted in such a way, that also for short-term exposure the values of MPTB, MPBB and MPC should not be exceeded.

The new basic recommendations of ICRP [2] point out that not the dose rate but the total dose to a tissue determines the risk of stochastic radiation effects in this tissue. With respect to incorporation of radionuclides the total dose equivalent H_T to a tissue T is defined as the time-integral over the dose equivalent rate $H_T(t)$ in this tissue after the intake of the radionuclide. This quantity which is proportional to the activity intake is called the "committed dose equivalent" to the considered tissue. ICRP recommends to use an integration period of 50 years for the evaluation of the committed dose.

ICRP defines therefore the dose factor for a tissue T as the committed dose equivalent H (in 50 years) in this tissue divided through the activity intake I to the human body [3]:

(3) $$h_T \text{ (ICRP 30)} = \frac{H_T \text{ (in 50 years)}}{I}$$

The first step in the evaluation of these dose factors concerns the calculation of the 50 y-time-integral of activity in the relevant tissues per unit intake which is equal to the total number of decays in these tissues in 50 years per unit of intake. By multiplication with the specific effective energies the (committed) dose factor for the considered target tissue can be obtained.

In addition the old concept of the "critical tissue" which has its origin in the historical threshold concept is replaced now by the "effective dose"-concept, which takes into account the dose to all irradiated tissues. Following the new ICRP-recommendations the effective dose equivalent H_E by an intake I of a radionuclide is defined by the equation

$$(4) \qquad H_E = \sum_T w_T H_T = \sum_T w_T \cdot h_T \cdot I$$

where w_T are the weighting factors for the tissues T recommended by ICRP (Publ. 26, 1977) and h_T the committed dose factors given by equation (3).

From equation (4) an effective dose factor

$$(5) \qquad h_E = \frac{H_E}{I} = \sum_T w_T h_T$$

can be derived which defines the effective dose equivalent in 50 years per unit of activity intake.

It should be noted that the weighting factors w_T recommended by ICRP were derived from the risk data given in the report of UNSCEAR [4] and represent mean values averaged over all ages and both sexes. Thus they can be applied also to members of the public which are chronically exposed to natural radionuclides during their whole life-time.

NEW DOSIMETRIC MODELS AND DOSE FACTORS RECOMMENDED BY ICRP

In its new recommendations on internal dosimetry [3] the ICRP proposes improved models for the distribution, retention and dosimetry of inhaled or ingested radionuclides, based on the experience obtained in the 20 years since the publication of ICRP 2 [1]. Of general importance are the new models for the respiratory tract and the gastrointestinal tract, which allow a more realistic estimate of the dose to these tissues and the activity transfer to other tissues.

Of special interest for the evaluation of dose factors for natural radionuclides in the U- and Th-decay chain are:

(1) The recommendation of a quality factor Q = 20 for α-radiation
(2) The new dosimetric model for bone

In the past Ra-226 was used as standard for bone-seeking radionuclides. The comparison with Ra-226 was made on the basis of the mean dose to total bone, which led to the introduction of a modifying factor $N \neq 1$ for the evaluation of the dose equivalent to bone for other radionuclides than Ra-226.

With respect to cancerogenesis the target tissues in the skeleton are the red bone marrow and the sensitive cells on bone surfaces, particularly the osteogenic cells on endosteal surfaces and certain, epithelial cells close to bone surfaces. On the basis of studies by SPIERS et al. [5, 6], WHITWELL et al. [7] and THORNE [8] the ICRP has derived now a dosimetric model for bone, which allows a direct estimation of the dose to the mentioned two target tissues (red bone marrow, bone surfaces) from the activity deposited in trabecular and cortical bone.

This model distinguishes two distribution classes of radionuclides in bone: Bone class V for radionuclides which are nearly uniformly distributed in the bone volume; and bone class S for radionuclides which are mainly deposited on bone surfaces. The association of natural radionuclides to these two bone classes is given in table 2 [3].

Table 2: Classification of natural radionuclides in bone

Emitter Uniform in Volume (Bone Class Y) 1)	Emitter on Bone Surfaces (Bone Class S)
U-238, 235, 234	U-237
-	Th-232, 230, 228
Ra-228, 226	Ra-224
Pb-210	Pb-212

1) Activity ratio Cortic./Trabec. Bone (Class V): 1 : 1 for Ra; 4 : 1 for other elements

It should be recognized that the long-lived Th-nuclides are deposited on bone surfaces, similar like Pu. On the other hand the long-lived nuclides of U, Ra and Pb are considered to be uniformly distributed in the bone volume.

As this model yields the dose to the two target tissues in bone no additional modifying factor N has to be inserted in the evaluation of the dose equivalent to these target tissues: N = 1 for all nuclides in bone.

It is obvious that the old and new dosimetric model for bone lead to different types and values of dose factors for bone. As an example, in table 3 the dose factors for ingestion of Ra-226 are compared which result from the old and new ICRP-model for bone. It should be pointed out that both models also proceed from different values of the quality factor for α's and the uptake factor f_1 from the GI-tract to blood.

Table 3: Dose equivalent to Bone Tissue per Bq ingested Ra-226

ICRP Recommendation	Target Tissue	Dose Factor for Target Tissue
Publ. 2 (1959) $Q_\alpha = 10$ $f_1 = 0.3$	Total Bone m = 5 kg	$\dot{\bar{H}}_{BONE}/\dot{I}_{ING} \approx 80 \cdot 10^{-6}$ Sv/Bq [1]
Publ. 30 (1979) $Q_\alpha = 20$ $f_1 = 0.2$	Bone surfaces m = 0.12 kg	$\bar{H}_{B,SURF.}/I_{ING} \approx 7.1 \cdot 10^{-6}$ Sv/Bq [2]

1) Dose rate after 50 y chronical intake per unit of ingestion rate
2) Committed Dose in 50 y per unit of ingested activity

From Table 3 follows that the new dose factor for bone surfaces by ingestion of Ra-226 is about a factor 10 lower than the dose factor for total bone derived from ICRP 2. Taking into account the dose factor for bone marrow and the dose to the soft tissues in the remaining body it results from the new ICRP-model for Ra-226 an effective dose factor for the adult reference man of

(6) $$h_E = 3.2 \cdot 10^{-7} \text{ Sv per Bq ingested Ra-226}$$

In the same way the effective dose factors for other natural radionuclides can be calculated on the basis of the new ICRP-recommendations for internal dosimetry [3]. In table 4 the expected tissue doses for ingestion of hexavalent compounds ($f_1 = 0.05$) and tetravalent vompounds ($f_1 = 0.002$) of U-238 and the resulting effective dose factor are given.

3)

Table 4: Committed tissue doses and effective dose per Bq ingested U-238 (occupational exposure)

Target Tissue (T)	w_T	H_T (10^{-8} Sv) per Bq	
		$f_1 = 0.002$	$f_1 = 0.05$
Bone Surfaces	0.03	4.0	100
Red Bone Marrow	0.12	0.3	7.5
Kidneys	0.06	1.7	41
ULI-wall	0.06	2	1
LLI-wall	0.06	5	3
Residual Body	0.67	ca. 0.02	ca. 0.5
Effective Dose 10^{-8} Sv per Bq		0.7	7.0

In figure 2 the effective dose factors for the ingestion of the relevant radionuclides of the U- and Th-decay seriesare summarized which follow from the new metabolic and dosimetric models recommended by ICRP [3]. It has to be emphasized that these values refer to the adult "reference man" and are recommended by ICRP for occupational exposure.

With respect to the exposure of members of the public these dose factors have to be corrected. Firstly the age-variation of tissue masses and retention parameters has to be taken into account. In addition the uptake factor f_1 from the GI-tract to blood may be different for natural radionuclides which are biologically incorporated in the diet.

For the estimation of the age-correction factor it has to be taken into account that a considerable fraction of the committed dose from these long-lived radionuclides is delivered a long time after the intake. This is demonstrated by figure 1 where the relative variation of the committed effective dose from ingested natural radionuclides as function of the time after intake is shown., normalized to the dose value for T = 50 years.

From this time distribution of the committed dose it can be followed that for chronical exposure to these long-lived nuclides the influence of age-dependent parameters on the life-time averaged, committed dose factor, which is the deciding quantity with respect to cancer risks, may not be substantial.

On the other side ingestion dose factors for members of the public can be directly derived from the comprehensive data about the natural activity of these radionuclides in the diet and in body tissues. In the following the results of such an analysis of the natural exposure data are summarized.

INGESTION DOSE FACTORS DERIVED FROM THE NATURAL EXPOSURE OF THE POPULATION

Data on the observed natural activity in food and the resulting specific activity in relevant tissues of the human body are summarized in several review papers, particularly in the report of UNSCEAR [4, 9].

The natural intake of U, Ra and Th results mainly by ingestion. Therefore from the estimated values for dietary intake rate $\dot{I}_{ing}$ of these nuclides and the resulting specific activities a_S in body tissues (source tissue S), mean values of the ratio $a_S/\dot{I}_{ing}$ can be obtained. Effective energies $\varepsilon_{T \leftarrow S}$ were calculated applying the methods described in the new ICRP-recommendations on internal dosimetry [3]; similar methods were used in the UNSCEAR-report [4].

In the tables 5 - 7 the mean ingestion dose factors

$$h_{T,ing} = \sum_S \varepsilon_{T \leftarrow S} \cdot \frac{m_S}{m_T} \cdot (a_S/\dot{I}_{ing}) \qquad (7)$$

for relevant target tissues T are listed, which are resulting from the observed natural exposure data for U-238, Ra-226 and Th-232. On the bottom of these tables the resulting values of the effective dose per Bq ingested are given, applying the weighting factors recommended by ICRP.

In a similar way ingestion dose factors for the population can be derived for other natural radionuclides. The evaluation of dose factors for the ingestion and inhalation of Pb-210 and Po-210 from natural exposure data was described in detail in a previous paper [10].

In figure 2 the effective dose factors for ingestion of nuclides of the U- and T-decay serie, derived from natural exposure data, are compared with those recommended by ICRP for workers [3].

Table 5: Mean Activity and Dose Rate per Unit of Ingestion Rate of U-238 with the diet derived from the natural exposure of the population ($\bar{I} \approx 6$ Bq/y $\hat{=}$ 500 µg/y)

Source Tissue (s)	$a_s/\dot{I}_{ing}$ Bq/kg per Bq/y	Target Tissue (T)	$\dot{H}_T/\dot{I}_{ing}$ 10^{-8} Sv per Bq
Bone	0.02	Bone Surface	500
Bone Marrow	0.0008	RBM	30 30 } 60
Kidneys	0.008	Kidneys	300
Residual Body	≈ 0.0008	Other Tissues	≈ 30
Effective Dose ca. $60 \cdot 10^{-8}$ Sv per Bq 1)			

1) ICRP 30 (1979): $H_E/I_{ing} = 7 \cdot 10^{-8}$ Sv/Bq ($f_1 = 0.05$)

Table 6: Dose Equivalent per Bq Ingested Ra-226, derived from the mean natural Ra-226-activity in body tissues (UN 1977)

$\bar{a}_{Bone} \approx 6$ Bq/ka, $\bar{a}_{soft\ tissue} \approx 0.1$ Bq/kg per Bq/d ingested

Target Tissue	Dose Equivalent (10^{-7} Sv) per Bq $H(Q_\alpha = 20)$	wH
Bone Surfaces	130 1)	3,9
Red Bone Marrow	16	1,9
Other Soft Tissues	1,4	1,2
Effective Dose Equivalent $H_E = \Sigma w H =$		7,0 2)

1) ICRP 2 (1959) : $\bar{H}_{Bone} \approx 800 \cdot 10^{-7}$ Sv per Bq with $Q_\alpha = 10$

2) ICRP 30(1979) : H_E ($T > 30$ y) = $3,2 \cdot 10^{-7}$ Sv per Bq for workers with $f_1 = 0.2$ and f_{Rn} (bone) = 0.3

Table 7: Mean Activity and Dose Rate per Unit of Ingestion Rate of Th-232 with the diet, derived from the natural exposure of the population ($\bar{I} \approx 1.5$ Bq/y)

Source Tissue (S)	$a_S/\dot{I}_{ing}$ Bq/kg per Bq/y	Target Tissue (T)	$\dot{H}_T/\dot{I}_{ing}$ 10^{-6} Sv/Bq
Bone	0.01	Bone Surface	70
		RBM	10
Bone Marrow	0.001		
Residual Body	0.0003 - 0.001	Other Tissues	1 - 3
Effective Dose ca. $5 \cdot 10^{-6}$ Sv per Bq *1)*			

1) ICRP 30 (1979): $H_E/I_{ing} = 0.8 \cdot 10^{-6}$ Sv per Bq ($f_1 = 2 \cdot 10^{-4}$)

The ingestion dose factors for the natural exposure of the population are in general higher than the ICRP-values for workers. This tendency reflects partly the different age adistribution in both groups, but will be probably mainly caused by a higher f_1-value for dietary intake.

In the case of Ra-226 and Pb-210 the difference is relative small and within the error limits of the natural exposure data. However for U- and Th-isotopes and for Po-210 the mean ingestion dose factor for intake with the diet seems to be 5 - 8 times higher than the ICRP-value for workers.

FINAL REMARKS

With respect to the assessment and limitation of the exposure of members of the public by long-lived natural radionuclides it has to be taken into account that the committed dose from a short-term or chronical exposure is distributed over the whole life-time. Therefore it might be not appropriate to set up dose factors or intake limits for different age groups of the population. Considering the total life-time risk it seems more reasonable to lay down average dose factors and average annual limits of intake, which are averaged over the whole life-time. For this purpose the ingestion dose factors, derived from the natural exposure, might be adequate.

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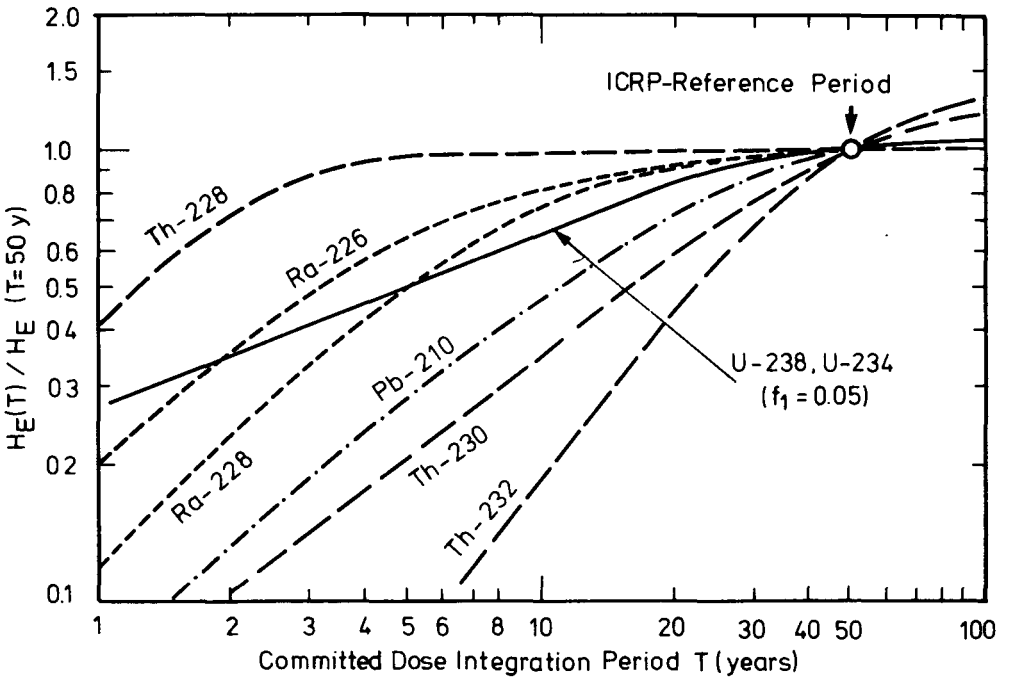

Fig. 1: Relative variation of the committed, effective dose with the integration period

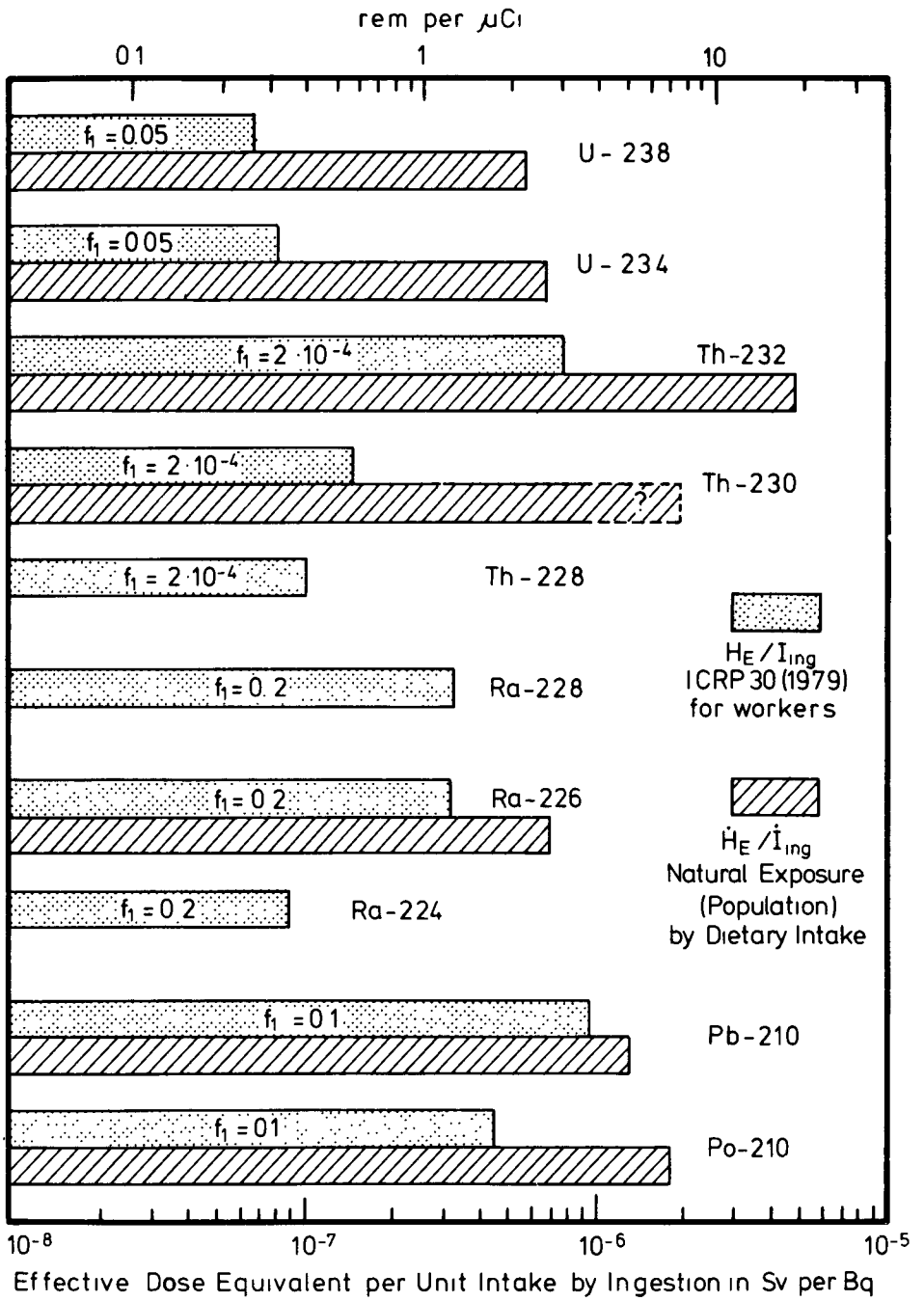

Fig. 2: Comparison of ingestion dose factors derived from the natural exposure with those recommended by ICRP for workers

QUELQUES OBSERVATIONS A PROPOS DE LA RADIOACTIVITE NATURELLE DE L'ALIMENTATION EN FRANCE

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S.C.P.R.I.
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RESUME. Le Service Central de Protection contre les Rayonnements Ionisants effectue régulièrement dans le cadre de la surveillance de l'environnement, des mesures de radioactivité naturelle (Potassium, Uranium, Radium-226 et Thorium) dans les constituants essentiels de la chaîne alimentaire. Ces mesures sont réalisées sur des prélèvements issus de différentes stations réparties sur tout le territoire français.
Les données essentielles concernent:
- les eaux: eaux superficielles, eaux souterraines, et plus de 600 eaux minérales,
- le régime alimentaire complet,
- différents constituants du régime alimentaire (céréales, poissons, fruits, légumes...).

Les auteurs exposent les méthodes analytiques utilisées pour la détermination de ces radioéléments et présentent les valeurs moyennes correspondantes.

KURZFASSUNG. MESSUNGEN DER VOM MENSCHEN AUFGENOMMENEN NATÜRLICHEN RADIOAKTIVITÄT IN FRANKREICH. Der "Service Central de Protection contre les Rayonnements Ionisants" (S.C.P.R.I.) führt im Rahmen der Umweltüberwachung regelmässig Messungen der natürlichen Radioaktivität (Kalium, Uran, Radium-226 und Thorium) in den wichtigsten Komponenten der Nahrungsmittelkette durch. Die Messungen werden an Proben vorgenommen, die aus verschiedenen über das französische Hoheitsgebiet verteilte Stationen stammen.
Die wichtigsten Daten betreffen:
- das Wasser: Oberflächengewässer, Grundwasser und mehr als 600 Mineralwässer,
- das vollständige Nahrungsmittelangebot,
- einzelne Bestandteile des Nahrungsmittelangebots (Getreide, Fisch, Obst, Gemüse usw.).

Die für die Bestimmung dieser Radionuklide benutzten Analysenmethoden werden erläutert und die gewonnenen Mittelwerte angegeben.

SUMMARY. SOME OBSERVATIONS ON NATURAL RADIOACTIVITY IN FOOD IN FRANCE. In the context of environmental studies, the S.C.P.R.I. (Service Central de Protection contre les Rayonnements Ionisants) performs regular measurements of natural radioactivity (potassium, radium and thorium) in the principle components of the diet. The measurements are carried out on samples coming

from various sampling locations covering the whole of France.

The principal data concern:

- water: surface waters, underground waters, and more than 600 mineral waters,
- the diet taken as a whole,
- principal components of the diet (cereals, fish, fruit, vegetables,..).

The authors explain the analytical procedures used for the determination of the radionuclides and present the mean values obtained.

INTRODUCTION

Le corps humain est soumis à une exposition interne permanente et inéluctable, consécutive à la présence des radioéléments incorporés durant la croissance et pendant toute la vie de l'individu. Elle correspond à plusieurs centaines de milliers de désintégrations par minute pour l'organisme entier. Si l'on exclut les très faibles activités du Césium 137 et de quelques autres radioéléments d'origine artificielle (qui ont beaucoup décru depuis 1961-62), la quasi-totalité de cette irradiation interne est donc due au Potassium 40, au Radium 226, et aux radioéléments d'origine cosmique, Tritium, Carbone 14, Béryllium 7, Sodium 22 : au total une trentaine de radioéléments sont impliqués dans l'exposition d'origine interne.

La mesure de ces divers radioéléments ne présente pas toujours le même intérêt. Celui-ci est fonction de leur nature, de leur abondance, de leur variabilité et bien sûr de leur radiotoxicité. Ainsi, d'une part la constance de la teneur en Potassium chimique du corps humain, soumise à travers la physiologie rénale à une rigoureuse homeostasie, d'autre part, l'abondance isotopique du Potassium 40, elle aussi d'une rigoureuse constance (1 atome radioactif sur 8 475 atomes stables), font qu'une étude systématique in vivo de cet élément n'aurait pas grand intérêt en elle-même.

Par contre, la mesure du Potassium 40 dans l'alimentation, notamment dans l'eau, présente l'intérêt de la mesure radioactive d'une substance chimique qui sert d'entraîneur pour d'autres éléments, le Césium 137 et le Césium 134 en particulier, du fait de l'identité des métabolismes.

Le rapport de 1977 du Comité Scientifique des Nations-Unies montre que l'exposition due au Potassium 40 à elle seule représente 88% de l'exposition de la moelle osseuse, 88% de celle des gonades, et 61% de celle de l'os. Ne fait exception que l'exposition du poumon qui est due essentiellement au Radon, mais pour laquelle le Potassium intervient encore pour 32%. Le Potassium est donc responsable pour l'essentiel de l'exposition interne naturelle du corps humain.

ORGANISATION DES MESURES

Dans le domaine bien particulier de la mesure de la radioactivité naturelle ingérée, les mesures effectuées par le SCPRI concernent essentiellement la surveillance des eaux d'alimentation et des produits alimentaires divers, poissons, fruits et légumes, récoltes des céréales, laits, miel, etc., l'étude des différentes sources hydrominérales du territoire, l'analyse des régimes alimentaires complets prélevés dans diverses collectivités.

En ce qui concerne la radioactivité naturelle du régime alimentaire, nous avons effectué 987 semaines de prélèvements dans 7 collectivités scolaires françaises de 1968 à 1978, soit 10 ans (tableau 1). L'organisation est la suivante : la personne responsable prélève, pendant une semaine chaque mois, la ration alimentaire d'un adolescent. Elle effectue une préparation sommaire sur place et nous envoie, d'une part, les aliments solides desséchés et, d'autre part, les liquides. Les prélèvements sont traités au laboratoire de radioanalyses du Vésinet. L'ensemble correspond donc à 6 900 rations quotidiennes d'élèves de 12 à 15 ans (aliments et boisson). Cet échantillonnage peut être considéré comme représentatif.

RESULTATS

Pour les aliments solides (tableau 2), l'activité quotidienne du Potassium 40 ingéré est en moyenne de 3 000 picocuries -pCi- par jour (avec un minimum de 1 800 et un maximum de 4 900). En Uranium, la moyenne est de 0,6 picocurie par jour. En Radium 226, l'activité ingérée se situe entre 0,8 et 2,9 pCi/jour.

Pour la partie liquide de l'ingestion, une importante contribution peut être due à la radioactivité naturelle des eaux d'adduction (qui restent le plus souvent bues en France). Le SCPRI a effectué depuis dix ans la mesure systématique réglementaire de la radioactivité sur tous les projets de nouveaux captages destinés à alimenter en eau potable une agglomération (tableau 3). Pour le Potassium 40, on distingue différentes classes de niveaux (en picocuries par litre -pCi/l-), de même que pour l'Uranium naturel et le Radium 226. Les activités maximales qui ont été relevées sont : 54 pCi/l pour le Potassium 40 dans une eau d'adduction bue couramment par une population du sud de la France ; 16 pCi/l pour l'Uranium naturel ; 19 pCi/l pour le Radium 226.

Par ailleurs, depuis quelques années, la consommation d'eaux minérales a beaucoup augmenté en France. Parmi les 650 sources hydrominérales que nous avons analysées, nous avons sélectionné les onze principales sources françaises d'eaux minérales de table (tableau 4). Pour chacune d'elle est indiqué le nombre de litres commercialisés par an arrondi au million. Ce sont les eaux d'Evian et de Contrexéville qui sont les plus consommées avec 720 et 700 millions de litres par an. Comme la France compte à peu près 50 millions d'habitants, chacun boit en moyenne 28 litres par an de ces eaux. Les chiffres les plus élevés en Potassium 40 sont ceux des sources de Saint-Yorre et de Saint-Sylvestre avec 120 milligrammes par litre. En ce qui concerne le Radium 226, c'est la source de Châteauneuf-les-Bains qui contient la plus forte proportion avec 26 pCi/l. Quant à l'Uranium, l'eau de Saint-Galmier-Badoit, de consommation assez courante puisqu'en moyenne chaque Français en boit un à deux litres par an, contient 79 microgrammes par litre d'Uranium naturel. Le Thorium, quant à lui, reste toujours inférieur à 11 microgrammes par litre.

En ce qui concerne le lait, l'étude a porté (tableau 5) sur 2 groupes de lait : d'une part les laits de ferme crus, qui sont prélevés sur 9 points de collecte et qui, par conséquent, ne correspondent pas à un très grand mélange du lait ; d'autre part sur le lait pasteurisé, collecté dans chaque département à la coopérative laitière la plus importante. L'on constate à peu près toujours les mêmes valeurs de la radioactivité du lait en Potassium 40.

Pour les céréales (tableau 6), nous analysons des prélèvements provenant d'environ 250 silos, dans différentes régions, et nos résultats portent aussi pratiquement sur 9 ans de contrôle. La concentration moyenne du Potassium 40 dans les grains entiers est de 3 300 picocuries par kilogramme de grain. Dans la farine, elle est de 1 100 picocuries par kilogramme.

Les résultats concernant les poissons de mer (tableau 7), qui représentent l'essentiel de l'alimentation en poisson, portent sur 840 prélèvements. Si l'on considère d'une part les poissons plats et d'autre part les poissons ronds, on voit, pour les poissons ronds, des valeurs légèrement supérieures pour le Potassium 40. Pour les poissons de rivières, qui sont naturellement beaucoup moins consommés, les chiffres sont nettement plus élevés en Uranium et Radium 226. On y trouve également une beaucoup plus forte concentration notamment des produits de la retombée des tests nucléaires que dans les

poissons d'eau de mer. L'explication est la suivante : l'eau de mer, qui est très chargée en éléments minéraux, est un milieu protecteur vis-à-vis de la radioactivité pour les organismes qui y vivent, car cette charge minérale joue le rôle d'entraîneur.

Enfin, les autres produits alimentaires (tableau 8) : fruits et légumes, coquillages et huile végétale. Les pommes de terre représentent, pour le Potassium 40, une activité relativement forte : 3 500 picocuries par kilogramme de pommes de terre. Pour les coquillages, l'activité est à peu près homogène : environ 2 200 pCi/kg. Un chiffre assez bas est celui des huiles végétales diverses : inférieur à 110 pCi/kg. Quant à un élément particulier de l'alimentation, le miel, la variabilité est considérable puisqu'on passe de 310 à 3 300 pCi/kg.

La radioactivité de l'alimentation est essentiellement d'origine naturelle et le Potassium 40 joue, là aussi, un rôle dominant. Mais ses variations parfois considérables d'un aliment à un autre n'entraînent par contre aucune variation résultante de l'exposition interne de l'organisme.

En effet, si l'exposition interne par le Potassium 40 est relativement importante (25% de l'exposition naturelle totale de l'homme), elle est remarquablement constante par suite de la fonction homeostatique rénale : tout apport de Potassium 40, si important soit-il, est immédiatement compensé par une élimination rénale équivalente, sous peine de désordres graves dans la régulation de l'équilibre plasmatique, et le Potassium reste constamment à 0,20% du poids du corps, soit 140 grammes pour l'homme-standard dont l'activité totale en Potassium 40 est donc de 110 000 picocuries.

La radioactivité artificielle, comparée à cette radioactivité naturelle, n'atteint jamais 1% de la radioactivité due au Potassium 40, et elle reste donc très inférieure aux seules fluctuations de la radioactivité naturelle totale du régime dues principalement à sa composition et à l'origine des divers aliments.

CONCLUSION

Pour conclure, il faut souligner qu'aucun travail sérieux n'a jamais établi de relations entre la présence de Radium, par exemple dans les eaux de boisson, même à des taux relativement élevés, et un éventuel accroissement de la fréquence des cancers et des leucémies dans quelque région que ce soit. Si des hypothèses ont été faites à partir d'un certain nombre de considérations purement théoriques, statistiques, et systématiquement pessimistes, elles n'ont jamais été, en aucun cas, confirmées par des constatations expérimentales sérieuses. Il est d'ailleurs essentiel de rappeler que, quand on avance un chiffre (par exemple celui de 100 cancers par an, par million d'habitants), il s'agit du risque statistique maximal, c'est-à-dire que ce nombre pourrait se situer entre 0 et 100, mais qu'il peut, en particulier, être aussi bien zéro.

Ce n'est d'ailleurs pas d'aujourd'hui qu'on parle de ces questions : un travail remarquable, qui reste toujours d'actualité, a été fait au Royaume-Uni par TURNER il y a une quinzaine d'années. Il est fondé sur des recherches et des enquêtes épidémiologiques très sérieuses qui n'ont, à notre connaissance jusqu'à présent, été menées qu'en Grande-Bretagne, sur les eaux potables distribuées à la population. L'absorption quotidienne moyenne du Radium 226 à partir de l'eau potable est plus de 30 fois plus importante dans les Cornouailles que dans le Pays de Galles. Elle est à peu près 1000 fois plus importante en ce qui concerne le Radon. L'on a donc, d'un côté les Cornouailles, où la radioactivité est élevée, et de l'autre le Pays de Galles où elle est beaucoup plus faible. Or aucune augmentation du taux normal de mortalité (ce que les Anglais appellent le Standardized Mortality Ratio - SMR) par cancer ou leucémie ne peut être mise en évidence dans la région des Cornouailles où, bien au contraire, le taux de mortalité correspondant à ces deux affections est même nettement inférieur au SMR. Par contre, bien que les Comtés du Pays de Galles choisis comme témoins soient alimentés en eau potable comportant une radioactivité naturelle particulièrement faible, il y est constaté, depuis de nombreuses années, un dépassement très important du SMR, notamment par cancers gastriques, atteignant 38%. Les eaux correspondantes proviennent généralement de drainages en surface, comportent un pH bas à l'origine, et sont très faiblement minéralisées (moins de 50 ppm de matière minérale dissoute), alors que les eaux des régions dans lesquelles le SMR reste normal bien qu'elles soient beaucoup plus radioactives, comportent plus de 200 ppm.

TURNER conclut en recommandant de porter attention, plus qu'à la radioactivité des eaux potables, à leur composition exacte en oligo-éléments minéraux, présents à l'état de traces dans les eaux dites douces (soft water), sur lesquelles le manque de connaissances, même encore à l'heure actuelle, au point de vue épidémiologique, reste total.

Des enquêtes du même type ont, depuis, été lancées notamment en Suède, au Canada et aux Etats-Unis, et on n'a pas encore réussi à trouver pourquoi les eaux légères sont ainsi plus dangereuses que les eaux très minéralisées. En tout cas, ce dont on est certain, c'est que la radioactivité n'y est pour rien.

Entre-temps d'ailleurs une brillante confrontation expérimentale, comme les Anglais savent en réaliser, a montré combien était juste l'interprétation de TURNER, car ils ont alimenté plusieurs Comtés du Pays de Galles (où la radioactivité était la plus faible avec un taux de mortalité plus élevé) avec de l'eau amenée par aqueducs depuis les Cornouailles : au bout de cinq ans, le taux de mortalité de ces seules régions s'est abaissé progressivement au taux de mortalité plus faible des Cornouailles.

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987 PRÉLÈVEMENTS EFFECTUÉS DANS 7 COLLECTIVITÉS SCOLAIRES DE 1968 À 1978

(CORRESPONDANT À 6 900 RATIONS QUOTIDIENNES D'ÉLÈVES DE 12 À 15 ANS, ALIMENTS ET BOISSONS COMPRISES).

TABLEAU 1 REPRESENTATIVITE DES ECHANTILLONS ANALYSES EN VUE DE LA MESURE DE LA RADIOACTIVITE NATURELLE DU REGIME ALIMENTAIRE

Radioélément Naturel	Activité Quotidienne Ingérée (picocuries)
Potassium 40	Moy. : 3 000 (min.: 1 800 - max.: 4 900)
Uranium	Moy. : 0,6 (min.: 0,2 - max.: 2)
Radium 226	Moy. : < 2 (min.: 0,8 - max.: 2,9)

Tableau 2 ELEMENTS RADIOACTIFS NATURELS MAJEURS DU REGIME ALIMENTAIRE

Radioélément	Différents niveaux (pCi/L)	Fréquence (% des Captages)	Activité maximale (pCi/L)
K 40	<0,9	21	54
	0,9 - 4,3	70	
	4,3 - 8,5	6	
	≥8,5	3	
U nat	<0,2	72	16
	0,2 - 1,7	27	
	≥1,7	1	
Ra 226	<2	98	19
	≥2	2	

Tableau 3 ELEMENTS RADIOACTIFS NATURELS MAJEURS DES EAUX D'ADDUCTION FRANCAISES

DEPT	SOURCE	K MG/L	U NAT µG/L	RA 226 PCI/L	TH NAT µG/L	MILLIONS DE LITRES PAR AN POUR 1979 (*)
74	EVIAN (CACHAT)	1,0	1,5	< 1	< 11	720
88	CONTREXÉVILLE	2,6	0,6	< 1	< 11	700
88	VITTEL (HÉPAR-GDE SOURCE)	2,5	1,1	< 1	< 11	540
30	PERRIER	3,4	3,4	< 1	< 11	130
03	SAINT YORRE (13 SOURCES)	120	1,3	12	< 11	110
42	ST GALMIER (BADOIT)	16	79	2,1	< 11	75
63	ST SYLVESTRE	120	1,4	10	< 11	56
03	VICHY (3 SOURCES)	100	0,35	17	< 11	43
22	PLANCOET (SASSAY)	2,6	< 0,6	< 1	< 11	15
63	CHATEAUNEUF-LES-BAINS	44	< 0,6	26	< 11	6
07	VALS (4 SOURCES)	40	< 0,6	2,2	< 11	6

TABLEAU 4 RADIOACTIVITE DES 11 PRINCIPALES SOURCES FRANCAISES HYDROMINERALES DE TABLE

(*) COMPTE NON TENU DES EXPORTATIONS

QUALITE ET ORIGINE DES LAITS	ANNÉE	K 40 EN pCi/L	
		MOYENNE ANNUELLE	VALEURS MIN. & MAX.
LAITS DE FERME CRUS	1971	1 290	1060 - 1530
	1972	1 280	935 - 1450
(216 PRÉLÈVEMENTS ANNUELS CORRESPONDANT À 9 POINTS DE COLLECTE)	1973	1 290	1020 - 1650
	1974	1 270	816 - 1480
	1975	1 320	1140 - 1480
	1976	1 260	850 - 1500
	1977	1 270	980 - 1500
	1978	1 250	880 - 1480
LAITS PASTEURISÉS	1971	1 310	1100 - 1480
	1972	1 290	1050 - 1430
(ENVIRON 360 PRÉLÈVEMENTS ANNUELS PROVENANT DE 89 CENTRES LAITIERS IMPORTANTS)	1973	1 300	1110 - 1520
	1974	1 310	1090 - 1660
	1975	1 310	1050 - 1530
	1976	1 280	880 - 1600
	1977	1 280	1070 - 1600
	1978	1 280	1030 - 1620

TABLEAU 5 POTASSIUM 40 DANS LE LAIT

ANNEES DE RECOLTE	K 40 EN pCi/KG FRAIS	
	GRAINS ENTIERS	FARINES
1969 A 1978	3 300	1 100

TABLEAU 6 MOYENNES DU POTASSIUM 40 DANS LE BLE

CATEGORIE DE POISSON	pCi/kg de chair (frais)		
	K 40	U nat	Ra 226
POISSONS PLATS (sur 390 prélèvements)	2 200	< 0,3	< 1
POISSONS RONDS (sur 450 prélèvements)	2 600	< 0,5	< 1

Tableau 7 — RADIOACTIVITE NATURELLE DES POISSONS D'EAU DE MER

PRODUIT ALIMENTAIRE	pCi DE K 40 PAR KG FRAIS
FRUITS ET LÉGUMES	
PÊCHE	1 100
POMME	770
TOMATE	1 700
MELON	2 300
RAISIN	1 400
ASPERGE	1 600
POMME DE TERRE	3 500
COQUILLAGES	
COQUILLE ST-JACQUES	2 200
HUÎTRE	2 200
TELLINE	2 100
HUILES VÉGÉTALES DIVERSES	< 110
MIEL	310 à 3 300

TABLEAU 8 POTASSIUM 40 DANS DIVERS PRODUITS ALIMENTAIRES

THE EXPOSURE OF THE ITALIAN POPULATION TO NATURAL RADIOACTIVITY IN DRINKING WATER AND FOOD

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SUMMARY. An extensive survey of some natural radionuclide levels in Italian drinking waters and foods has been carried out over the past years. Ra-226, Rn-222, Po-210 and Pb-210 levels have been measured in a large variety of water supplies, mineral waters, foods and diets, with special regard to the products employed in bottle-feeding in the first year of life.

The samples have been chosen in order to be representative of large population groups, or of groups characterized by different dietary habits.

The results obtained, while not showing exceptionally high levels, indicate that the use of some products (e.g. mineral waters) may give rise to exposure from Ra-226 higher than normal, when account is taken of the enhanced availability to the human metabolism of such an element when present in water.

RESUME. EXPOSITION DE LA POPULATION ITALIENNE A LA RADIOACTIVITE NATURELLE DE L'EAU POTABLE ET DES ALIMENTS. Une étude approfondie des niveaux de quelques radionucléides naturels présents dans les eaux et les aliments en Italie a été conduite ces dernières années. On a mesuré les niveaux de Ra-226, Rn-222, Po-210 et Pb-210 pour une grande variété d'eaux d'adduction, d'eaux minérales, d'aliments et de régimes avec attention particulière pour les produits entrant dans l'alimentation au biberon des nourrissons.

L'échantillonnage a été réalisé de façon à être représentatif de larges groupes de population ou de groupes caractérisés par des habitudes alimentaires différentes.

Les niveaux relevés quoique n'étant pas exceptionnellement hauts montrent que la consommation de certains produits (par ex. les eaux minérales) peut entraîner une exposition au Ra-226 plus élevée que la normale si l'on tient compte aussi de ce que, dans l'eau, cet élément est plus disponible pour le métabolisme humain.

KURZFASSUNG. DIE EXPOSITION DER ITALIENISCHEN BEVÖLKERUNG DURCH NATÜRLICHE RADIOAKTIVITÄT IN TRINKWASSER UND NAHRUNGSMITTELN. In den letzten Jahren wurde ein umfassender Überblick über die Pegel einiger natürlicher Radionuklide im italienischen Trink- und Tafelwasser sowie in italienischen Nahrungsmitteln gewonnen. Die Ra-226-, Rn-222-, Po-210- und Pb-210-Pegel wurden in einer Vielzahl von Trinkwasserproben und Mineralwässern sowie Nahrungsmitteln und Diätkost gemessen, wobei die für die Flaschenernährung des Säuglings im ersten Lebensjahr verwendeten Erzeugnisse besondere Beachtung fanden.

Bei der Auswahl der Proben wurde darauf geachtet, dass sie einen repräsentativen Querschnitt durch grosse Bevölkerungsgruppen oder Gruppen mit unterschiedlichen Ernährungsgewohnheiten darstellen.

Wenn auch die vorliegenden Ergebnisse keine aussergewöhnlich hohen Pegel erkennen lassen, so zeigt sich doch, dass der Genuss einiger Erzeugnisse (z.B. von Mineralwasser) zu einer überdurchschnittlichen Strahlenbelastung durch Ra-226 führen kann, wenn man zusätzlich berücksichtigt, dass ein solches Element dann in verstärktem Masse auf den menschlichen Stoffwechsel einwirkt, wenn es im Wasser auftritt.

INTRODUCTION

It is well known that under normal conditions the preminent contribution to human exposure from natural radioactivity is due to external irradiation of both cosmic and terrestrial origin (1). Internal irradiation is generally responsible for about 20% of the total exposure, but increases up to 40% if irradiation of particular organs (e.g. skeleton, lung) is considered (1). Nonetheless recent studies (1,2,3) evidentiated that under particular conditions of housing and/or dietary habits the internal exposure largely exceed the external one leading to annual dose values eventually larger than those established as maximum for irradiation after exploitation of pacific application of nuclear energy, and generally accepted as reference for exposures from any kind of activity. Previous results (4,5) related to dietary habits very widespread in Italy, indicated also that the use of mineral water might give a consistent contribution to dietary internal contamination due to Radium-226 and Lead-210. A study has been therefore carried out in past years to evaluate:

a) the normal level of these radionuclides in diet samples representative for the italian population;
b) the contribution eventually due to the use of mineral water as table water;
c) recognize possible population groups exposed to higher levels.

Such concern seemed justified as mineral water is widely used in bottle feeding of babies in the first year of life.

SAMPLING AND RESULTS

All the 207 mineral waters traded and consumed in Italy have been sampled. The locations of the sources are shown in Fig. 1 (single point may represent various springs). The Radon-222 content of mineral waters, as obtained from licensing data, the measured Radium-226 concentrations, the Radium-226/Ca ratio and the measured Lead-210 values (6) are given in Fig. 2,3,4 and 5 respectively. The values found, even if show high figure for some samples, are almost similar to those from other countries (7,8,9,10,11). The samples have been collected as they reach the consumer, as a preliminar study had showed that Radium-226 and Lead-210 absorption on the wall of the standard commercial bottles was almost negligible within the considered concentration range. Ten of the largest water supplies based on surface reservoir and large water tables have been also sampled in order to test their contribution to the daily intakes. Surface water showed Radium-226 concentrations below 0.04 pCi/l, while underground water ranged between 0.04 and 0.3 pCi/l. Lead-210 level was below the detection limit of 0.2 pCi/l.

Milk samples from 15 main factories of Italy have been also analyzed. The areas of origin of the samples together with their respective Radium-226 concentration and Radium-226/Ca ratio are given in Fig.6 and show values very similar to those found by others, in Italy(12) and in United States (13).

Adult diets have been collected in four towns (ten samples each) characterized by different geographical, social and pollution conditions, namely:

- a large town;
- an industrialized town;
- two towns located in rural areas, but with different socio-economical conditions.

The values found for Radium-226 range between 0.3 and 2 pCi/d for the daily intake and between 0.1 and 0.9 pCi Radium-226/g Ca, to be compared with the values measured in normal background areas of other countries ranging from 0.2 to 4 pCi/d and from 0.3 to 4 pCi Radium-226/g Ca (1,12,13,14,15,16,17). The values found for Lead-210 (18) range between 2.1 and 3.3 pCi/d to be compared with the values measured in other countries (Arctic Region excluded) ranging from 1.2 and 6.2 pCi/d (1, 13,19-28).

Teen-ager's diets have been collected in boarding schools of three large towns, in northern, central and southern Italy respectively. The values measured for Radium-226 range between 0.3 and 0.9 pCi/d for the daily intake and between 0.3 and 1 for Radium-226/g Ca ratio, and are very similar to those found in other European countries for adolescent diets ranging from 0.5 to 2.8 pCi/d for the daily intake and from 0.5 to 2.2 for Radium-226/g Ca (29).

For the first year's diet, 23 of the most widespread infant foods (powdered milk, cereal flour, food supplement) have been sampled, and according to the reccomended allowances, daily intakes have been evaluated relatively to the following periods:

0 - 1 month
0 - 3 months
0 - 6 months
0 - 12 months

For the last three periods a similar evaluation has been applied to diets based on cow milk, for which Radium-226 concentration has been derived from the values measured through the country. Since in these diets mineral water is normally used, Radium-226 concentrazion has been estimated on the basis of the reccomended types, in the range 0.1 - 4 pCi/l. The daily intake for each diet has been consequently derived as shown in Table 1. In Fig. 7 are shown the locations of the towns where adult and teen-ager's diet has been sampled together with Radium-226 daily intake and Radium-226 Ca ratio, while in Fig.8 are shown the results obtained on adult diet for Lead-210. In Fig. 9 all the results of Radium-226 are summarized as daily intake ranges. First year diets may cause intakes higher than normal, the lowest being those based on cow milk. This has to be accounted to the use of mineral water whose Radium-226 concentration may be up to 100 times higher than of tap water. Adult and teen-ager's diet samples containing only tap water, in fact, show the lowest values of daily intake. In order to evaluate the contribution of mineral water which is very often used as table water in the daily diet, an average consumption of 0.5 l/d may be assumed and its Radium-226 content added to the range of measured values. The normal range of daily intake results therefore enhanced up to about 10 pCi/d of Radium-226; a rather high value even if below the proposed limit of 20 pCi/d (30). For Lead-210 mineral water contribution resulted to be unrelevant when compared to dietary levels.

If the Radium-226/Ca ratio of the various samples is considered (see Fig.10), it may be seen that the widest range of variation is found for mineral water, while the other samples are well around the value 1 as found by others (12,14) and generally accepted as the normal one (1). On the basis of previous studies (31) the uptake of some elements by man seems to depend on the matrix in which they are found. Hence, owing to the higher exchange power of the elements in ionic form (as in water) as compared

to their availability when bound in organic matter, element uptake from water might be greater than from other items of the diet (32). Besides it has been recently proved that uptake of elements (e.g. lead) from soft water is larger than from hard water (33). Therefore it is reasonable to infer that the contribution of mineral water through a sistematic assumption may widely enhance Radium-226 intake and uptake by man.

DISCUSSION

On the basis of the results presented, annual doses have been estimated for internal exposure from Radium-226 and Lead-210 - Polonium-210 (18). Such values have been compared in Table 2 with annual dose estimates from external exposure as measured in Italy in a previous nationwide survey (34), and from internal exposure due ro cosmogenic radionuclides and to primordial ones other than Radium-226 and Lead-210 - Polonium - 210 (1). Annual dose estimated have been also carried out on the basis of diets including mineral water, and are reported in second coloumn of Table 2. As it can be seen the contribution of Radium-226 from the use of mineral water may lead to equivalent absorbed dose contributions even larger than those from external exposures, while contribution of Lead-210 is almost unaltered.

Natural background radiation exposure higher than those considered as normal (100 mrad/y) have been found in a large area of central Italy (34) (see Fig. 11). An average exposure of 180 mrad/y was measured in these regions and related to the volcanic rock formations which characterize Latium and Campania (35). Nevertheless the variability related to dietary composition resulted to be larger than that related to geographical and geological criteria. This might be explained as food is almost consumed elswhere its production area. This trade-induced averaging effect, which characterizes large towns more than villages, has the advantage that possible local food chain contaminations should be strongly reduced by the large scale product pooling and redistribution.

CONCLUSION

Internal exposure due to primordial radionuclides ingested through the diet resulted to be generally a small portion of the total exposure of man from natural sources also in Italy. Nevertheless it has been evidentiated that the use of mineral water, eventually characterized by high levels of natural radioactivity, may give place to higher exposure both in adult and in infant individuals. Even if such exposures are smaller than those often originated by indoor irradiation due to high natural radioactivity building materials, or high Radon-222 level tap water, it seems worthwhile that large consumption of mineral water should be regulated on the basis of derived limits, leaving those exceeding such values for special therapeutical applications.

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Table 1

Radium-226 daily intake and Radium-226 to calcium ratio ranges for first year diet

Period (Months)	Radium-226 daily intake (pCi/d)		Radium-226 to calcium ratio (pCi Ra-226/g Ca)	
0 - 1	0.07	2.2	0.30	13
0 - 3	0.17	3.2	.24	15
0 - 6	0.70	4.5	0.80	11
0 - 12	1.2	5.9	1.8	13

Table 2

Estimated annual doses from natural radioactivity absorbed in some tissues for italian normal population individuals (mrad/y)

Exposure	Normal Diet			Normal Diet + Mineral Water		
	Whole-Body	Skel.	Lung	Whole-Body	Skel.	Lung
External Exposure:						
Cosmic Radiation	36	36	36	-	-	-
Terrestrial Radiation	63-200	35-110	35-110	-	-	-
Internal Exposure:						
Cosmogenic Radionuclides	1.3	0.6	2	1.3	0.6	2
Primordial Radionuclides:						
Lead-210, Polonium-210	0.7	7	0.3	0.9	9	0.5
Radium-226	< 0.1	0.7	0.03	0.5	4	0.1
Others (Potassium-40 incl.)	16	19	52	16	19	52

LIST OF FIGURES

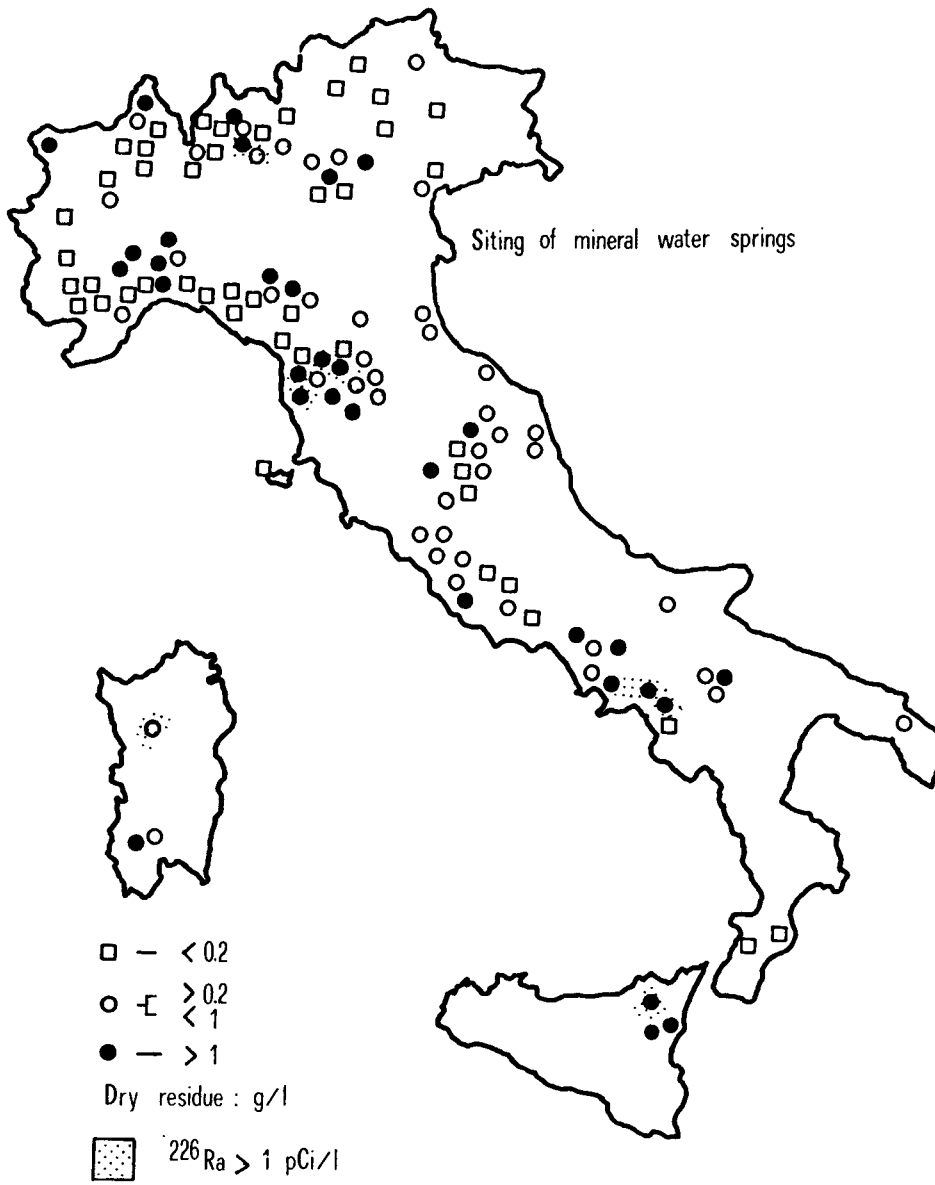
Siting of mineral water springs
□ — < 0.2
○ — > 0.2 < 1
● — > 1
Dry residue : g/l
226Ra > 1 pCi/l

Fig.1

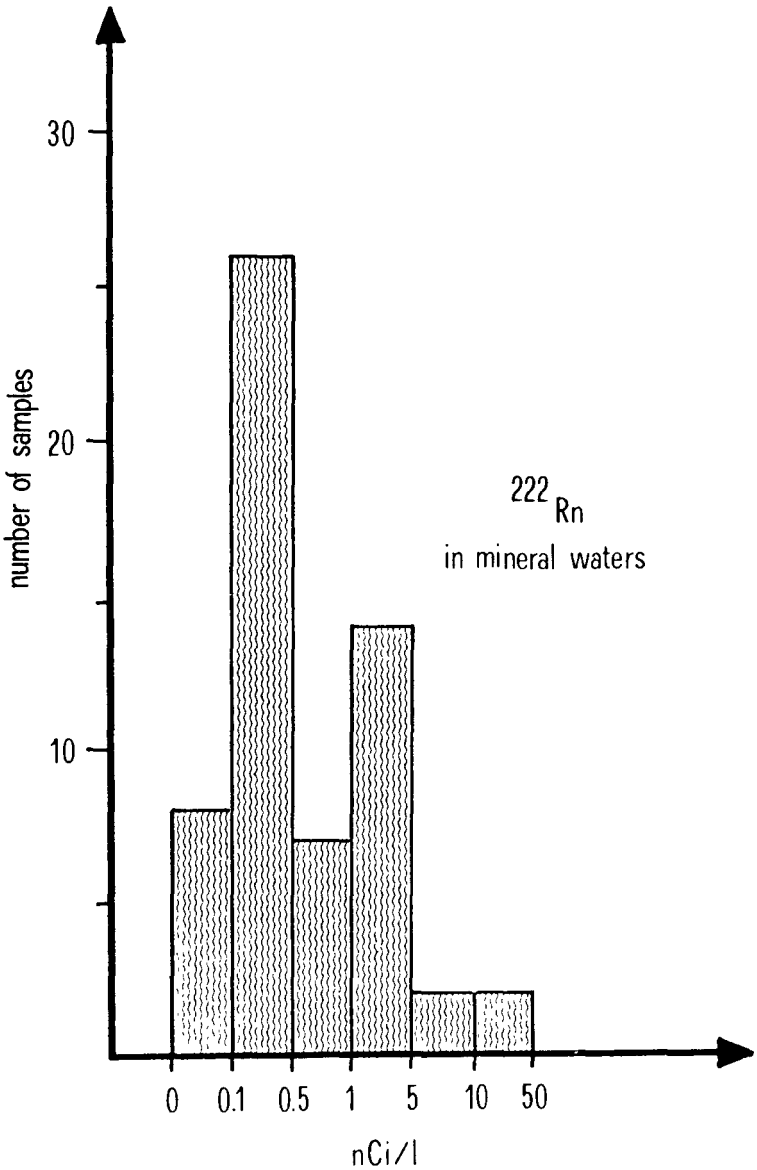
30
20
10
number of samples
222 Rn
in mineral waters
0
0.1
0.5
1
5
10
50
nCi/l

Fig. 2

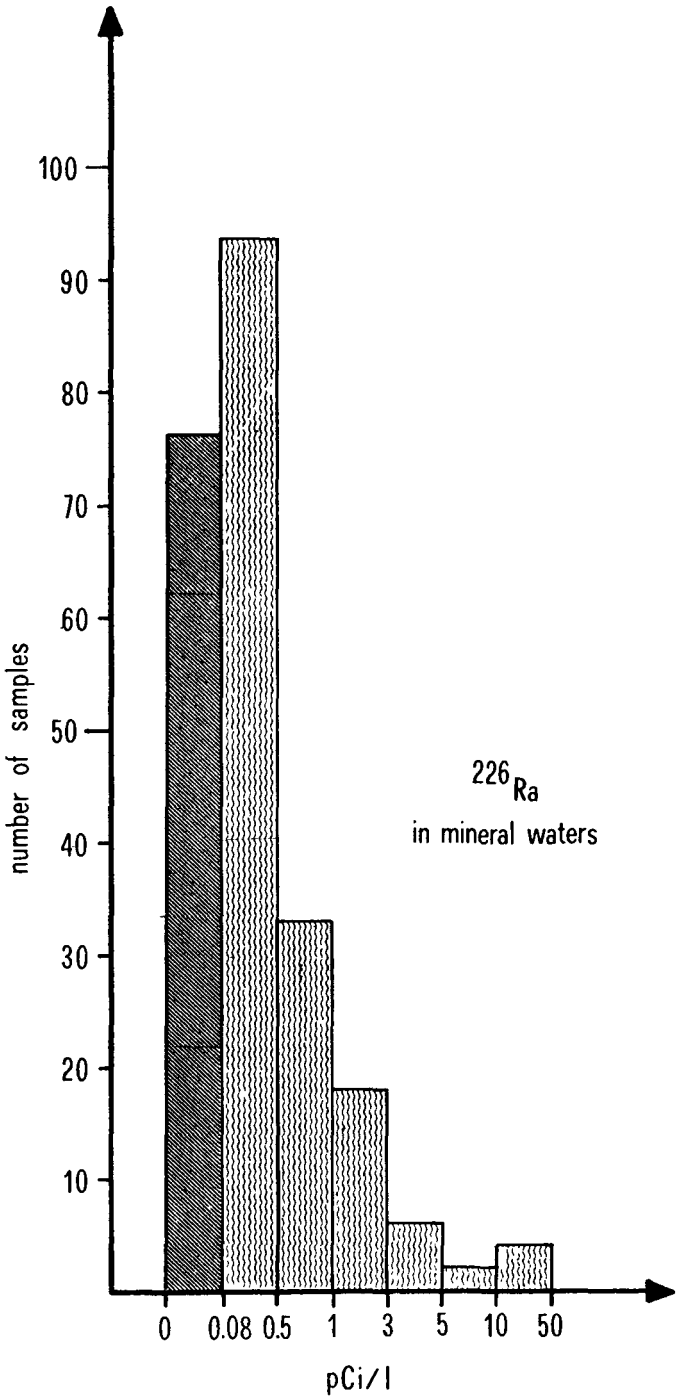
number of samples
100
90
80
70
60
50
40
30
20
10
^{226}Ra
in mineral waters
0
0.08
0.5
1
3
5
10
50
pCi/l

Fig.3

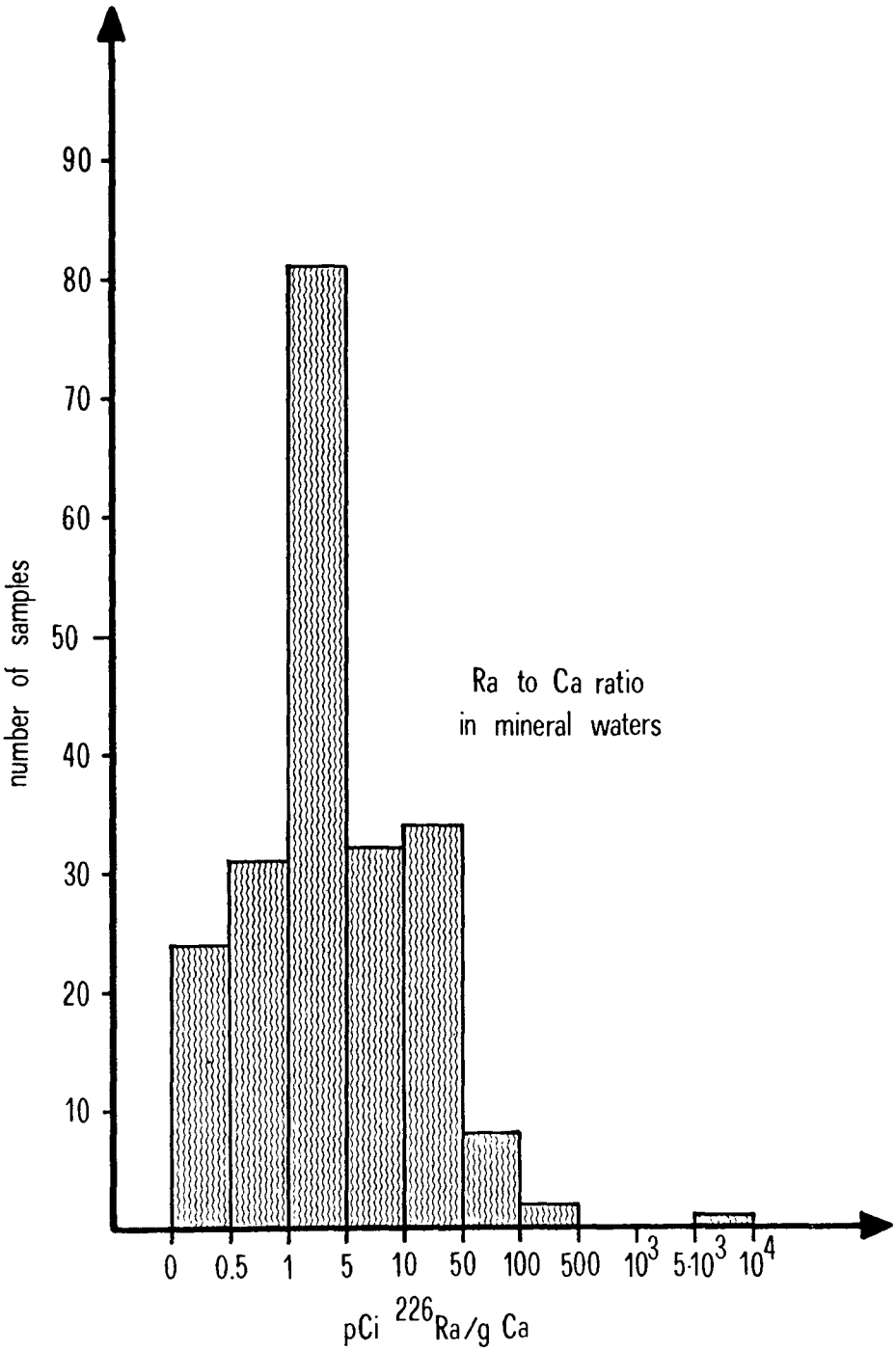
number of samples
90
80
70
60
50
40
30
20
10
Ra to Ca ratio
in mineral waters
0
0.5
1
5
10
50
100
500
10^3
$5 \cdot 10^3$
10^4
pCi ^{226}Ra/g Ca

Fig. 4

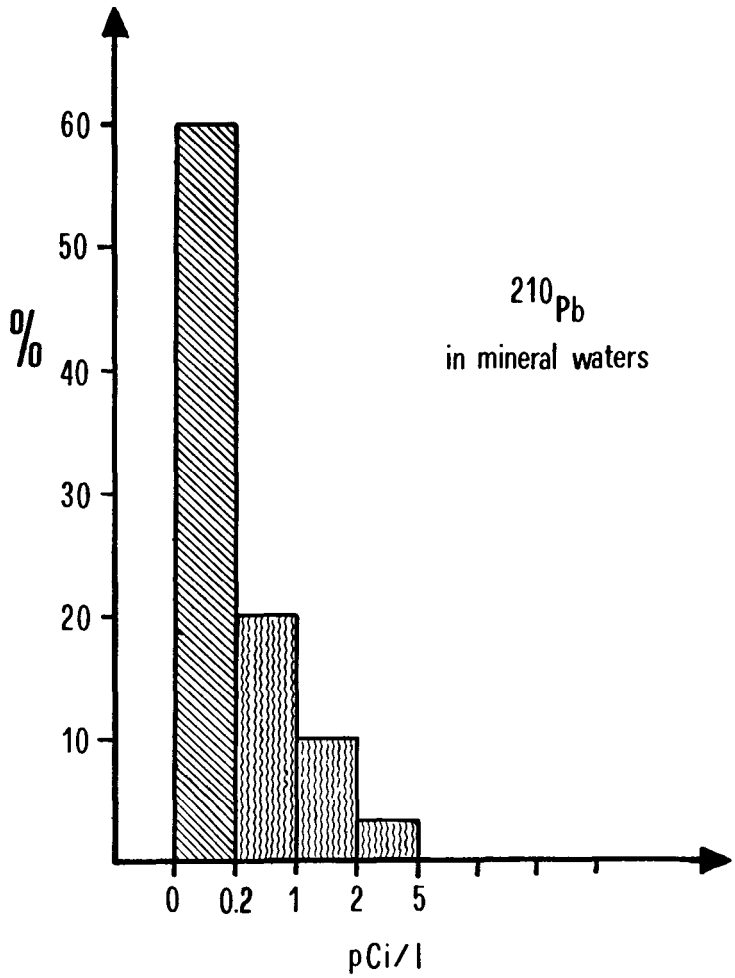

Fig.5

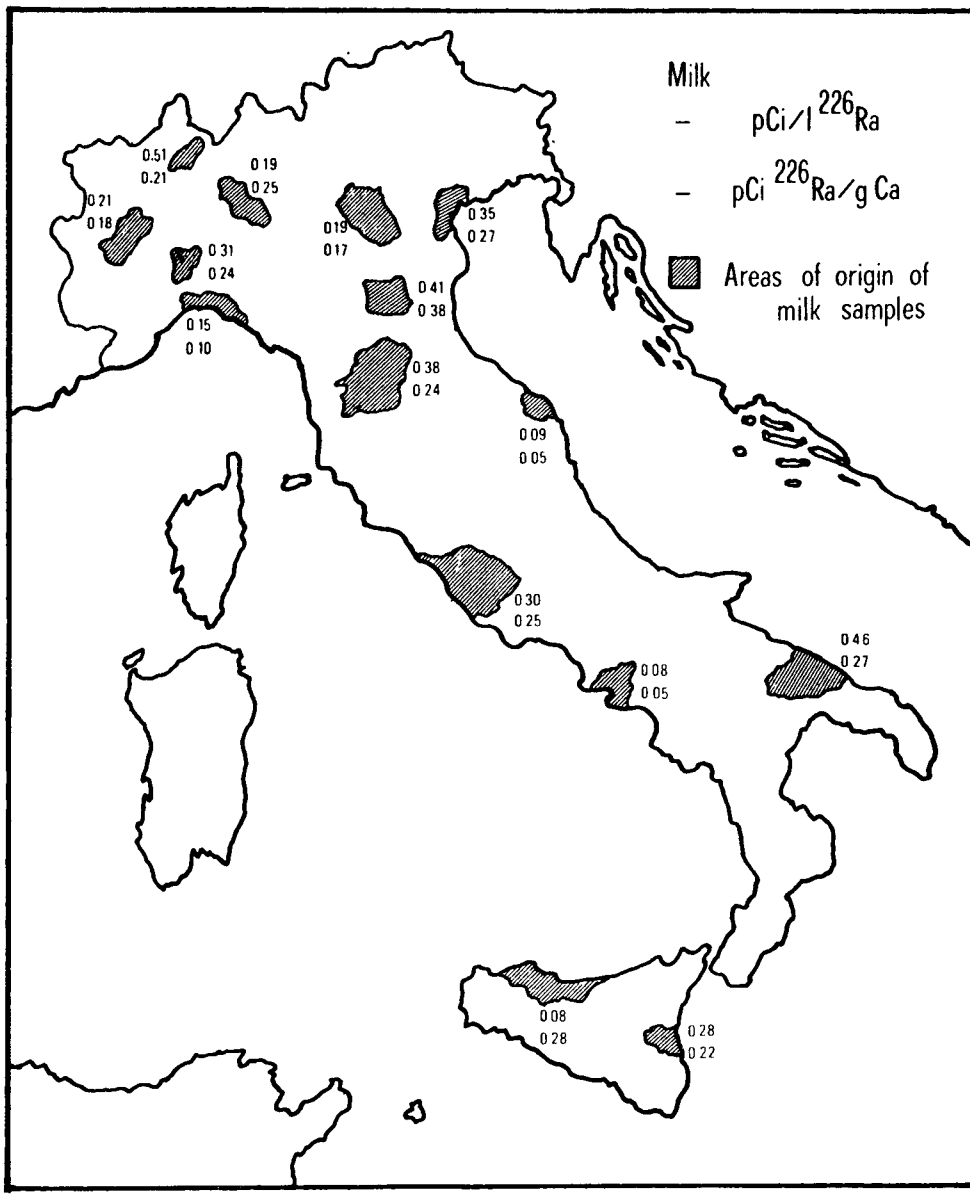
Milk
– pCi/l ^{226}Ra
– pCi ^{226}Ra/g Ca
Areas of origin of milk samples
0.51
0.21
0 19
0 25
0 21
0 18
0 31
0 24
0 15
0 10
0 19
0 17
0 35
0 27
0 41
0 38
0 38
0 24
0 09
0 05
0 30
0 25
0 46
0 27
0 08
0 05
0 08
0 28
0 28
0 22

Fig.6

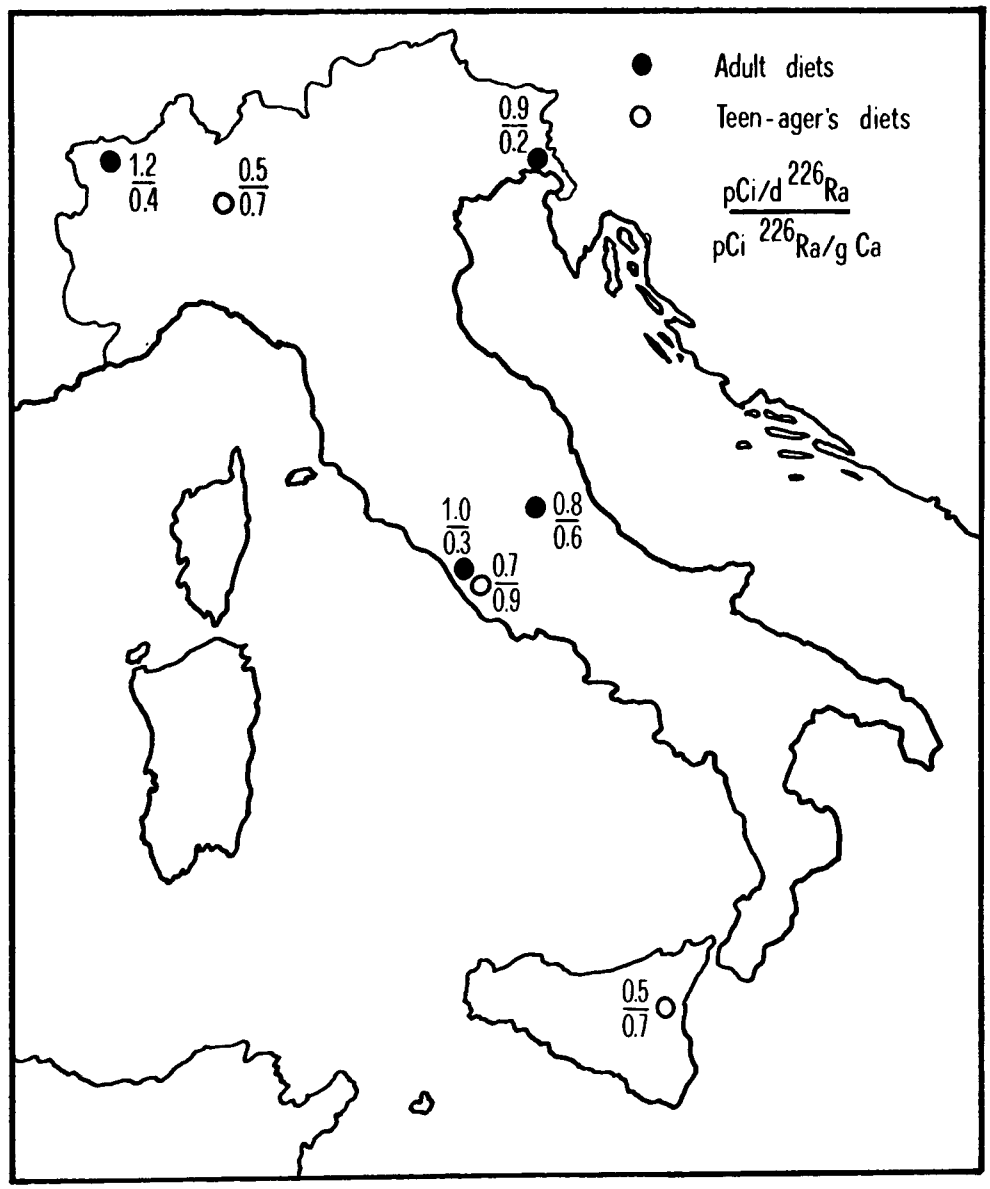
Adult diets
Teen-ager's diets
pCi/d ^{226}Ra / pCi ^{226}Ra/g Ca
0.9/0.2
1.2/0.4
0.5/0.7
1.0/0.3
0.8/0.6
0.7/0.9
0.5/0.7

Fig. 7

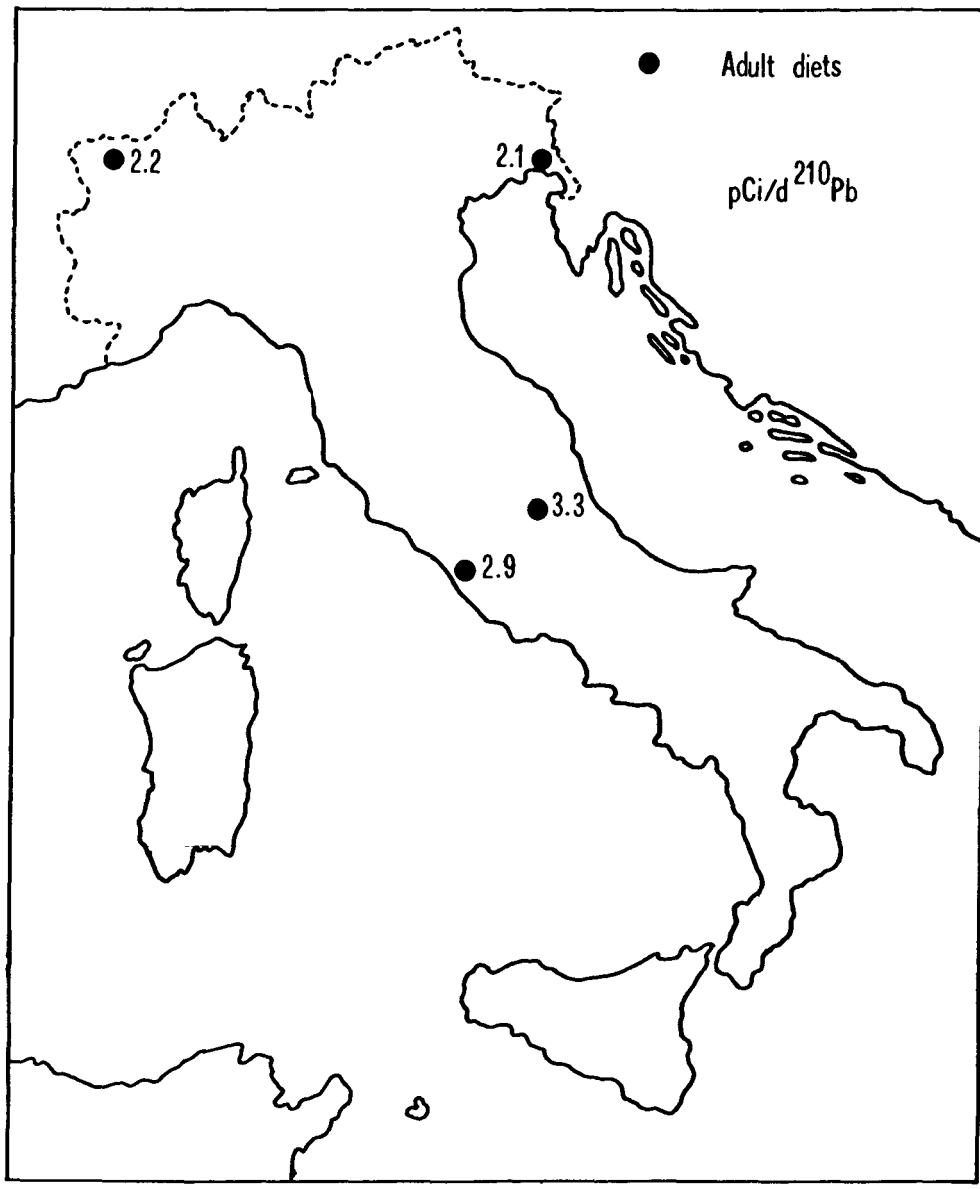

Fig.8

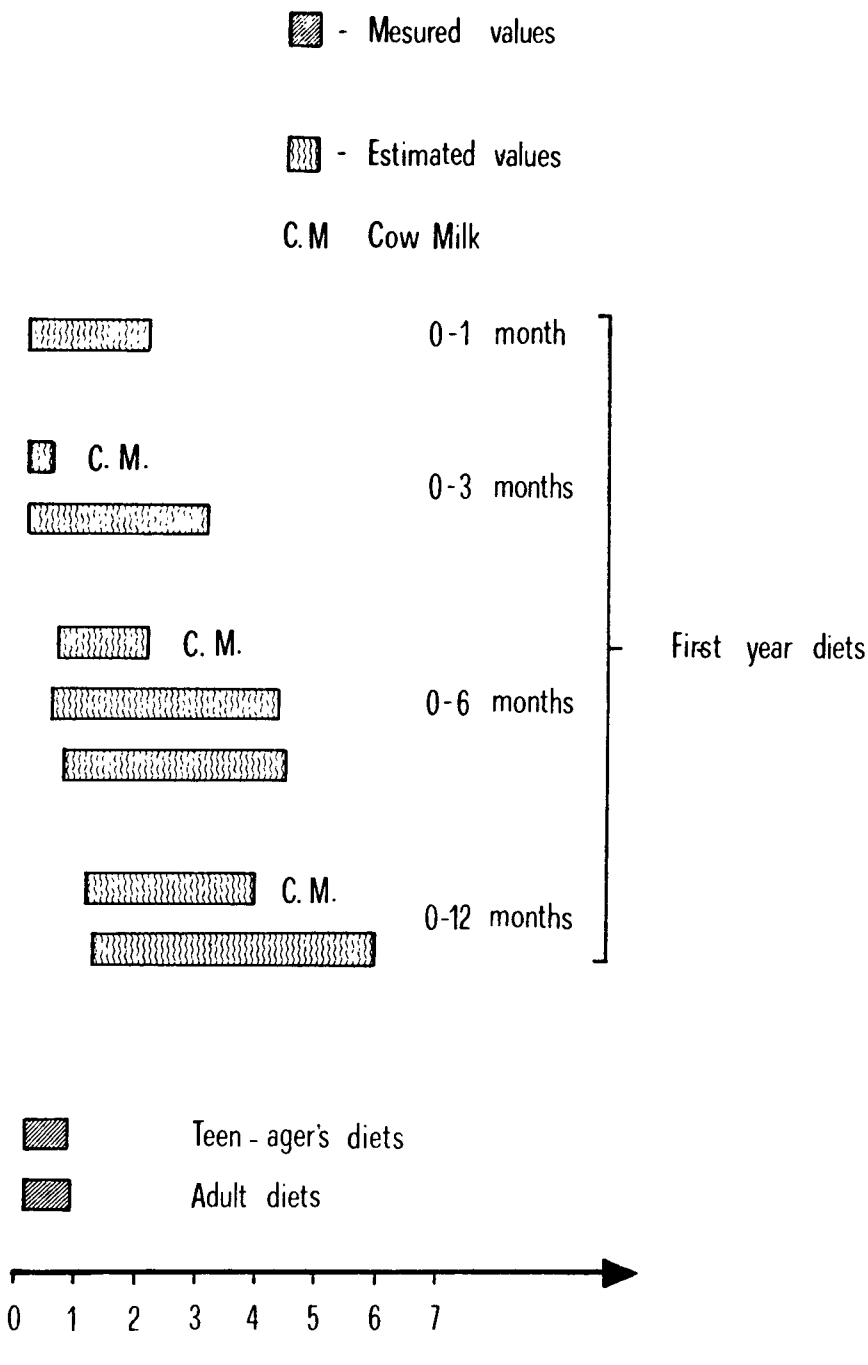
- Mesured values
- Estimated values
C.M Cow Milk
0-1 month
C. M.
0-3 months
C. M.
0-6 months
C. M.
0-12 months
First year diets
Teen - ager's diets
Adult diets
0 1 2 3 4 5 6 7
226Ra intakes
pCi/d

Fig. 9

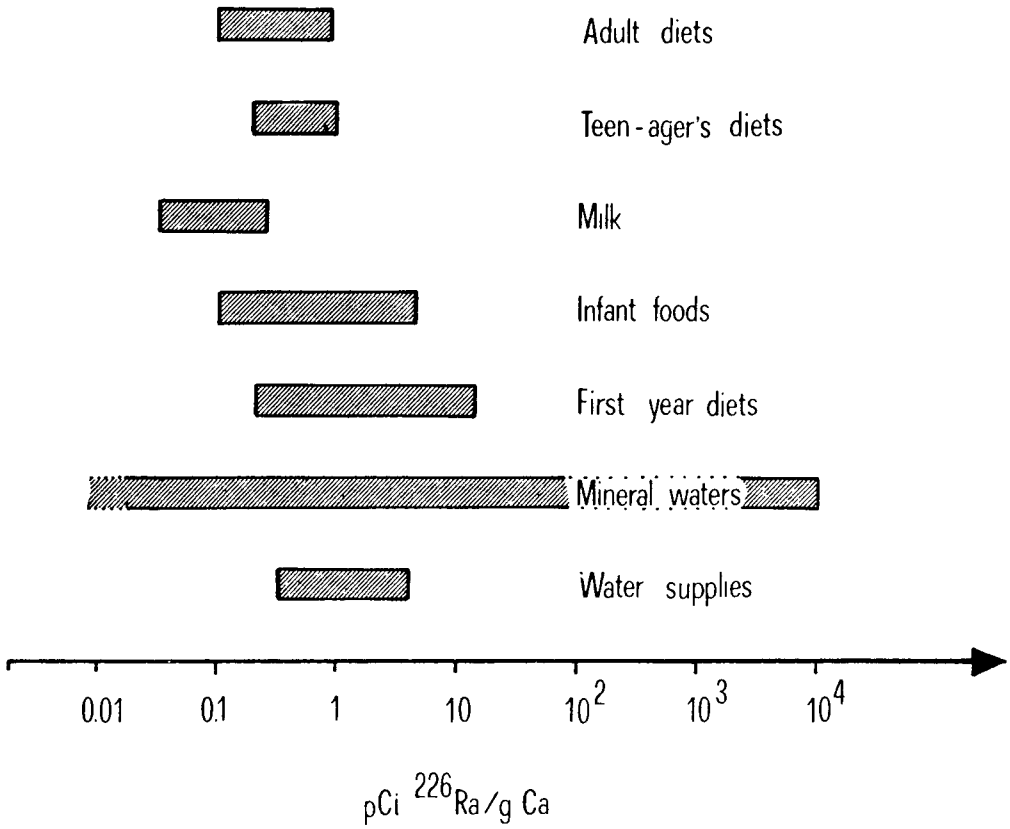
Adult diets
Teen-ager's diets
Milk
Infant foods
First year diets
Mineral waters
Water supplies
0.01
0.1
1
10
10^2
10^3
10^4
pCi ^{226}Ra/g Ca

Fig. 10

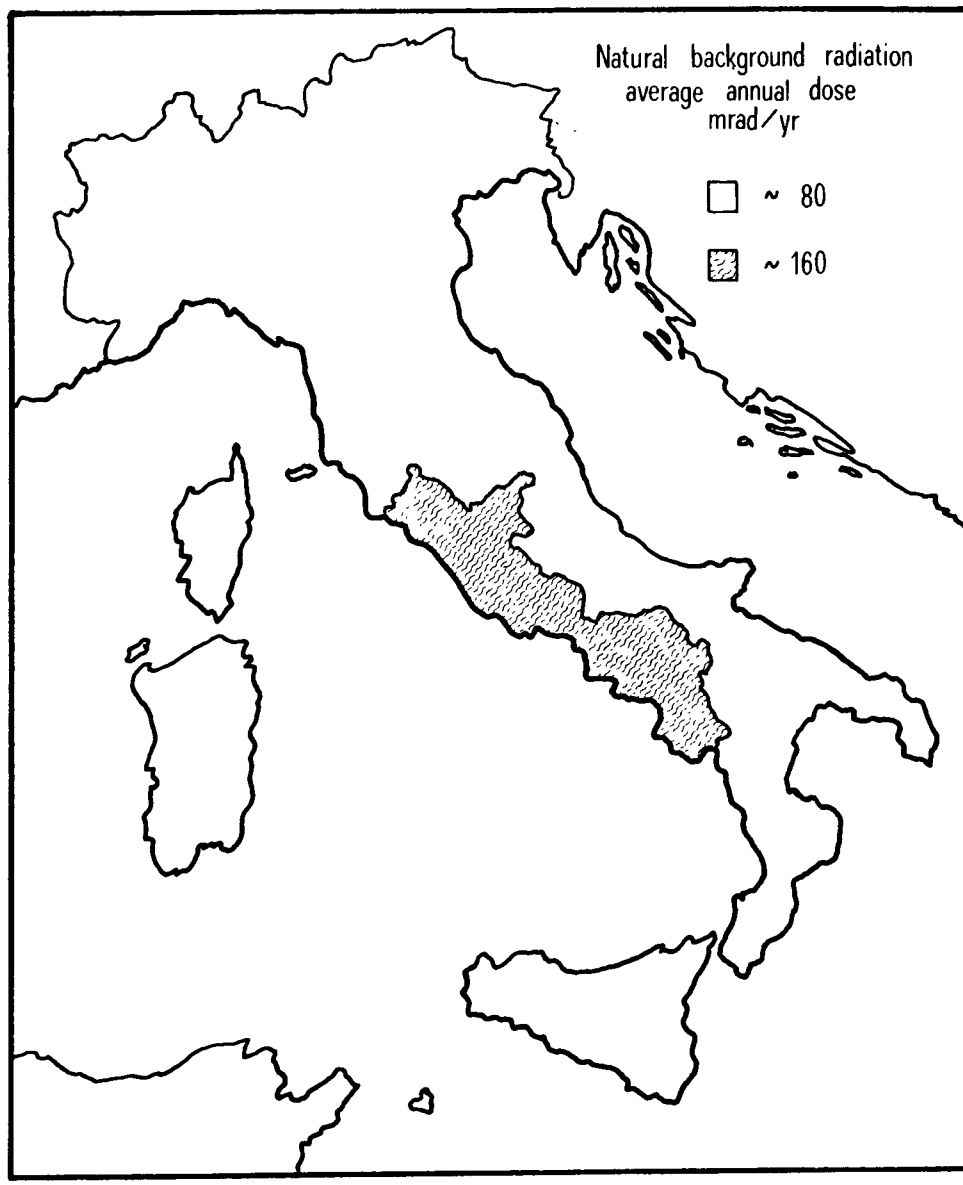
Natural background radiation
average annual dose
mrad/yr
~ 80
~ 160

Fig. 11

NATURAL RADIOACTIVITY IN SOME BRITISH WATERS

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SUMMARY. Measurements of naturally occurring radioactivity in British waters are given. Special reference is made to public water supplies although other waters are discussed. As is to be expected, some waters of underground origin contain most natural radioactivity. In their case the nuclide of greatest interest is radon-222 which, because of its ease of removal, does not present an intractable problem. Otherwise the human intake of naturally occurring radioactivity from water is very small and usually its radiological significance, if any, is not more than that from foodstuffs.

RESUME. RADIOACTIVITE NATURELLE DES EAUX BRITANNIQUES. On indique les résultats de mesures de la radioactivité naturelle des eaux en Grande-Bretagne. L'accent est mis sur l'eau des réseaux d'adduction quoique l'on considère aussi d'autres eaux. Comme on s'y attendait, certaines eaux souterraines présentent souvent une radioactivité naturelle. Le nucléide le plus intéressant est le radon-222 qui, facile à éliminer, ne pose pas un problème insoluble. Par ailleurs, l'incorporation humaine de radioactivité naturelle par l'eau est très faible et normalement, si elle a une signification radiologique elle n'est pas plus grande que par les denrées alimentaires.

KURZFASSUNG. NATÜRLICHE RADIOAKTIVITÄT IN EINIGEN BRITISCHEN GEWÄSSERN. Messergebnisse der natürlichen Radioaktivität in britischen Gewässern werden mitgeteilt. Das Hauptaugenmerk gilt dabei der öffentlichen Wasserversorgung, obwohl auch andere Gewässer erörtert werden. Erwartungsgemäss haben einige unterirdische Gewässer den grössten Gehalt an natürlicher Radioaktivität. Von besonderem Interesse ist dabei das Nuklid Radon-222, das, da es leicht zu beseitigen ist, keine unlösbaren Probleme aufwirft. Abgesehen davon ist die Aufnahme natürlich vorkommender Radioaktivität aus dem Wasser durch den Menschen sehr gering und ihre radiologische Bedeutung ist normalerweise nicht grösser als bei Nahrungsmitteln.

1. Water supplies in Britain are derived extensively from all three principal types of source, namely:

i. impounded waters

ii. abstraction from rivers

iii. underground. (See Table I)

Few people drink rainwater regularly. Some spa waters are drunk but they do not constitute what is usually meant by the term 'water supply'. Not surprisingly impounded waters contain only small amounts of naturally occurring radioactivity whilst some underground waters contain the most. This paper reviews and comments on such measurements as have come to the author's attention in the last twenty years. Much of the information quoted or referred to has been published by other authors in the technical literature; a little more comes from experience in the Radiochemical Inspectorate. Although tritium and ^{14}C are both naturally occurring, their man-made origins are of prime importance and so they are excluded from discussion here. There is an extensive literature on beta/gamma activity in water but the results are of course dominated by fall-out or other man-made activity; consequently in this review we are largely concerned with alpha emitting nuclides and their daughters. It must be borne in mind that where 'total alpha' figures are quoted they refer (unless otherwise stated) to activity measured after evaporation: radon has been driven off.

2. Impounded waters

Turner et al, (1) in a comprehensive survey reported 0.01 pCi to 0.20 pCi of radium per litre in surface waters, both impounded and rivers, excluding Cornwall. Other measurements since that time are in accordance. During the period when there was an extensive programme to monitor fall-out from weapons-testing a number of water supplies were examined for alpha activity as well as for beta emitters and the results were reported in a series of Government publications (2, 3, 4) from which the information in tables II, III and IV is obtained. Further unpublished results obtained by the Radiochemical Inspectorate confirm the general picture that as a rule the 'total alpha' activity in impounded waters is much below 1 pCi/litre, often being less than the limit of detection as determined by the counting statistics. On a few occasions measurements slightly above 1 picocurie per litre have been recorded eg

a. Cardiff water supply sampled during the second quarter of 1966, where water from impounding reservoirs in the mountains showed

0.42 pCi/l 'total alpha' but after passage through a long concrete pipe showed 1.4 pCi/litre. (4)

b. Belfast water supply (within the United Kingdom but not strictly speaking within Great Britain), second quarter 1964, 'total alpha' 1.73 pCi/litre. (2)

These Belfast samples were collected as water enters the aqueduct from the reservoirs in the Mountains of Mourne. A spot sample of the water on 17 August 1964 was measured at 2.18 pCi per litre but the radium content was shown to be much lower, the concentrations of radium-224 and of radium-226 being quoted as 0.095 and 0.25 pCi/litre respectively. Presumably impounding reservoirs which receive waters rapidly from bare hillsides may have a radioactive content much and immediately affected by natural activity in rain (it is known for example that ^{210}Pb in rain may reach about 3 pCi/litre) or by disturbance of sediment. Table V is given to show some recent typical results; it may be that it also illustrates the phenomenon just mentioned.

Potassium in impounded waters has been quoted in some specific detailed analyses (Refs 2, 3, 4). Eg

Lleyn (Cwmystradllyn) North Wales	0.22 pCi/l
Glasgow (Loch Katrine)	0.22 pCi/l
Leeds	1.23 pCi/l
Manchester (Haweswater)	0.45 pCi/l

It may be of interest to note that the total alpha activities were, descending the table, 0.13, 0.59, about 0.28 and about 0.43 pCi/litre respectively. It is not proposed to give here an extensive survey of the potassium content of waters, whether impounded or otherwise. The figures quoted above may be regarded as typical for impounded and are for comparison.

Radon measurement has been made in two extensive surveys: that of Turner, Radley and Mayneord to which reference has already been made (1) and that of Kenny et al (5) in 1962. The latter of these two was directed towards supplies in which radium and radon might be expected whereas the former was not. Even so, the highest radon concentration that was found by Kenny and his co-workers in reservoir water was 700 pCi/litre in a reservoir on granite. As is usually the case the radium content was much less: 1.0 pCi/litre of ^{226}Ra and 0.4 of ^{224}Ra.

3. Water supplies from rivers

One would expect two characteristic features to be shown by rivers:

i. Those that flow through good agricultural land and those which receive sewage from large populations ought to have enhanced potassium content.

ii. Radon ought to be considerably removed (if any present initially) by the natural agitation and aeration of the water.

Those expectations are realised in practice.

Thus in the granite areas of SW England where underground water often contains substantial quantities of radon the rivers have been reported to have a much lower content, for example some measurements by the National Radiological Protection Board (6) gave the following results:

Wolf River	70 pCi/litre
Thrushel River	70 pCi/litre
Tamar River	70 pCi/litre
Tavy River	50 pCi/litre

Kenny et al listed measurements in a number of streams and rivers used for water supply, some of them in granite areas. Their radon content ranged from 0 to 310 pCi/litre. (The same paper refers to a case where the feeder streams are at a lower concentration than is the water in the receiving impounding reservoir.) Radon values for surface waters generally, but excluding Cornish water, have been quoted as being in the range 400 pCi/litre (1).

Radium and alpha activity other than radon do not present any problems either. For example we have (7):

London drinking water (Thames derived): Radium 0.1 pCi/litre

London drinking water (Lee derived): Radium 0.12 pCi/litre

Numerous measurements of total alpha activity of the River Thames over the years have shown it to be less than 1 pCi/litre. At the instigation of the Radiochemical Inspectorate some other major rivers which provide water have been analysed by the Laboratory of the Government Chemist using daily samples bulked for a quarter (8). The results are shown in Table VI. The River Severn is at present being investigated. In the Cornish rivers and streams the highest concentration of ^{226}Ra reported by Kenny et al was 1.5 pCi/litre accompanied by 0.9 pCi/litre of ^{224}Ra.

4. Underground waters excluding spa water

Much interest has been focussed on the high radon-222 content of some underground waters, particularly those from within or near granite areas. Not all granite areas appear to produce large radon concentrations and it seems that the highly mineralised areas and their neighbourhoods are the most significant - for example Cornwall and Devon in SW England. In the survey by Kenny et al (5) the highest radon concentrations found in water sources used for supply were 21,500 and 19,000 pCi/litre, although one unused spring contained 29,100 pCi/litre. The same authors measured radon in tap waters sampled from all the public supplies drawn from granite areas where they had found more than 2000 pCi/litre in the water before being put into supply. The highest value found was 20,000 pCi/litre in the water provided to a single cottage; the others were all less than 7000 pCi/litre. The highest concentration of ^{226}Ra found in the supply waters was 4.7 pCi/litre. In recent years the National Radiological Protection Board has measured radon in Cornish mine waters, finding concentrations of up to about 5000 pCi/litre (6).

In areas other than Cornwall, ground waters from chalk strata have been shown to contain up to 1.22 pCi/litre of long lived alpha activity and up to 200 pCi/litre of radon, whilst ground waters from other than chalk strata showed long lived activity up to 2.9 pCi/litre and radon up to 700 pCi/litre (1).

The National Radiological Protection Board has given me an unofficial, provisional figure for the ALI by ingestion for ^{222}Rn of 2×10^8 Bq. It seems unlikely that such an intake would be achieved from the above concentrations, particularly because much of the water consumed in a household has been boiled before drinking, the radon thereby being driven off.

Indeed it seems that inhalation of radon is likely to be the critical risk, since much of the radon delivered in water to a household must be liberated by aeration (splashing in a sink or bath), heating for various purposes and boiling to drink and cook etc. According to the NEA (9) water having a concentration of 10^3 pCi/litre might add about 20% to the incremental exposure to radon daughters from representative building materials. It seems therefore that in most parts of Britain the increment from water is of little relative importance. In the worst areas the contribution from water might at the highest be a few times that from 'representative' building materials.

Uranium. Water from the Hilton Well, which is deep in Bunter sandstone and which supplies the town of Wolverhampton was found to have a total alpha activity of about 10.5 pCi/litre (Tables III and IV). Further examination showed the bulk of that activity to be due to uranium, ^{226}Ra being measured at no more than 0.6 pCi/litre. Some information on uranium in diet is given in reference (10).

5. Spa waters

Measurements (11) in the thermal spring waters of Bath have showed the ^{226}Ra content to be in the range 10.1–14.2 pCi/litre, the highest radon value being 2399 pCi/litre. Earlier measurements on various spa waters showed long lived alpha activity in the range 4.5 to 37 pCi/litre and radon up to 2000 pCi/litre (1). Some data obtained by the National Radiological Protection Board (6) on water from a British spa are similar to the foregoing and are shown in Table VII.

6. Removal of natural radioactivity from water

Improvement of water quality by the removal of the two most important nuclides is a fairly simple matter - radon by aeration, radium by a number of standard methods which are used to 'soften' water (12, 13).

In Britain there are no water supplies which are specially treated in order to remove radium.

Early in the last decade a water supply being developed in Devon was deemed to need removal of carbon dioxide to render it less "aggressive", and when it was discovered that water at the well-head contained a radon concentration of 15,000 pCi/litre it was decided to increase the efficiency of aeration in order to reduce radon levels (14, 15). It should be noted that when radon is removed deliberately by aeration it is necessary to consider the potential risk from radon in air.

7. Conclusion

Natural radioactivity levels in British water supplies are lower than some reported from other countries. In most parts of Britain the ingestion of natural radioactivity can be no more, and is probably considerably less than that from foodstuffs. (See for example data in refs 16, 17.) In this context it is worth remembering that potassium is accumulated in the human body. Because the quantity in an adult individual is more or less constant the concepts of maximum permissible concentration and ALI are not applicable, but the amount in the body is about 0.1 μCi, giving an estimated annual dose equivalent of 170 μSv (18, 19).

The inhalation of radon daughters from radon in water is not a serious problem: at the worst the dose to the bronchial epithelium and the whole lung could be of the same order of magnitude as that from building materials in extreme cases.

Neither radium nor radon in water need present an intractable problem, since radon can easily be removed by aeration should it be considered necessary and radium may be removed by some common water softening processes.

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TABLE I

APPROXIMATE DISTRIBUTION OF POPULATION BETWEEN THE THREE TYPES OF WATER SUPPLY IN THE UNITED KINGDOM

i.	Impounded Waters	21×10^6 people
ii.	Streams and Rivers	13×10^6 people
	(the figure includes about 5×10^6 obtaining water from the R Thames)	
iii.	Underground from wells and boreholes	19×10^6 people

[Information from reference 4]

TABLE II

Water	Results in picocuries/litre					Type
	Sr90	Cs137	Total Beta	Total Alpha	Potassium 40	
					(natural activity)	
East Devon, Wilmington Spring	0.0086	0.009	4.4	< 0.38	0.91	Spring
Lee Valley, North Mimms	1.53	0.051	12.9	< 0.51	4.16	Well in chalk
Bristol, Cheddar Spring	0.15	0.017	2.6	0.70	0.91	Spring
Scarborough, Irton Bore	0.75	0.045	6.0	< 0.41	1.29	Deep well
Scarborough, River Derwent	1.40	0.088	12.1	< 0.34	1.44	Recharge water

TABLE III

Supply	Period of sampling in 1964	Alpha activity (pc/litre)	
		Soon after preparation of source	Recount four weeks later
Impounded waters			
Belfast	2nd Qtr	1.73	-
Birmingham	2nd Qtr	0.25	-
Edinburgh	2nd Qtr	0.19	-
Glasgow	2nd Qtr	0.23	-
Hanningfield	4th Qtr	< 0.42	< 0.43
Leeds	4th Qtr	< 0.16	< 0.15
Liverpool	4th Qtr	< 0.09	0.11
Llandudno	4th Qtr	0.12	0.18
Lleyn	4th Qtr	< 0.08	0.16
Lurgan	2nd Qtr	< 0.20	-
Manchester	4th Qtr	0.14	0.11
Newcastle	4th Qtr	< 0.15	0.15
Newport	4th Qtr	< 0.11	0.15
Rhyl	4th Qtr	< 0.20	< 0.20
Rivers and springs			
Aberdeen	2nd Qtr	0.43	-
Coventry	3rd Qtr	< 0.48	< 0.65
Exeter	3rd Qtr	0.11	0.15
London	2nd Qtr	< 0.39	-
Norwich	3rd Qtr	< 0.49	< 0.48
Oxford	2nd Qtr	< 0.42	-
Southampton	3rd Qtr	0.95	2.70
Wells and boreholes			
Brighton	3rd Qtr	< 0.55	< 0.72
Folkestone	3rd Qtr	< 0.22	< 0.27
South Lincs	3rd Qtr	0.93	2.67
Wolverhampton	3rd Qtr	7.57	8.28
Wolverhampton	4th Qtr	10.5	10.5

TABLE IV

Water	Periods of sampling	Results in pCi/litre				Type
		Total alpha (1)	Total alpha (2)	Total beta	Potassium 40 (natural activity)	
East Yorks (Millington)	19/10/65– 29/11/65	< 0.47	< 0.58	< 1.95	0.34	Springs
East Yorks (Newbald)	12/7/65– 15/8/65	0.31±0.15	< 0.55	< 1.55	0.58	Borehole
Hull (Keldgate)	3/8/65– 31/8/65	0.4	–	< 2.1	0.72	Borehole
Lee Valley (North Mimms)	1/6/65 28/6/65	< 0.67	–	11.2±2.0	4.2	Well, in chalk
Mimms Hall Brook	1/6/65– 28/6/65	< 0.55	–	26.8±2.0	6.4	Brook
M.W.B. (Hoddesden)	20/12/65– 15/1/66	0.88±0.58	0.56±0.48	4.8±1.6	4.1	Well, in chalk
Sheffield (Rearne Valley)	6/7/65–	3.53±1.59	< 1.95	13.5±4.4	8.7	From a colliery

Notes. Total alpha (1) The alpha activity measured soon after preparation of the source.

Total alpha (2) The alpha activity measured at least 4 weeks after preparation of the source.

TABLE IV (Cont)

Water	Periods of sampling	Results in pCi/litre				Type
		Total alpha (1)	Total alpha (2)	Total beta	Potassium 40 (natural activity)	
South Derbyshire (Belper Meadows)	7/9/65– 7/10/65	< 0.52	< 0.52	2.14±1.60	2.12	Borehole
South Derbyshire (Meerbrook Sough)	16/9/65– 17/10/65	1.47±0.62	2.02±0.64	3.8 ±2.2	0.76	Drainage tunnel
S. W. Devon (Ranney Collector)	28/6/65–	0.72±0.13	0.60±0.13	5.7 ±0.8	0.87	Shallow well close to R Dart
River Dart (near Totnes	28/6/65– 29/7/65	1.01±0.16	1.25±0.17	9.4 ±0.7	0.79	
Wolverhampton (Hilton)	1st Qtr 1965	–	11.1 ±0.9	9.2	4.2	Deep well
	3rd Qtr 1965	11.1 ±0.8	11.4 ±1.1	–	–	
	4th Qtr 1965	9.0 ±0.6	9.0 ±0.7	–	–	

Notes. Total alpha (1) The alpha activity measured soon after preparation of the source.

Total alpha (2) The alpha activity measured at least 4 weeks after preparation of the source.

TABLE V

TOTAL ALPHA ACTIVITY IN SOME LAKE WATERS IN THE ENGLISH LAKE DISTRICT - 1978

Lake	Date	Activity in pCi/litre	Remarks
Buttermere	28 February	0.9	
Wastwater	28 February	1	Used as Water Supply
	19 October	0.6	" " " "
Crummock Water	28 February	2 ± 1	
Loweswater	28 February	2 ± 1	
	16 May	0.4	
Ennerdale Water	16 May	0.4	Used as Water Supply
Whitehaven tap water	28 February	0.8	(from Ennerdale Water)

TABLE VI

RADIOACTIVITY IN RAW RIVER WATERS (ANNUAL MEAN OF QUARTERLY SAMPLES)

River	Gross α activity Becquerels/ litre	Uranium Becquerels/ litre	Gross β activity Becquerels/ litre	Potassium-40 Becquerels/ litre	Radiocaesium Becquerels/ litre	Radio-strontium Becquerels/ litre	Tritium Becquerels/ litre
Tyne 1975	ND	2 x 0.037	ND	1.5 x 0.037	0.1 x 0.037	1.1 x 0.037	ND
*Ouse 1976/7	4 x 0.037	2 x 0.037	20 x 0.037	8 x 0.037	0.04 x 0.037	0.16 x 0.037	144 x 0.037
Lune 1977/8	1.5 x 0.037	2 x 0.037	9 x 0.037	1.5 x 0.037	0.04 x 0.037	0.83 x 0.037	150 x 0.037

ND = Not Determined

Reproduced from Reference 8

*Note: This river is the Ely Ouse.

TABLE VII

Radiocuclide	Activity Concentration Typical	pCi/l Large
^{226}Ra	5	5-7[b]
^{222}Rn	2000[c]	1700-2200[c]
^{210}Pb	200	150-230[d]

a. Small number of samples.

b. Filtered sample 3.6 pCi/l.

c. At time of collection.

d. Filtered sample 20 pCi/l.

NATÜRLICHE RADIOAKTIVITÄT IN TRINKWASSER, NAHRUNGSMITTELN UND IM MENSCHEN IN DEUTSCHLAND

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KURZFASSUNG. Erste relevante Ergebnisse über den natürlichen Radiumgehalt und das Ra-226/Ca-Verhältnis in Nahrungsmitteln, Trinkwasser und verschiedenen Organen und Geweben des menschlichen Körpers in Deutschland wurden vor mehr als 20 Jahren veröffentlicht (H. Muth, A. Schraub, K. Aurand, H.J. Hantke, 1957; H. Muth, B. Rajewsky, H.J. Hantke, K. Aurand, 1960). Für die mittlere tägliche Radiumaufnahme des Menschen ergab sich damals der Wert 3 pCi. Neuere Messungen der Ra-226-Konzentration im Trinkwasser ergaben in guter Übereinstimmung mit den früheren Messwerten Konzentrationen zwischen 0,02 bis 1 pCi/Liter, vereinzelt können auch Werte bis zu einigen pCi/Liter auftreten. Die Ra-226-Konzentration im Mineralwasser liegt im Mittel deutlich höher (1-20 pCi/Liter).
Die Wässer einiger untersuchter Flüsse und Seen weisen dagegen Ra-226-Konzentrationen von 0,03 bis 0,8 pCi/Liter auf (I.Gans, J. Porstendörfer, 1977, 1978). Zum Teil wesentlich höhere Werte sowohl in Quell- und Oberflächenwässern als auch in lokal produzierten Nahrungsmitteln wurden dagegen in Uranabbaugebieten (Schwarzwald) gemessen. Die Radium-Gesamtkörperaktivität von Menschen aus solchen Bereichen überschreitet jedoch nicht den landesweiten Mittelwert von etwa 10^{-10} Ci (H. Kiefer, 1979).
Es wird über neue Messergebnisse der Ra-226-, Pb-210- und Po-210-Konzentration in menschlichen Knochenproben und die Altersabhängigkeit der Ra-Konzentration im Vergleich zur Konzentration von Sr-90 berichtet (B. Glöbel, H. Muth). Ergebnisse von Messungen der Konzentrationen von Th-232 und Th-228 in menschlichen Gewebeproben (Stahlhofen, 1964) werden den Radiumwerten gegenübergestellt.

SUMMARY. NATURAL RADIOACTIVITY IN DRINKING WATER, FOODSTUFFS AND MAN IN GERMANY. The first significant results in respect of the natural radium content and the Ra-226/Ca relationship in foodstuffs, drinking water and various human organs and tissues in Germany were published more than 20 years ago (H. Muth, A. Schraub, K. Aurand, H.J. Hantke, 1957: H. Muth, B. Rajewsky, H.J. Hantke, K. Aurand, 1960). At that time the average individual's daily intake of radium was found to be 3 pCi. More recent measurements of the Ra-226 content of drinking water have produced values close to those measured earlier, between 0.2 and 1 pCi/litre, with

values of up to several pCi/litre being found in individual cases. The average Ra-226 concentration in mineral water is considerably higher, (1-20 pCi/litre). While, by way of contrast, the water of several of the rivers and lakes investigated showed Ra-226 concentrations of 0.03-0.8 pCi/litre (I. Gans, J. Porstendörfer, 1977, 1978) the values were higher (in some cases considerably higher) in spring-water, surface waters and locally produced foodstuffs in uranium mining areas (Black Forest). However, the whole-body radium burden in such areas does not exceed the national average of about 10^{-10} Ci (H. Kiefer, 1979). New results are presented for Ra-226, Pb-210 and Po-210 concentrations in human bone samples and for the influence of the age on the relationship between Ra and Sr-90 concentration values (B. Glöbel, H. Muth). Results of measurements of Th-232 and Th-228 concentrations in samples of human tissues (Stahlhofen, 1964) are compared with the values for radium.

RESUME. RADIOACTIVITE NATURELLE DE L'EAU POTABLE, DES ALIMENTS ET DE L'ORGANISME, EN ALLEMAGNE. Les premiers résultats pertinents de mesures, en Allemagne, de la teneur naturelle en radium et du rapport des éléments Ra-226/Ca contenus dans les aliments, l'eau potable et les différents organes et tissus du corps humain, ont été publiés il y a plus de 20 ans (H. Muth, A. Schraub, K. Aurand, H.J. Hantke, 1957; H. Muth, B. Rajewsky, H.J. Hantke, K. Aurand, 1960). La valeur quotidienne moyenne, alors mesurée, de l'ingestion de radium par l'homme était de 3 pCi. Des mesures récentes de la teneur de l'eau potable en Ra-226 ont permis d'établir des concentrations comprises entre 0,02 et 1 pCi/litre, chiffres qui concordent avec les résultats antérieurs; dans des cas isolés, les valeurs obtenues peuvent aller jusqu' à quelques pCi/litre. La concentration de Ra-226 dans l'eau minérale est en moyenne nettement supérieure (1 à 20 pCi/litre).

Les eaux de quelques-uns des cours d'eau et des lacs étudiés contiennent des concentrations en Ra-226 allant de 0,03 à 0,8 pCi/litre (I. Gans, J. Porstendörfer, 1977, 1978). Dans des régions d'exploitation de l'uranium (Forêt-Noire), des concentrations parfois considérablement supérieures ont été mesurées, tant dans les eaux de source et de surface que dans les aliments de la production locale. Toutefois, l'activité du radium dans tout l'organisme des habitants de ces régions ne dépasse pas la moyenne nationale, qui est d'environ 10^{-10} Ci (H. Kiefer, 1979). Le rapport donne de nouveaux résultats de mesures des concentrations de Ra-226,

Pb-210 et Po-210 dans des échantillons d'os humains, ainsi que la relation de dépendance entre l'âge et la concentration en radium, comparée à la concentration en Sr-90 (B. Glöbel, H. Muth). Les résultats de mesures des concentrations de Th-232 et de Th-228 dans des échantillons de tissus humains sont confrontés aux valeurs obtenues pour le radium (Stahlhofen, 1964).

Erste orientierende Untersuchungen und Messungen zur Frage der natürlichen Radioaktivität des menschlichen Körpers wurden in Deutschland auf Veranlassung von B. Rajewsky bereits in den Dreißiger Jahren von A. Krebs durchgeführt (1, 2, 3). Krebs hat diese Messungen sowohl an Proben von Krematoriumsaschen als auch an Proben von veraschten Organen und Geweben vorgenommen. Als Ergebnis von Messungen an Proben von insgesamt 18 "Normalpersonen" gibt er für den Gesamtkörpergehalt an ^{226}Ra 1×10^{-9} bis 4×10^{-8} g Ra an mit einem Mittelwert von $1{,}4 \times 10^{-8}$ g Ra. Wie spätere, von anderen Autoren durchgeführte Messungen zeigten (siehe Abschnitt 3), lag dieser Wert für ^{226}Ra zu hoch. Die Gründe hierfür dürften einmal in der relativ kleinen von Krebs erfaßten Personenzahl, zum anderen vor allem aber in der Meßmethode zu suchen sein. Krebs hat die vorher entsprechend präparierten Aschenproben in einen "Spitzenzähler" eingeführt und die "gesamte Alphastrahlenaktivität" (also nicht nur ^{226}Ra + Folgeprodukte!) gemessen. Außerdem war der damals verwendete selbstgebaute Spitzenzähler ein sehr subtiles, störanfälliges Gerät, dessen Bedienung nicht einfach war. Trotzdem gaben diese Pionierarbeiten von Krebs in Deutschland erste Hinweise auf die Größenordnung der gesamten natürlichen Alphastrahlenaktivität im Körper des Menschen.

1. Natürliche Radioaktivität in Trinkwasser.

a) Ältere Messungen.

Erste relevante Ergebnisse über den natürlichen Radiumgehalt und das $^{226}Ra/Ca$ - Verhältnis in Trinkwasser in Deutschland wurden vor mehr als 20 Jahren veröffentlicht (4, 5, 6).

In Tabelle I sind einige Ergebnisse dieser älteren Messungen für Leitungswasser (Trinkwasser), in Tabelle II für Quellwässer sowohl aus tiefen (400 - 500 m) als auch aus weniger tiefen (30 - 50 m) Quellen aus verschiedenen Gegenden Deutschlands und in Tabelle III aus vier deutschen Flüssen zusammengestellt. Der gemessene ^{226}Ra-Gehalt von Trinkwasser (0,03 - 0,34 pCi/l) und Quellwasser (10 - 20 pCi/l für tiefe Quellen und 0,07 - 0,5 pCi/l für weniger tiefe Quellen) stand in guter Überein-

stimmung mit Werten, die damals in verschiedenen Bereichen der USA gemessen worden waren. Die Werte der Flußwässer und der Quellwässer aus weniger tiefen Quellen lagen zwischen 0,07 und 0,8 pCi/l, also im gleichen Bereich wie beim Leitungswasser. In Tabelle I ist auch ein Wert für Badgastein/Österreich genannt, der erkennen läßt, daß dort im normalen Leitungswasser kein erhöhter ^{226}Ra-Gehalt vorliegt.

b) Neuere Messungen.

Auf Veranlassung des Bundesministers des Innern und mit dessen Unterstützung wurden während der letzten Jahre (1977/78) zunächst stichprobenartig Messungen des Ra-Gehaltes von Trink-, Quell- und Oberflächenwässern von den folgenden beiden Stellen durchgeführt:

Institut für Wasser-, Boden- und Lufthygiene des Bundesgesundheitsamtes Berlin und

Institut für Biophysik, Strahlenzentrum der Justus-Liebig-Universität Gießen.

Außerdem wurden im Jahre 1978 von einer Arbeitsgruppe des Kernforschungszentrums Karlsruhe systematische Untersuchungen und Messungen in der unmittelbaren und weiteren Umgebung einer Uranuntersuchungsgrube im Schwarzwald (Menzenschwand im Gebiet des Feldberges) als Muster für einen Bereich mit erhöhter natürlicher Radioaktivität vorgenommen. Das Meßprogramm des Instituts für Wasser-, Boden- und Lufthygiene des Bundesgesundheitsamtes Berlin wurde so angelegt, daß einerseits größere Wasserversorgungen erfaßt wurden, in denen im allgemeinen aufgrund des verwendeten Rohwassers und spezieller Aufbereitungsverfahren niedrigere Ra-Konzentrationen zu erwarten sind, und daß andererseits in bestimmten Gegenden mit wahrscheinlich höheren Konzentrationen gezielte Probenahmen auch bei kleineren Wasserwerken durchgeführt wurden (7, 8). Insgesamt wurden zunächst mehr als 300 Wasserproben [Leitungswasser (Trinkwasser), Oberflächenwasser (Flüsse, Kanäle, Seen, Abwässer), Quell-, Brunnenwasser] auf ^{226}Ra und z. T. auch auf ^{222}Rn untersucht. Repräsentativ für Wasserversorgungen mit Aufbereitungsverfahren sind die gemessenen Radiumkonzentrationen in München mit

0,05 pCi/l, in Berlin mit 0,1 bis 0,3 pCi/l und in Bonn und Marburg mit 0,2 pCi/l. Um die Gegenden mit zu erwartenden höheren Konzentrationen zu lokalisieren, wurden in einer Studie die Regionen der Bundesrepublik ermittelt, deren geologischer Untergrund relativ hohe Konzentrationen an Uran aufweist (Schwarzwald, Franken, nordostbayerisches Grundgebirke, Saar-Nahe-Senke, südliches Niedersachsen) (Abb. 1). Im Fichtelgebirge wurde eine größere Probenahme durchgeführt. Tabelle IV zeigt einige Ergebnisse dieser Messungen. Die höchsten Konzentrationen sowohl für ^{226}Ra mit 2,3 pCi/l bzw. 1,5 pCi/l als auch für ^{222}Rn mit 7,9 nCi/l bzw. 7,0 nCi/l wurden in den Orten Fichtelberg und Kirchenlamitz gefunden. Eine noch höhere ^{226}Ra-Konzentration, nämlich 4 pCi/l, konnte bisher nur bei einer Einzelwasserversorgung in der Oberpfalz/Bayern festgestellt werden. Eine erste Untersuchung zur Frage der Reduktion des Radiumgehaltes durch die Trinkwasseraufbereitung wurde in zwei Berliner Wasserwerken durchgeführt. Wie sich aus Tabelle V ergibt, wird der ^{226}Ra-Gehalt durch Belüftung (Versprühung) des Wassers und anschließende Filterung um den Faktor 2 bis 4 reduziert, während der ^{222}Rn-Gehalt beim Belüften zunächst herabgesetzt und im Filter allerdings durch den Zerfall des sich dort absetzenden ^{226}Ra wieder erhöht wird.

Von der Arbeitsgruppe des Instituts für Biophysik, Strahlenzentrum der Universität Gießen, wurden zunächst schwerpunktmäßig Untersuchungen im Bundesland Hessen vorgenommen (9, 10, 11). Insgesamt wurden Messungen an 205 aus dem Leitungsnetz der öffentlichen Wasserversorgung entnommenen Proben durchgeführt. Die Werte liegen zwischen 0,01 pCi/l und 3 pCi/l. Eine histographische Darstellung der Ergebnisse (Abb. 2) zeigt eine asymmetrische Verteilung. Es ist daher nicht zweckmäßig, einen arithmetischen Mittelwert anzugeben, sondern besser den Medianwert, d. h. den Wert, der die Gesamtheit der Stichproben in zwei gleich große Klassen teilt. Er liegt für das untersuchte Trinkwasser bei 0,11 pCi/l. Besonders hohe Konzentrationen wurden in Arolsen (3,0 pCi/l), Rüdesheim (1,38 pCi/l) und Montabaur (1,14 pCi/l) gefunden. Es wird darauf hingewiesen, daß diese weit über dem Durchschnitt liegenden Werte u. U. dadurch zu erklären sind, daß diese Orte ihr Trinkwasser zum Teil aus

Tiefbrunnen beziehen. Außer Leitungswasser wurden von der Gießener Gruppe auch Mineralwässer, Quellwässer, Brunnenwässer, Trinkheilwässer sowie Proben aus Flüssen und Binnenseen untersucht. Während die Werte für Quell-, Brunnen-, Fluß- und Seewasser im Bereich der Werte für Leitungswasser liegen, weisen Mineralwässer (in Flaschen) und Trinkheilwässer um mehr als das zehnfache höhere Werte auf. Tabelle VI gibt eine Zusammenfassung der Werte für die verschiedenen untersuchten Wässer.

Von der Arbeitsgruppe des Kernforschungszentrums Karlsruhe wurde die Strahlenexposition der Bevölkerung in der Umgebung einer Uranuntersuchungsgrube in Menzenschwand im Schwarzwald durch natürliches und aus der Grube emittiertes ^{226}Ra untersucht (12). Unter anderem wurden hierbei die ^{226}Ra-Konzentrationen in der Umwelt und in Nahrungsmitteln des Südschwarzwaldes bestimmt. Die hierdurch bedingte Strahlenexposition der Bevölkerung wurde abgeschätzt. Ebenso wurden Transferfaktoren für die wichtigsten Transportprozesse des Radiums in der Biosphäre angegeben. Tabelle VII zeigt die ^{226}Ra-Konzentrationen im Abwasser der Uranuntersuchungsgrube, im Bach ("Krunkelbach") vor und nach Einleitung der Grubenwässer und auch im Trinkwasser von Menzenschwand, das aus mehreren Quellen des Menzenschwander Tales stammt, keine Verbindung zum Grubenwasser hat und daher normale Werte aufweist. Außerdem wurden die ^{226}Ra-Konzentrationen von weiteren Quell- und Oberflächenwässern im Menzenschwander Tal und in der näheren und weiteren Umgebung des Tales in einer großen Zahl von Proben bestimmt. Der höchste singuläre Wert lag bei 1549 pCi/l ^{226}Ra! ("Wüstengraben, Quelle 2"). Zum Teil wurden auch relativ hohe Radonkonzentrationen in diesen Wässern gemessen (20 - 33 nCi/l ^{222}Rn!) ("Wüstengraben, Quelle 1 und Quelle 2").

Tabelle VIII bringt eine Zusammenstellung der mittleren Aktivitätskonzentrationen derjenigen natürlichen Radionuklide, die vornehmlich für eine mögliche Strahlenexposition bei Ingestion mit dem Trinkwasser in Frage kommen (13,14). Da die Konzentrationen der Nuklide der Uran-Radiumreihe sehr schwanken, sind deren Mittelwerte als Angabe der Größenordnung zu verstehen. Zweifellos kommt von den Radionukliden der Uran-Radiumreihe dem ^{226}Ra mit Abstand die größte Bedeutung zu.

2. Natürliche Radioaktivität in Nahrungsmitteln

Auch zur Ermittlung der natürlichen Radioaktivität in Nahrungsmitteln wurden erste Untersuchungen in Deutschland bereits vor mehr als 20 Jahren durchgeführt (4, 5, 6). In den Tabellen IX, X und XI sind einige dieser älteren Ergebnisse zusammengestellt. Die ^{226}Ra-Konzentration variiert zwischen o,1 und 6,0 fCi/g Ausgangsmaterial. Aufgrund dieser Meßergebnisse wurde damals die mittlere tägliche Radiumaufnahme durch den Menschen unter "normalen" Umweltbedingungen auf 3 pCi geschätzt, wobei etwa 90 % dieser Aktivität durch die Nahrung und etwa 10 % durch das Trinkwasser zugeführt werden. Auch diese Daten standen in befriedigender Übereinstimmung mit den Angaben amerikanischer Autoren.

Während Radium, Thorium und Uran ausschließlich aus dem Boden unmittelbar in pflanzliche Nahrungsmittel gelangen, gibt es für ^{210}Pb (RaD) auch die Möglichkeit der Aufnahme aus der Atmosphäre, in der es als Folgeprodukt des natürlicherweise in der Luft vorhandenen ^{222}Rn gebildet wird. Die Pflanzen nehmen das ^{210}Pb nicht aus dem Boden auf, sondern über die oberirdischen Pflanzenteile, da es sich als "natürlicher fallout" anbietet. Sowohl die kurzlebigen Folgeprodukte des ^{222}Rn [^{218}Po (RaA), ^{214}Pb (RaB), ^{214}Bi (RaC), ^{214}Po (RaC') und ^{210}Tl (RaC'')] als auch ^{210}Pb lagern sich an die in der Atmosphäre vorhandenen Aerosole an. Die Aerosole werden auf den Pflanzen abgelagert und so kommt es zur Aufnahme der kurzlebigen Folgeprodukte bzw., wenn die Sedimentationsgeschwindigkeit klein genug ist, zur Aufnahme des ^{210}Pb. So wurden in der pflanzlichen Nahrung relativ hohe Aktivitäten von ^{210}Pb und seinem Folgeprodukt ^{210}Po nachgewiesen (15, 16). Vor allem B. Glöbel (17, 18) hat umfangreiche Untersuchungen hierzu durchgeführt. In Tabelle XII sind einige seiner Ergebnisse zusammengestellt. Sie geben einen Überblick über die durchschnittliche tägliche Aufnahme von ^{210}Po und ^{210}Pb durch einen "Normalverbraucher". Die Meßergebnisse sind, bedingt durch die Art der Probenentnahme, Mittelwerte und können daher von den an Einzelproben ermittelten Werten beträchtlich abweichen. Es zeigt sich, daß Trinkwasser, Bier und Wein gegenüber der Milch und den festen Nahrungsmitteln

bei der Bilanz der ^{210}Pb- und ^{210}Po-Zufuhr vernachlässigt werden können. Der abgeschätzte Mittelwert für die tägliche Aufnahme von 4,6 pCi für beide Nuklide wurde noch dadurch überprüft, daß die Ausscheidung bei einer Einzelperson über längere Zeit gemessen wurde. Die für die Ausscheidung im Stuhl gefundenen Werte von 1,1 bis 9,5 pCi/Tag geben eine Bestätigung des Mittelwertes für die Aufnahme. Auch hier bestand gute Übereinstimmung mit amerikanischen und britischen Angaben. Die Bedeutung der beiden natürlichen Radionuklide ^{210}Pb und insbesondere ^{210}Po für die natürliche Strahlenexposition des Menschen war damit offensichtlich. Auch aus anderer Sicht, im Hinblick auf die öffentlichen Diskussionen über die Möglichkeiten einer Gefährdung der Menschen durch nicht radioaktives Blei in der Umwelt, kommt den Kenntnissen über das Verhalten des radioaktiven Blei eine besondere Bedeutung zu (vgl. auch 19).

Im Rahmen der bereits in Abschnitt 1 zitierten Untersuchungen im Schwarzwald (12) wurde u.a. die ^{226}Ra-Konzentration von Forellen aus den Bächen (Krunkelbach und Menzenschwander Alb), in die das Abwasser der Uranuntersuchungsgrube eingeleitet wird, gemessen. Der ^{226}Ra-Gehalt der Fische des Krunkelbaches liegt bei 60 pCi/kg Fischfleisch und der Fische der Menzenschwander Alb bei 5 pCi/kg. Die ^{226}Ra-Aktivität in Fischen ist zu je ein Drittel auf die Eingeweide, das Skelett und das Fleisch verteilt. Außerdem wurde die ^{226}Ra-Konzentration in Heu und Gras (11 Proben), Pflanzen (8 Proben), Lebensmitteln (21 Proben), Wurzelerde verschiedener pflanzlicher Nahrungsmittel in verschiedenen Bodentiefen (8 Proben) und vor allem in Milch gemessen. Für Menzenschwand ergab sich ein Mittelwert von 1o,2 pCi/l Milch während in Milchproben von Landwirten aus der Umgebung von Menzenschwand Werte zwischen 0,33 bis 32,0 pCi ^{226}Ra/l gefunden wurden. In Tabelle XIII sind die Ergebnisse der ^{226}Ra-Konzentrationen in Molkereimilch angegeben. Während Freiburg und Karlsruhe normal niedrige ^{226}Ra-Konzentrationen aufweisen, lassen die Ergebnisse für Karlsruhe/Südschwarzwald und Waldshut/Tingen den Einfluß des radiumreichen Granits des Südschwarzwaldes erkennen. In Tabelle XIV sind die wichtigsten im Südschwarzwald gemessenen Werte der ^{226}Ra-Konzentrationen (12)zum Vergleich mit Angaben aus der Literatur (13) zusammengestellt.

Es zeigt sich, daß sie ausnahmslos höher liegen als die Vergleichswerte.

3. Natürliche Radioaktivität im Menschen

Schon bei den bereits zitierten ersten größeren Untersuchungsreihen zur Frage des natürlichen ^{226}Ra-Gehaltes des Menschen und seiner Umgebung (4, 5, 6) wurden neben Untersuchungen an Krematoriumsasche auch Messungen an menschlichen Gewebeproben (Knochen, Weichteilgewebe, auch Testis, Placenta und Foetus) durchgeführt. Für die ^{226}Ra-Konzentration im Knochen ergab sich ein Mittelwert von 5 fCi/g Frischgewicht, entsprechend etwa $1{,}2 \cdot 10^{-14}$ Ci pro 1 g Knochenasche. Unter Berücksichtigung der gemessenen mittleren Konzentrationen in Organen und im Weichteilgewebe (1,1 fCi/g Frischgewicht) ließ sich für den Gesamtradiumgehalt des Menschen ("Reference Man") ein Mittelwert von $1{,}3 \cdot 10^{-10}$ Ci abschätzen in guter Übereinstimmung mit amerikanischen Meßwerten (Hursh und Gates; Stehney und Lucas).

Von W. Stahlhofen (20, 21, 22) wurden sehr subtile und umfangreiche Untersuchungen zur Ermittlung des Gehaltes des menschlichen Körpers an ^{226}Ra, ^{228}Th und ^{210}Po durchgeführt. Messungen des ^{226}Ra-Gehaltes von Knochen und Organen in Abhängigkeit vom Alter sollten Aufschluß geben über den Einbau von ^{226}Ra in den menschlichen Organismus bei Dauerzufuhr kleiner Mengen. Bei menschlichen Knochen verschiedenen Alters (Knochen von Totgeburten bis zu Knochen von 80 Jahre alten Menschen) sowie bei Foetenknochen zwischen Mens IV und Mens X wurde keine Korrelation zwischen ^{226}Ra-Gehalt und Alter gefunden. Sowohl für Knochen von erwachsenen Menschen als auch für Foetenknochen ergab sich die gleiche ^{226}Ra-Konzentration in der Knochenasche von $1{,}3 \cdot 10^{-14}$ Ci/g. Die ^{226}Ra-Konzentration des Weichteilgewebes zeigt sich ebenfalls als unabhängig vom Alter und betrug im Mittel 0,1 fCi/g Frischgewicht bzw. $7 \cdot 10^{-13}$ Ci/g Ca. Für den "Reference Man" errechnet sich daraus ein ^{226}Ra-Gehalt pro Gesamtkörper von $0{,}4 \cdot 10^{-10}$ Ci,von dem sich etwa 85 % im Skelett und 15 % im Weichteilgewebe befinden. Dieser Wert stimmt überein mit ebenfalls von W. Stahlhofen sehr sorgfältig durchge-

führten Messungen an Krematoriumsaschen, nach denen der ^{226}Ra-Gehalt pro Gesamtkörper im Mittel bei $0{,}47 \cdot 10^{-10}$ Ci liegt. Der von Stahlhofen gefundene Gesamt-Radiumgehalt des Menschen liegt somit deutlich niedriger als der oben zitierte Wert der älteren deutschen und amerikanischen Untersuchungen. Der Verfasser führt diese Diskrepanz darauf zurück, daß die früher gemessenen Krematoriumsaschen Anteile des Krematoriumsbodens enthielten, dessen ^{226}Ra-Konzentration nach amerikanischen Messungen mit 5 pCi/g relativ hoch ist. Eigene Untersuchungen von Stahlhofen an Steinstaubproben ergaben ^{226}Ra-Konzentrationen der gleichen Größenordnung.Messungen von placentarem Gewebe ergaben eine ^{226}Ra-Konzentration von 0,16 fCi/g Frischgewicht bzw. $3{,}5 \cdot 10^{-13}$ Ci/g Ca. Hieraus wird gefolgert, daß die Placenta für ^{226}Ra keine Schranke darstellt.

In weiteren Messungen wurde der ^{210}Po- und ^{210}Pb-Gehalt von Femur- und Tibiaproben und das Aktivitätsverhältnis von ^{210}Po zu ^{210}Pb im menschlichen Skelett bestimmt. Die Mittelwerte betragen $3{,}7 \cdot 10^{-14}$ Ci ^{210}Pb/g Frischgewicht bzw. $3{,}1 \cdot 10^{-14}$ Ci ^{210}Po/g Frischgewicht. Daraus ergibt sich ein Aktivitätsverhältnis von ^{210}Po zu ^{210}Pb im Skelett des Menschen von 0,8. 60 % des insgesamt im Körper vorhandenen ^{210}Po ist im Skelett abgelagert. Für die ^{228}Th-Konzentration von menschlichen Knochenaschen ergab sich im Mittel 4 fCi/g und für das Verhältnis ^{228}Th zu ^{226}Ra im menschlichen Knochen im Mittel 0,4.

Eine umfangreiche und systematische Studie zum Problem der natürlichen und künstlichen Radioaktivität des Menschen liegt von E. Oberhausen vor (23): Seine Untersuchungen geben Aufschluß über den Gesamtgehalt des Menschen an ^{226}Ra, ^{228}Ra, ^{228}Th, ^{210}Pb, ^{210}Po, ^{40}K und ^{137}Cs und ihre Verteilung im Körper. Die Ergebnisse stimmen, wenn man die Fehlerbreiten der angewandten Meßmethoden und die beträchtlichen biologischen Schwankungsbreiten berücksichtigt, relativ gut mit den von Stahlhofen angegebenen Mittelwerten überein. Beide Autoren nehmen eine Abschätzung der durch ^{226}Ra, ^{228}Th und ^{210}Po bedingten natürlichen Strahlenexposition von Knochen und Weichteilgewebe vor. Die Werte sind zum Vergleich in Tabelle XV zusammengestellt. Sie enthält auch die von E. Oberhausen berechneten Werte der natürlichen Strahlenexposition durch ^{40}K. Es zeigt sich, daß die durch ^{226}Ra und die im

Skelett verbleibenden 30 % seiner kurzlebigen Folgeprodukte bedingte Strahlendosis etwa gleich groß ist der durch ^{228}Th und seine Folgeprodukte. Die Dosis durch die ^{210}Po-Konzentration im Knochen ist dagegen etwa 3 mal höher als die durch ^{226}Ra und seine kurzlebigen Folgeprodukte. Von E. Oberhausen wurde auch an einer großen Zahl von Personen die Altersabhängigkeit des Kaliumgehaltes getrennt nach Geschlechtern über die Gammastrahlung des ^{40}K gemessen (23, 24, 25). Abbildung 3 zeigt die Ergebnisse. Man sieht, daß die Kaliumkonzentration bei Kindern am größten ist und dann mit zunehmendem Lebensalter abnimmt. Das bei Jüngeren im Alter von 15 Jahren auftretende Maximum ist auf die stärkere Ausbildung der Skelettmuskulatur während der Pubertät zurückzuführen (siehe auch 26). Zusammenfassende Darstellungen zum Problem der internen natürlichen Strahlenexposition des Menschen finden sich auch in den folgenden deutschen Veröffentlichungen: (27, 28, 29, 30, 31, 32, 33).

Abschließend sei noch auf zwei neuere Untersuchungen zur Frage der natürlichen Strahlenexposition durch ^{226}Ra eingegangen. Im Rahmen der bereits in den Abschnitten 1 und 2 diskutierten Studie zur natürlichen Radioaktivität im Südschwarzwald (12) wurden aus den Ergebnissen der Messungen der ^{226}Ra-Konzentrationen in Boden, Wasser und Nahrungsmitteln (siehe Tabelle XIV) die verschiedenen Transferfaktoren[+] berechnet. In Tabelle XVI sind die jeweiligen Mittelwerte zusammengestellt. Aus den gemessenen ^{226}Ra-Konzentrationen der Nahrungsmittel und des Trinkwassers und dem jeweiligen pro Kopf-Verbrauch (entnommen aus dem Ernährungsbericht 1976 der Deutschen Gesellschaft für Ernährung, Frankfurt a.M.) errechnet sich unter der Annahme, daß nur die lokal produzierten Nahrungsmittel der Ernährung dienen, eine mittlere Jahresaufnahme von 7,0 nCi ^{226}Ra. Nach der gültigen Deutschen Strahlenschutzverordnung beträgt der Grenzwert der jährlichen Aktivitätszufuhr für ^{226}Ra durch

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[+]Transferfaktoren sind die Verhältnisse der Aktivitätskonzentrationen der Radionuklide (pCi/kg) im lebenden und im abiotischen (Boden, Wasser) Material

Ingestion nur 580 pCi ^{226}Ra. Die abgeschätzte Jahresingestion im Raum Menzenschwand von 7,0 nCi ist also 12 mal größer als dieser Grenzwert. (Die Grenzwerte der Strahlenschutzverordnung dürfen grundsätzlich allerdings nicht auf die natürliche Strahlenexposition angewandt werden).

Die Messung von 28 ausgesuchten Bürgern von Menzenschwand, die alle länger als 15 Jahre dort gelebt hatten, im Ganzkörperzähler des Kernforschungszentrums in Karlsruhe erbrachte einen Mittelwert für die ^{226}Ra-Gesamtkörperaktivität der gesamten Gruppe von 0,3 $\pm$ 0,7 nCi ^{226}Ra. Dieser Wert liegt zwar höher als für Bewohner in Bereichen normaler natürlicher Radioaktivität (31 pCi ^{226}Ra-Gesamtkörperaktivität), müßte aber aufgrund der abgeschätzten Jahresingestion noch wesentlich größer sein. Die Erklärung für diesen Unterschied dürfte darin liegen, daß - wie nicht anders zu erwarten - nur ein Teil der gesamten aufgenommenen Nahrungsmittel aus Menzenschwand und seiner unmittelbaren Umgebung selbst stammt.

Wir hatten während der letzten Jahre (1977/78/79) Gelegenheit, an unserem Institut an einer großen Zahl von Proben Mittelwerte der natürlichen ^{226}Ra-Konzentration im menschlichen Knochen (Zahl der Proben 805), in Placenten (Zahl der Proben 2130), in Blutplasma (7 l) und in einer Standarddiät (insgesamt 235 Tagesrationen mit zusammen $\approx$ 330 kg) zu bestimmen (B. Glöbel, H. Muth, 34) (B. Glöbel, 35). Die Proben wurden uns von O. Pribilla vom Institut für Gerichtliche und Soziale Medizin der Universität Kiel zur Verfügung gestellt. An den gleichen Proben war dort die ^{137}Cs- und vor allem die ^{90}Sr-Konzentration gemessen worden (O. Pribilla, 36). Tabelle XVII gibt eine Zusammenstellung der von uns gefundenen Mittelwerte und zum Vergleich die Mittelwerte der älteren deutschen Messungen (6, 23). Es zeigt sich, daß die früheren Werte durch die neuen Untersuchungen an einer großen Zahl von Proben im wesentlichen bestätigt wurden.

Die uns überlassenen Knochenproben waren bereits nach 4 Altersgruppen mit der oberen Grenze 40. Lebensjahr eingeteilt. Proben von Personen älter als 40 Jahre standen bisher nicht zur Verfügung. In Abbildung 4 sind die gemessenen Mittelwerte der ^{226}Ra-Konzentration in der Knochenasche als Funktion des Alters in

Jahren wiedergegeben. Abbildung 5 zeigt in gleicher Weise die von O. Pribilla (36) gemessenen ^{90}Sr-Konzentrationen des gleichen Probenmaterials. Aus den beiden Abbildungen ist zu entnehmen, daß der ^{226}Ra-Gehalt im Knochen eine dem ^{90}Sr-Gehalt ähnliche Altersabhängigkeit zeigt. Die beiden Maxima jeweils in den ersten Lebensjahren und dem 9. bis 15. Lebensjahr entsprechen den Wachstumsphasen des menschlichen Skeletts. Es ist bekannt, daß in diesen Lebensabschnitten das Anwachsen des Calciumbestandes im Menschen jeweils stark beschleunigt ist. Wahrscheinlich werden in diesen Wachstumsphasen ^{226}Ra, ^{90}Sr und Ca ohne Diskriminierung im Verhältnis, wie es im Plasma vorliegt,in den Hydroxylapatitkristall des Knochens eingebaut. In den Phasen der stationären Knochenmasse beherrschen Diffusions- und Umkristallisationsvorgänge das Geschehen, in deren Verlauf ^{90}Sr und ^{226}Ra leichter ausgeschieden werden als Ca, was zu einer Verminderung der ^{226}Ra- und ^{90}Sr-Konzentration im Knochen während des Lebensalters von 2 bis 9 Jahren und ab dem 15. Lebensjahr führt. Wie sich die ^{226}Ra-Konzentration im Knochen bei höherem Lebensalter ($>$40 a) entwickelt, ist noch nicht abzusehen. Unsere Untersuchungen werden in Zusammenarbeit mit dem Institut für Wasser-, Boden- und Lufthygiene des Bundesgesundheitsamtes in München-Neuherberg weitergeführt, wobei vor allem auch der Lebensabschnitt $>$40 a Berücksichtigung finden wird.

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Tabelle I: ^{226}Ra in Leitungswasser (Trinkwasser) nach (6)

Ort	pCi ^{226}Ra/l	Calcium (g/l)	Ci ^{226}Ra/g Ca ($\times 10^{-12}$)	Ci Rn/l ($\times 10^{-10}$)
Frankfurt a.M.	0,22	0,040	5,5	1,3
München	0,09	-	-	-
Northeim	0,34	0,078	4,5	2,2
Inheiden	0,20	-	-	-
Bochum	0,03	-	-	-
Erlangen	0,15	-	-	-
Badgastein (Österreich)	0,27	0,016	17,0	-

Tabelle II: ^{226}Ra in Quellwässern nach (6)

Ort	pCi ^{226}Ra/l	Calcium (g/l)	Ci ^{226}Ra/g Ca ($\times 10^{-12}$)	Ci Rn/l ($\times 10^{-10}$)
Quellen 400 - 500 m tief				
Hassia-Sprudel Bad Vilbel	9,16	0,227	33,7	6,5
Friedrich-Karl-Spr. Bad Vilbel	11,7	-	-	-
Kronthal/Taunus	18,4	0,257	71,5	10,6
Quellen 30 - 50 m tief				
Erlbach bei Donauwörth	0,26	-	-	-
Bismarckquelle Friedrichsruh	0,13	0,122	1,09	-
Hermannsquelle Mölln	0,07	0,065	1,08	-
Hitzacker	0,14	0,104	1,3	-
Ratzeburg	0,52	0,094	5,56	-

Tabelle III: ^{226}Ra in Flußwasser nach (6)

Fluss	pCi ^{226}Ra/l	Calcium (g/l)	Ci ^{226}Ra/g Ca ($\times 10^{-12}$)
Main	0,07	0,079	0,9
Isar	0,7	-	-
Leine	0,84	0,118	7,1
Regnitz	0,12	-	-

Tabelle IV: ^{226}Ra und ^{222}Rn in Wässern des Fichtelgebirges nach (7)

Ort der Probenahme	Wasserart[1]	^{226}Ra 2) (pCi/l)	^{222}Rn (nCi/l)
Fichtelberg	TW	2,3	7,9
Kirchenlamitz	TW	1,5	7,0
Ochsenkopf	Br	0,76	4,7
Fichtelsee	Br	0,72	1,7
	O	0,64	0,08
Bischofsgrün	TW	0,58	7,6
Alexanderbad	TW	0,54	0,9
Weißer Main	0	0,35	0,08
Bad Berneck	TW	0,30	0,48
Wunsiedel	Br	0,29	5,0
	Br	< 0,05	0,03
	TW	< 0,05	0,43
Weißenstadt	TW	0,26	4,6
	Br	0,18	2,3
	O	0,07	0,03
Münchberg	TW	0,07	4,3

1) TW = Trinkwasser
Br = Brunnen
O = Oberflächenwasser

2) relativer Streubereich der Meßwerte (95 %) ca. 30 %

Tabelle V: Auswirkungen der Wasseraufbereitung auf den Gehalt an ^{226}Ra und ^{222}Rn in zwei Berliner Wasserwerken nach (7)

Wasseraufbereitung	Wasserwerk B		Wasserwerk J	
	^{226}Ra (pCi/l)+)	^{222}Rn (nCi/l)	^{226}Ra (pCi/l)+)	^{222}Rn (nCi/l)
Rohwasser	0,15	0,07	0,44	0,09
Rohwasser nach Belüftung	0,20	0,02	0,38	0,03
Reinwasser nach Filtern	0,09	0,05	0,11	0,11
Reinwasser vor Netzeinspeisung	0,12	0,05	0,11	0,07

+) relativer Streubereich (ca. 95 %) ca. 30 %

Tabelle VI: Zusammenfassung gemessener Konzentrationen von ^{226}Ra in verschiedenen Wässern nach (11)

Art des Wassers	Zahl der Proben	Medianwert (pCi/l)	Schwankungsbereich (pCi/l)
Leitungswasser	205	0,11	0,01 - 3,0
Mineralwasser (Flaschen)	19	1,73	0,12 - 14,6
Trinkheilwasser (Kur)	10	1,45	0,14 - 6,9
Quell- und Brunnenwasser	12	0,16	0,05 - 0,9
Fluß- und Seewasser	23	0,08	0,01 - 0,5
Schnee und Regenwasser	3	0,02	0,01 - 0,15

Tabelle VII: Minimale, maximale und mittlere Konzentrationen von ^{226}Ra im Krunkelbach, im Abwasser der Uranuntersuchungsgrube, in der Menzenschwander Alb und im Trinkwasser von Menzenschwand nach (12)

Probenahme	^{226}Ra-Konzentration in pCi/l		
	Minimum	Maximum	Mittelwert
Krunkelbach, 150 m vor Einleitung der Grubenwässer	0,07	4,4	0,5±0,3
Abwasser der Uranuntersuchungsgrube Krunkelbach	4,1	128	23±6
Krunkelbach, 150 m nach Einleitung des Abwassers	0,39	23,1	4±1
Krunkelbach, 100 m vor Zusammenfluß mit der Feldberger Alb	0,8	8,2	2,1±0,6
Menzenschwander Alb, 100 m nach Einmündung der Feldberger Alb	0,59	8,3	1,1±0,1
Trinkwasserversorgung Menzenschwand	0,11	0,57	0,31±0,03

Tabelle VIII: Mittlere Aktivitätskonzentration wichtiger natürlicher Radionuklide im Trinkwasser nach (14)

Nuklid	Konzentration (pCi/l)
^{238}U	0,05
^{226}Ra	0,1
^{222}Rn + kurzl. Folgeprodukte	10-100
^{210}Pb	0,02
^{210}Po	0,01
^{40}K	5
^{3}H	5

Tabelle IX: ^{226}Ra in verschiedenen Nahrungsmitteln tierischen Ursprungs nach (6)

Nahrungsmittel	Ausgangs-material (g)	Ca im Ausgangs-material (g)	fCi ^{226}Ra pro 1 g Ausgangs-material	^{226}Ra pro 1 g Ca (x 10^{-13} Ci)
Schweinefleisch	1000	0.079	0.8	97
	1000	0.085	1.5	178
Schweineleber	300	0.030	0.8	795
Rindfleisch	1050	0.096	0.8	88
	880	0.068	0.8	107
Rinderblut	2000	0.117	0.2	41
Milch	1000	1.280	0.3	2.1
Milchpulver	984	11.700	1.7	1.4
20 Eier	940	0.432	3.1	69
Butter	980	0.538	0.3	5.5
Grüne Heringe	975	3.890	2.8	7
(einschl.Gräten)	985	4.140	4.0	10
Kabeljau (ohne Gräten)	600	0.158	4.0	150
Schellfisch (ohne Gräten)	635	0.209	6.3	190
Lebertran	1000	0.022	4.9	2240

Tabelle X: ^{226}Ra in verschiedenen Nahrungsmitteln pflanzlichen Ursprungs nach (6)

Nahrungsmittel	Ausgangs-material (g)	Ca im Ausgangs-material (g)	fCi ^{226}Ra pro 1 g Ausgangs-material	^{226}Ra pro 1 g Ca (x 10^{-13}Ci)
Roggen-Mischbrot	1475	0,283	2,6	138
Weißbrot	896	0,840	3,3	34
	1181	0,850	1,7	24
Weizenmehl	500	0,102	2,7	136
Margarine	986	0,288	0,1	3,4
Karotten	796	0,260	1,6	49
	787	0,264	1,7	52
	1240	0,460	6,1	229
Kohl	810	0,758	2,4	26
	823	0,621	1,0	13
Äpfel	1618	0,085	0,9	150

Tabelle XI: ^{226}Ra in Kartoffeln aus Nord- und Süddeutschland nach (6)

	Zahl der Proben	Durchschnitts-gewicht einer Probe (g)	fCi ^{226}Ra pro 1 g Frischgewicht	^{226}Ra pro 1 g Ca (x 10^{-13} Ci)
Norddeutschland	22	1500	1,0	238
Süddeutschland	17	2000	0,6	123

Tabelle XII: ^{210}Pb und ^{210}Po in Nahrungsmitteln nach (18)

Nahrungsmittel	Tägl. Aufnahme (g)	Mittlere ^{210}Po-Konzentration (pCi/g)	Wertebereich pCi/g	Zahl der gemessenen Proben	Tägl. aufgenommene ^{210}Po-Aktivität in pCi	Verhältnis $^{210}Pb/^{210}Po$
Salat	1o	0,04	0,1 -0,01	10	0,4	1
Spinat	3	0,03	0,2 -0,01	5	0,09	1
Bohnen	7	0,02	0,06 -0,005	10	0,14	1
Eier	0,5 St.	0,5 St.	1,0 -0,05	5	0,25	1
Erbsen	1	0,02	0,05 -0,008	5	0,02	1
Rinderniere	2	0,2	0,2 u. 0,8	2	0,4	0,1 - 0,5
Schweineniere	2	0,1	-	1	0,2	1
Rindfleisch	11	0,02	0,05 -0,001	10	0,22	1
Schweinefleisch	11	0,01	0,02 -0,001	10	0,11	1
Kalbfleisch	1	0,01	-	2	0,01	1
Fisch	11	0,04	0,1 -0,005	5	0,44	1
Zigaretten	1,3St.	0,1/St.	0,2 -0,005	20	0,13	1
Milch	250cm^3	0,0005	0,002-0,0001	10	0,125	1
Kohl	16	0,07	0,1 -0,007	5	1,12	1
Kartoffeln	200	0,001	0,005-0,0006	10	0,2	1
Brot	160	0,0025	0,005-0,001	10	0,4	1
Reis	4	0,003	0,009-0,0005	10	0,012	1
Zucker	40	0,002	0,004-0,001	10	0,08	1
Öl	5	0,002	0,006-0,0008	5	0,01	1
Honig	1,6	0,0018	0,003-0,0008	5	0,0029	1
Mehl	27	0,01	0,0015-0,004	10	0,27	1
Wein	16cm^3	0,00055 pro cm^3	0,001-0,00015	5	0,0088	1
Trinkwasser	1500cm^3	0,0028/1	0,006-0,0001	10	0,0042	10

Tabelle XII: Fortsetzung nach (18)

Nahrungsmittel	Tägl. Aufnahme (g)	Mittlere ^{210}Po-Konzentration (pCi/g)	Wertebereich pCi/g	Zahl der gemessenen Proben	Tägl. aufgenommene ^{210}Po-Aktivität in pCi	Verhältnis $^{210}Pb/^{210}Po$
Bier	85 cm^3	0,0004/l	0,006-0,0001	5	0,0034	1
Gras	-	0,3	0,5 -0,1	20	0,00	1
			Mittelwerte:	200	4,65	

Tabelle XIII: ^{226}Ra-Konzentrationen, gemessen in Milch verschiedener Molkereien nach (12)

Probennahme	^{226}Ra-Konzentration in pCi/l
Karlsruhe	0,92
Freiburg	0,91
Karlsruhe/Südschwarzwald	3,5
Waldshut/Tingen	5,7

Tabelle XIV: ^{226}Ra-Konzentrationen in Umweltproben aus Menzenschwand und Umgebung verglichen mit Literaturangaben (13) nach (12)

Probenmaterial	^{226}Ra-Konzentration in pCi/kg Menzenschwand (12)	(13)
Boden	1200 - 1500	150 - 3100
Wasser: Fluß- und See-	0,03 - 2,5	0,002 - 62
Grund- und Quell-	0,1 - 1549	0,001 - 237800
Trink-	0,11 - 0,57	0,005 - 50
Nahrungsmittel:		
Kartoffel	30 - 40	0,8 - 2,8
Getreide, Mehl	20 - 240	1,9 - 2,8
Fleisch	2	0,01 - 1,1
Milch	0,3- 48	0,3
Gemüse	5 - 170	0,5 - 3,8
Fisch	1,4- 211	5,1
Eier	80	3,1 - 6,1
Innereien	10 - 200	0,1

Tabelle XV: Die Strahlenexposition "von innen" durch natürliche Radionuklide im Menschen.
Nach Stahlhofen (20, 21, 22) und Oberhausen (23)

Radionuklid	Organ	Strahlendosis in mrad/Jahr
^{226}Ra und 30 % seiner kurzlebigen Folgeprodukte	Knochen	1,0 (20) 1,0 (23
	Weichteilgewebe	0,02 (20) 0,05 (23)
^{228}Th im radioaktiven Gleichgewicht mit seinen Folgeprodukten	Knochen	0,8 (20) 1,0 (23)
	Weichteilgewebe	0,03 (20) 0,07 (23)
^{210}Po	Knochen	3,1 (20) 2,5 (23)
	Weichteilgewebe	0,3 (20) 0,05 (23)
^{40}K	Knochen	11 (23)
	Muskel	24 (23)
	Weichteilgewebe außer Muskel	16 (23)

Tabelle XVI: Transferfaktoren für ^{226}Ra nach (12)

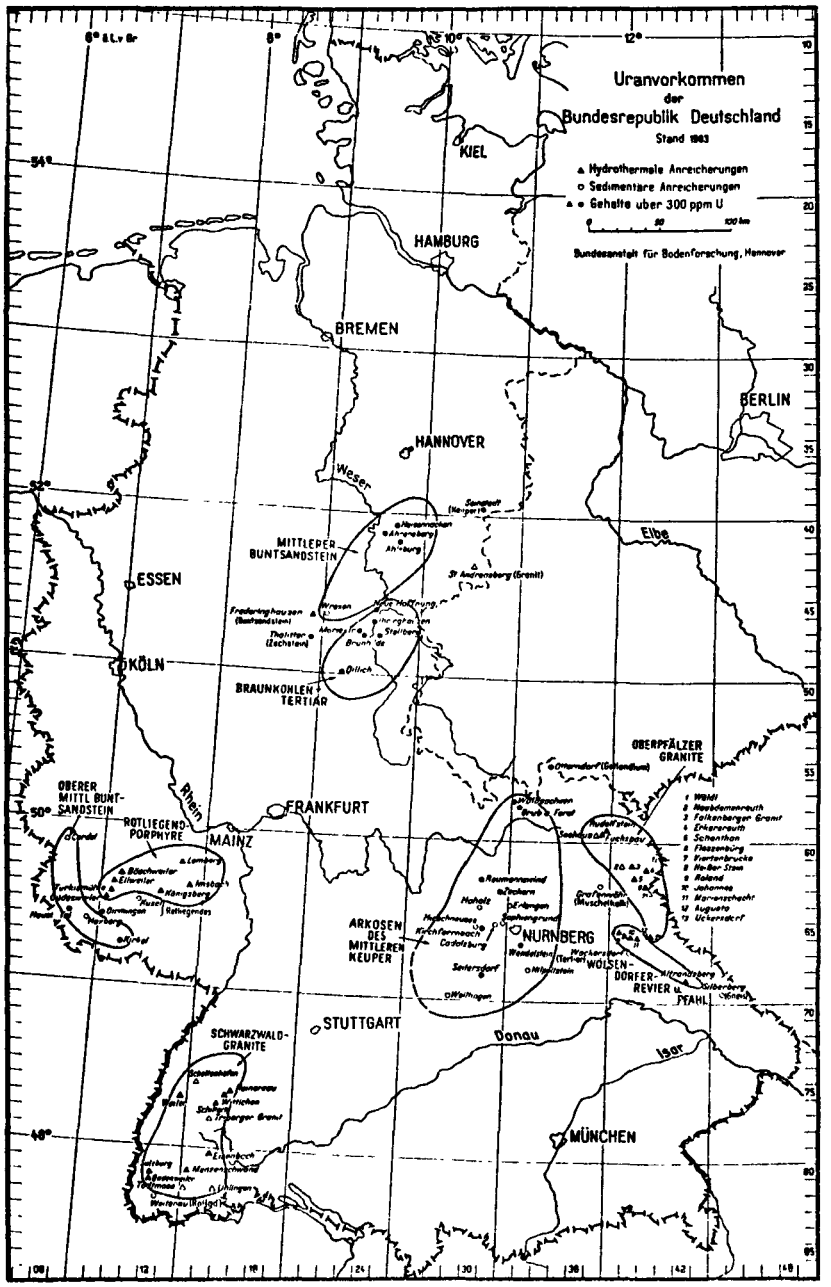
Material	Transferfaktor	Bemerkungen
Wasser/Sedimente	$(11 \pm 4) \cdot 10^{-5}$	Im 3. Absetzbecken der Uranuntersuchungsgrube
Wasser/Sedimente	$(26 \pm 6) \cdot 10^{-5}$	In den Menzenschwander Bächen
Gras/Boden	$0{,}027 \pm 0{,}005$	$\frac{\text{in pCi/g Gras, frisch}}{\text{pCi/g Boden, trocken}}$
Nahrungsmittel/Boden	0,001 (Rindfleisch) bis 0,2 (Weizen, Gerste, Heidelbeeren)	$\frac{\text{in pCi/kg Nahrungsmittel, frisch}}{\text{pCi/kg Boden, trocken}}$
Milch/Gras	$0{,}2 \pm 0{,}05$	
Forelle/Wasser	28 ± 12	bezogen auf das Gesamtgewicht
Forelle/Wasser	22 ± 14	bezogen nur auf das Fischfleisch

Tabelle XVII: Mittlere Konzentration von ^{226}Ra und Ca in menschlichen Geweben und in der Nahrung, bezogen auf Frischgewicht nach (34, 35)

	^{226}Ra in fCi/g			Ca in g/g
	1978 (34, 35)	1960 (6)	1964 (23)	Geigy
Knochen[1]	7,1	5,2	10,0	$1,4 \cdot 10^{-1}$
Placenten[2]	0,8	0,8	-	$2,5 \cdot 10^{-4}$
Blutplasma	0,5	0,2	0,12	$7,0 \cdot 10^{-5}$
Nahrung	0,3	0,2	0,3	$7,3 \cdot 10^{-4}$

[1]Zahl der Proben: 805

[2]Zahl der Proben: 2130


Uranvorkommen
der
Bundesrepublik Deutschland
Stand 1983
Hydrothermale Anreicherungen
Sedimentare Anreicherungen
Gehalte uber 300 ppm U
Bundesanstalt für Bodenforschung, Hannover
KIEL
HAMBURG
BREMEN
HANNOVER
BERLIN
Weser
Elbe
MITTLEPER BUNTSANDSTEIN
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KÖLN
BRAUNKOHLEN TERTIÄR
OBERPFÄLZER GRANITE
OBERER MITTL BUNT-SANDSTEIN
ROTLIEGEND-PORPHYRE
Rhein
FRANKFURT
MAINZ
ARKOSEN DES MITTLEREN KEUPER
NÜRNBERG
WOLSEN-DORFER-REVIER
PFAHL
STUTTGART
SCHWARZWALD-GRANITE
Donau
Isar
MÜNCHEN

Abbildung 1

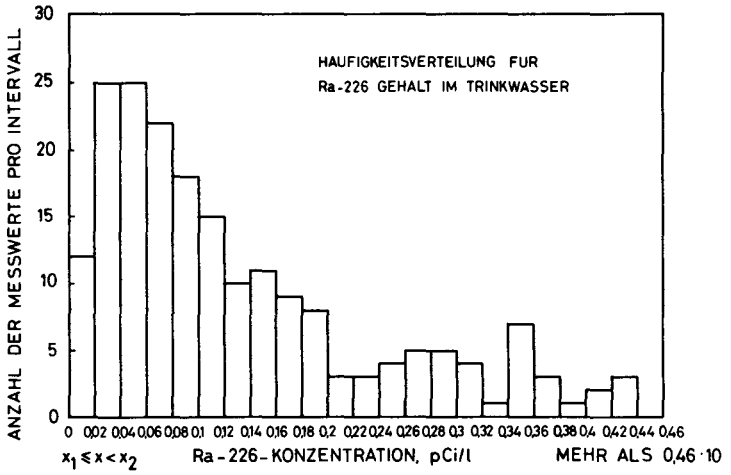

Abbildung 2: nach (11)

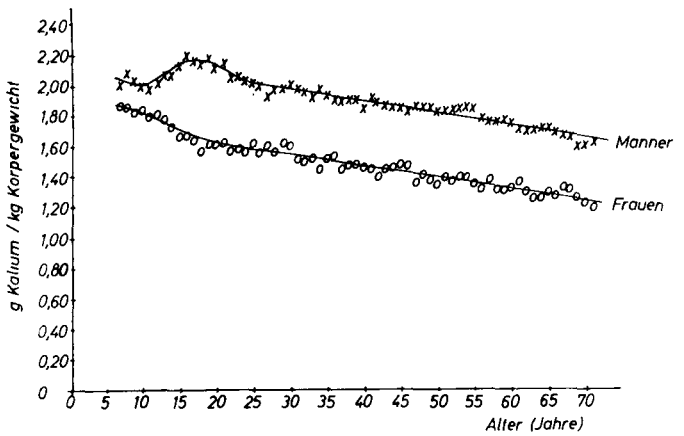

Abbildung 3: Mittlere Kaliumkonzentration in Abhängigkeit vom Alter nach (23)

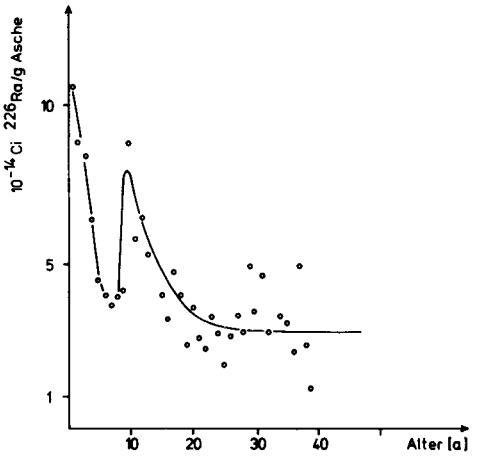

Abbildung 4: Altersabhängigkeit der ^{226}Ra-Konzentration in menschlicher Knochenasche nach (34)

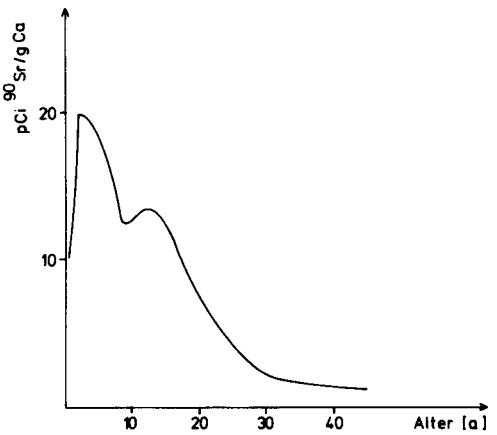

Abbildung 5: Altersabhängigkeit der spezifischen ^{90}Sr-Aktivität in menschlichen Knochen nach (36)

ÜBERBLICK ÜBER NATÜRLICH RADIOAKTIVE STOFFE IN ABFÄLLEN VON KONVENTIONELLEN KRAFTWERKEN UND DIE DARAUS RESULTIERENDE STRAHLENEXPOSITION

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KURZFASSUNG. Die Frage der Emission natürlich radioaktiver Stoffe bei konventionellen Kraftwerken und die daraus resultierende Strahlenexposition ist in den letzten Jahren, nicht nur in zahlreichen wissenschaftlichen Veröffentlichungen, sondern auch in der Öffentlichkeit, lebhaft diskutiert worden.
Die Arbeit gibt einen Überblick über die vorliegenden Messergebnisse des Gehaltes von Kohle, Flugasche und Granulat an natürlich radioaktiven Stoffen und über die auf Grund dieser Ergebnisse durchgeführten Berechnungen zur Abschätzung der Strahlenexposition. Die für die Berechnung verwendeten Annahmen werden erläutert und auf die Unsicherheiten der Berechnungen hingewiesen.
Abschliessend werden die Probleme bei Verwendung von Öl und Erdgas als Energiequelle diskutiert.

SUMMARY. SURVEY OF NATURALLY OCCURRING RADIOACTIVE SUBSTANCES DISCHARGED FROM CONVENTIONAL POWER STATIONS AND THE CONSEQUENT RADIATION EXPOSURE.
The emission of naturally occurring radioactive substances from conventional power stations and the resultant radiation exposure have been the subject of lively debate in recent years not only in numerous scientific publications, but also among the general public. The study gives an outline of the available measurements of naturally occurring radioactive substances in coal, fly-ash and off-gases, and of the radiation exposure estimated on the basis of these results. The assumptions and uncertainties underlying the calculations are explained.
The problems which arise when oil and natural gas are used as sources of energy are subsequently discussed.

RESUME. REVUE DES SUBSTANCES RADIOACTIVES NATURELLES CONTENUES DANS LES DECHETS DES CENTRALES CONVENTIONNELLES ET DE L'EXPOSITION EN RESULTANT.
La question de l'émission, par les centrales conventionnelles, de substances radioactives naturelles, et de l'exposition en résultant a suscité ces dernières années, de vives discussions non seulement dans de nombreuses publications scientifiques, mais aussi dans l'opinion publique.

Ce rapport fournit un aperçu des résultats actuellement disponibles de mesures de la teneur du charbon, des cendres volantes et des produits granulés en substances radioactives naturelles ainsi que de l'exposition aux rayonnements estimée sur base de ces résultats. On explicite les hypothèses de calcul utilisées et on signale les incertitudes de calculs. On discute pour conclure les problèmes posés par un recours au pétrole et au gaz naturel comme sources d'énergie.

Die Frage der Emission natürlich radioaktiver Stoffe aus konventionellen Kraftwerken und ihr Beitrag zur zivilisatorisch bedingten Veränderung der Strahlenexposition aus natürlichen Strahlenquellen hat in den letzten Jahren in der Bundesrepublik wissenschaftliche Gremien und die Öffentlichkeit beschäftigt. Bis zum Jahre 1976 lagen aber nur wenige Meßergebnisse über den Gehalt von Kohle, Flugstaub und Granulat an natürlich radioaktiven Stoffen vor, die zu einer Abschätzung der Strahlenexposition herangezogen werden konnten. Dies hat sich in der Zwischenzeit in der Bundesrepublik wesentlich geändert, nicht zuletzt dadurch, daß die Strahlenschutzkommission beim Bundesminister des Innern im April 1978 eine Arbeitsgruppe eingesetzt hat, die die für eine abschließende Beurteilung noch offenen Fragen klären sollte. Diese waren:

1.) Spezifische Aktivität und Radionuklidzusammensetzung des emittierten Reingasstaubes

2.) Partikelgröße der emittierten Staubteilchen

3.) Löslichkeit der natürlich radioaktiven Stoffe der emittierten Staubteilchen

4.) Dosisfaktoren für natürlich radioaktive Stoffe.

Nach der Vorstellung der Arbeitsgruppe sollten die Untersuchungen in der Bundesrepublik eine Beurteilung für die Umgebung eines modernen Steinkohlekraftwerkes und eines modernen Braunkohlekraftwerkes ermöglichen. Die Arbeiten der Gruppe sind zwar noch nicht abgeschlossen, doch liegen Ergebnisse vor, die hier zur Diskussion gestellt werden können.

Kohleverbrauch und Staubemissionen moderner Kraftwerke.

Ein 1000 MW-Kohlekraftwerk verbraucht im Jahr etwa $1,5 \times 10^6$ t Kohle. Wenn man von einem Aschegehalt von ca. 8% ausgeht, bedeutet dies, daß jährlich ca. $1,2 \times 10^5$ t Asche bei einem 1000 MW-Kohlekraftwerk anfallen. Diese Asche könnte mit der Abluft in die Umgebung abgegeben werden. Der Staubanteil in einem Normkubikmeter Reingas würde dann ca. 8g betragen, wenn man davon ausgeht, daß bei der Verbrennung je Tonne Kohle ca. 10^4 Kubikmeter Reingas bezogen auf den Normalzustand entstehen. In der Bundesrepublik Deutschland ist die Staubemission gesetzlich aber auf maximal 150 mg pro Normkubikmeter Rauchgas

begrenzt, sodaß ein Kohlekraftwerk Rückhaltevorrichtungen mit hohem Wirkungsgrad benötigt. Durch den Einsatz von mehrstufigen Elektrofiltern beträgt die tatsächliche Staubemission in der Bundesrepublik bei einem modernen Steinkohlekraftwerk nur ca. 60 mg pro Normkubikmeter und die bei einem modernen Braunkohlekraftwerk ca. 30 mg pro Normkubikmeter. Um diese Werte zu erreichen, benötigt man bei Steinkohlekraftwerken eine zweistufige, bei Braunkohlekraftwerken eine dreistufige Filteranlage. Trotz aller Rückhaltevorrichtungen kann aber davon ausgegangen werden, daß bei modernen Kohlekraftwerken jährlich größenordnungsmäßig 1000 t Flugstaub über den Schornstein emittiert werden. Der Wert für Steinkohlekraftwerke liegt dabei im allgemeinen über 1000 t pro Jahr, der für Braunkohlekraftwerke unter 1000 t pro Jahr.

Spezifische Aktivität und Radionuklidzusammensetzung.
Die spezifische Aktivität von Kohle an Radium 226 und Thorium 232 liegt im allgemeinen mit $< 0,5$ nCi/kg unter der der Erdkruste, für die eine spezifische Aktivität von 1 bzw. 1,2 nCi/kg angenommen wird. In den Verbrennungsrückständen ist auf Grund des Aschegehaltes der Kohle eine etwa um den Faktor 10 höhere spezifische Aktivität zu erwarten. Wenn man davon ausgeht, daß sich Uran/Radium und seine Folgeprodukte sowie Thorium und seine Folgeprodukte in der Kohle im Gleichgewicht befinden, reichern sich bei der Verbrennung die leicht flüchtigen Bestandteile aus den natürlichen Zerfallsreihen im Flugstaub leicht an. Für eine vergleichende Untersuchung wurden vom TÜV-Rheinland Flugascheproben aus der Vorreinigung, Mittelreinigung und Nachreinigung bei einem modernen Braunkohlekraftwerk und aus der Vorreinigung und Nachreinigung von einem modernen Steinkohlekraftwerk entnommen und über α und γ-Messungen bei der Gesellschaft für Strahlen- und Umweltforschung und beim Institut für Strahlenhygiene des Bundesgesundheitsamtes analysiert. Als Ergebnis dieser Analysen kann festgestellt werden, daß die spezifische Aktivität des Flugstaubes der Mittel- und Nachreinigung immer größer ist, als bei der Vorreinigung. Die Daten sind in den Tab. 1 und 2 zusammengestellt. Besonders auffällig ist, daß die spezifische Aktivität von Pollonium 210 im Flugstaub der Nachreinigung beim Stein-

kohlekraftwerk fast dreimal so hoch ist wie die der Vorreinigungsproben. Beim Flugstaub des Braunkohlekraftwerks ist dieser Faktor sogar fast 7. Insgesamt sind die spezifischen Aktivitäten der Flugaschen des Braunkohlekraftwerkes aber niedriger als beim Flugstaub von Steinkohlekraftwerken. Da die spezifische Aktivität des Flugstaubes aus der letzten Elektrofilterstufe nicht mit der des mit dem Reingas aus dem Schornstein des Kraftwerkes emittierten Flugstaubes übereinstimmen muß, wurden auch Proben aus dem Reingasstaub entnommen und nach chemischer Aufarbeitung durch die Gesellschaft für Strahlen- und Umweltforschung über eine -Messung analysiert. Die Ergebnisse zeigen für beide Kraftwerkstypen eine recht gute Übereinstimmung der spezifischen Aktivität des Flugstaubes aus dem letzten Elektrofilter mit der aus dem Reingasstrom. Die Reingasstromproben konnten vom TÜV-Rheinland nur mit erheblichem technischen Aufwand entnommen werden. Die Probenahme für eine Probenmasse von 1 g dauerte einige Tage! Der Aufwand hat sich aber gelohnt durch die Tatsache, daß man die spezifische Aktivität des Flugstaubes der letzten Filterstufe mit guter Übereinstimmung gleich der spezifischen Aktivität der mit dem Reingasstrom über den Schornstein emittierten Staubteilchen setzen kann. Aus den gemessenen spezifischen Aktivitäten in Flugstaubproben läßt sich für die beiden Kraftwerkstypen eine mittlere jährliche Aktivitätsemission berechnen. Die entsprechenden Daten sind in Tabelle 3 zusammengestellt.

Partikelgröße der emittierten Staubteilchen.

Da für die Berechnung der Strahlenexposition die Partikelgröße der Staubteilchen einen Einfluß hat, wurde auch dieser Frage nachgegangen. Der TÜV-Rheinland hat ermittelt, daß ca. 25% der emittierten Partikel beim Steinkohlekraftwerk und ca. 20% der emittierten Partikel beim Braunkohlekraftwerk größer als 10 µm sind. Eine Bestimmung der spezifischen Aktivität von Flugaschen aus den Elektrofiltern, die in 4 Gruppen nach Korngrößen aufgeteilt waren, hat ferner gezeigt, daß die spezifische Aktivität mit zunehmender Korngröße abnimmt. Hierdurch läßt sich auch die unterschiedliche spezifische Aktivität in den einzelnen Filterstufen des Elektrofilters erklären, denn die größeren Teilchen werden sicher zuerst abgeschieden.

Löslichkeit

Für die Abschätzung der Strahlenexposition ist auch entscheidend, ob die natürlich radioaktiven Stoffe in den Flugstaubpartikeln löslich oder unlöslich sind. In der Literatur finden sich Angaben zwischen 6 und 35% Löslichkeit. Die Arbeitsgruppe hält eine Löslichkeit von 15% für realistisch.

Dosisfaktoren für natürlich radioaktive Stoffe.

In der Bundesrepublik Deutschland sind für die Ermittlung der Strahlenexposition durch Emission radioaktiver Stoffe mit der Abluft die vom Bundesminister des Innern im Jahre 1971 herausgegebenen "Allgemeinen Berechnungsgrundlagen" verbindlich. Diese enthalten auch für natürlich radioaktive Stoffe Dosisfaktoren, die auf der Grundlage von ICRP 2 berechnet wurden. Jacobi hat aber gezeigt, daß gerade der in den "Allgemeinen Berchnungsgrundlagen" angegebene Dosisfaktor für Radium-226 nach neueren wissenschaftlichen Erkenntnissen eine etwa 10-fache Überschätzung der Strahlenexposition des Knochens zur Folge hat. Die Diskussion, ob es sinnvoll ist, für die Berechnung der Strahlenexposition durch Emissionen von Kohlekraftwerken überhaupt die "Allgemeinen Berechnungsgrundlagen" zu verwenden oder ob man besser dazu einen Vergleich mit dem natürlichen Gehalt radioaktiver Stoffe im Körper durchführen sollte, ist in der Arbeitsgruppe noch nicht abgeschlossen. - Die Arbeitsgruppe ist sich aber darüber einig, daß sowohl die Strahlenexposition durch Emission natürlich radioaktiver Stoffe mit der Abluft aus Kohlekraftwerken als auch die Strahlenexposition durch Emission radioaktiver Stoffe aus Kernkraftwerken im Normalbetrieb im Vergleich zur Strahlenexposition aus anderen Strahlenquellen ohne Bedeutung ist.

Die Ausführungen haben gezeigt, daß die Strahlenexposition durch Emissionen natürlich radioaktiver Stoffe aus Braunkohlekraftwerken niedriger ist als die durch Emissionen aus Steinkohlekraftwerken. Öl-gefeuerte Kraftwerke haben noch niedrigere Aktivitätsemissionen. Bei Benutzung von Erdgas als Energiequelle ist nach einer Niederländischen Untersuchung die Radon-Emission das Hauptproblem. Ein sehr viel größeres Radon-Problem haben wir aber in Wohnungen, wie die Berichte des gestrigen Tages gezeigt haben. - Literatur kann vom Verfasser angefordert werden.

Tabelle 1: Natürlich radioaktive Stoffe in Flugaschen eines Steinkohle–Kraftwerkes in nCi/kg (Mittelwerte)

nach Messungen der GSF:

Proben-bezeichnung	Nuklid	spezifische Aktivität
Vorreinigung	K 40	22
	U 238/Th 234*	6,6
	Ra 226/Pb 214/Bi 214*	5,7
	Pb 210	38
	Po 210	39
	Ac 228/Pb 212/Tl 208*	2,7
Nachreinigung	K 40	23
	U 238/Th 234*	8,8
	Ra 226/Pb 214/Bi 214*	6,4
	Pb 210	64
	Po 210	108
	Ac 228/Pb 212/Tl 208*	3,0
nach γ–Messungen des Bundesgesundheitsamtes:		
Vorreinigung	K 40	18
	Ra 226	5,7
	Th 232	2,5
Nachreinigung	K 40	18
	Ra 226	6,8
	Th 232	2,9

* radioaktives Gleichgewicht in der Probe angenommen

Tabelle 2: Natürlich radioaktive Stoffe in Flugaschen eines Braunkohle–Kraftwerkes in nCi/kg (Mittelwerte)

nach Messungen der GSF:

Proben-bezeichnung	Nuklid	spezifische Aktivität
Vorreinigung	K 40	4,3
	U 238/Th 234*	1,2
	Ra 226/Pb 214/Bi 214*	0,7
	Pb 210	1,2
	Po 210	0,7
	Ac 228/Pb 212/Tl 208*	0,3
Mittelreinigung	K 40	4,1
	U 238/Th 234*	1,6
	Ra 226/Pb 214/Bi 214*	0,8
	Pb 210	1,4
	Po 210	1,3
	Ac 228/Pb 212/Tl 208*	0,6
Nachreinigung	K 40	7,6
	U 238/Th 234*	2,8
	Ra 226/Pb 214/Bi 214*	1,4
	Pb 210	2,4
	Po 210	4,8
	Ac 228/Pb 212/Tl 208*	1,0
nach γ–Messungen des Bundesgesundheitsamtes:		
Vorreinigung	K 40	3,3
	Ra 226	0,8
	Th 232	$<$ 0,5
Mittelreinigung	K 40	5,5
	Ra 226	1,3
	Th 232	0,7
Nachreinigung	K 40	9,8
	Ra 226	1,6
	Th 232	0,8

* radioaktives Gleichgewicht in der Probe angenommen

Tabelle 3: Berechnete mittlere jährliche Aktivitätsemissionen bei Steinkohle bzw. Braunkohle gefeuerten 1 000 MW Kraftwerken in mCi

Nuklid	Steinkohle	Braunkohle
U 238	10	3
U 234	10	3
Th 230	10	2
Ra 226	10	2
Pb 210	100	3
Po 210	150	5
Th 232	5	1
Th 228	5	1

NATURAL RADIOACTIVITY LEVELS IN RELEASES FROM COAL FIRED POWER PLANTS IN ITALY

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SUMMARY. The importance of coal in energy production is growing fast owing to the controversies encountered in the development of nuclear power plants and to the projection of oil shortages and increasing prices in the next ten years.

Thus in Italy the National Electricity Production Board (ENEL) is planing to expand the use of coal by a factor of about four in the eighties, either through increased exploitation of Italian brown-coal mines, or through increasing imports from foreign countries.

Against this background, the present paper presents data on levels of natural radionuclides measured in Italian coals and those reported in the literature for coals of foreign origin. Such levels vary widely with the geographical location of the mine and the type of coal.

An evaluation is also reported of the actual and potential radiological impacts of coal fired power plants in Italy, which impacts are currently somewhat limited but might be enhanced up to ten-fold as a consequence of the planned developments.

RESUME. NIVEAUX DE RADIOACTIVITE NATURELLE DANS LES REJETS DES CENTRALES AU CHARBON EN ITALIE. Le rôle du charbon dans la production d'énergie croît rapidement devant les controverses soulevées par la multiplication des centrales nucléaires et les perspectives d'une pénurie de pétrole et d'une hausse des prix ces dix prochaines années.

En Italie aussi, la Société Nationale de Production Electrique (ENEL) prévoit de quadrupler environ l'utilisation de charbon dans les années 80, soit en intensifiant l'exploitation des mines de lignite italiennes, soit en augmentant les importations de l'étranger.

Dans ce contexte, le présent rapport fournit des données sur les niveaux de radionucléides naturels mesurés dans les charbons italiens et sur ceux cités dans la littérature pour les charbons étrangers. Ces niveaux varient largement selon les sites des mines et les types de charbon.

On y évalue aussi l'impact radiologique réel et potentiel des centrales au charbon en Italie, lequel, plutôt limité actuellement, pourrait jusque décupler par suite des développements prévus.

KURZFASSUNG. NATÜRLICHE RADIOAKTIVITÄT IN ABLEITUNGEN AUS KOHLEKRAFTWERKEN IN ITALIEN. Die Bedeutung der Kohle für die Energieerzeugung nimmt wegen des umstrittenen Ausbaus von Kernkraftwerken und wegen der für die nächsten zehn Jahre erwarteten Ölverknappung und Preissteigerung rasch zu.

Auch in Italien will die staatliche Elektrizitätsgesellschaft ENEL den Kohleeinsatz in den achtziger Jahren in etwa vervierfachen und hierfür entweder die Braunkohleförderung in italienischen Bergwerken steigern oder grössere Mengen aus dem Ausland einführen.

Vor diesem Hintergrund bringt das vorliegende Referat Daten über den Gehalt an natürlichen Radionukliden, der bei italienischer Kohle gemessen wurde, sowie Angaben aus der Literatur für Importkohle. Je nach der geographischen Lage des Bergwerks und nach dem Kohletyp unterliegen diese Werte grossen Schwankungen.

Ausserdem wird über die potentiellen radiologischen Auswirkungen kohlebefeuerter Kraftwerke in Italien berichtet, die zur Zeit noch ziemlich begrenzt sind, sich jedoch als Folge der geplanten Entwicklungen verzehnfachen könnten.

INTRODUCTION

Coal, as a geological material, contains small quantities of naturally occurring radionuclides. Its direct combustion, or conversion to other fuels, results in the concentration of non combustible mineral matter, including most of the said natural radionuclides, in the ash or in the gaseous residues. The release of these residues to the environment, either directly through the stack or indirectly from ash pile, results in a potential exposure of workers and population living around coal-fired power plants.

In past times coal has been widely used for producing both mechanical and electric power. Till the outcome of oil it has been almost the unique fossile fuel employed. After an eclypse due to the large availability of low cost oil in the years fifties and sixties, coal is again on the foreground of the energy sceneries for the future (1).

Coal worldwide production is actually at the second place, amongst the fossil fuels, covering about 30% of the world fuel consumption (1). its production trend, however, is only 2% to be compared with an average 7% for other fossil fuels (1). Actually such trend is slowly increasing, owing to the fact that for the present knowledge the ratio known reserves/yearly production is 200 for coal while only 36 for oil and natural gas (1). The attractiveness of coal has therefore focussed industry efforts in order to increase its production and field of use, from direct power generation to liquid hydrocarbon production.

COAL POWER PRODUCTION IN ITALY

Such guidelines, often stated by international competent bodies (2) have been also applied in Italy in the energy planning for next decade (3). Coal consumption for power production has been in fact increased from 2 10^6 tons/y to the actual 7 10^6 tons/y and is planned to reach 20 10^6 tons/y in next future (3). Italian coal production is limited to lignite ore and will be enhanced from the actual 2 10^6 tons/y to 4 10^6 tons/y in the next future (3). Italy therefore will be depending for about 80% on coal imported from various countries (Australia, Poland, South Africa, India). Coal produced electric energy will be therefore increasing from 8% of the total in 1973 to a planned 16%, taken into account of the installed power increase. In Fig. 1 are shown the locations of the coal power plants and of the lignite ore mines in Italy, including those in operation and those in construction. In Fig. 1 the power plants are also showed with different symbols depending on whether they are fired with italian lignite or import coal. Almost all these plants are located near large urban areas where some million of people reside, and there are consistent troubles caused by the environmental pollution produced. Coal power plants infact haven't ever encountered the favour of the public opinion owing to the strong alteration of the landscape they introduce especially on the coastal sites where most of them are located in order to be economically linked to the sea terminals necessary for receiving large amounts of imported coal. Even before the nuclear struggle had raised, amongst the public, a generalized feeling against large power plants, independently on how they are fueled, there had been a pressure to change from coal to oil, which is undoubtely less pollutant. Actually, however, economic convenience suggests the reemployement of coal, and public opinion requires to be ensured about the

environmental and public health impact of coal produced energy. A program therefore has been started in Italy to evaluate such impact in relation to both stable and radioactive substances which are disposed through the whole coal fuel cycle, and that might endanger the health of populations living in the vicinity of coal-fired power plants. This program includes:

a) a study of natural radionuclide content of coal ore and their distribution in various size fractions of ash particles;
b) the evaluation of both gaseous and particulate emissions, and their deposition in the surroundings of the plant sites;
c) a study of the geochemical features of the sites for determining the circulation of the deposited radionuclides and their impact on food-chain;
d) evaluation of the exposure of the various population groups via airborne, water and soil pathwais.

SAMPLING AND RESULTS

As a first step of this study two coal power plants have been considered. They are located in central Italy and are fed with lignite ore from nearby located open pit mines. Natural radionuclide content of both lignite and fly-ash from the last stage of electrostatic filter has been measured by various techniques (emanation for Radium-226, fluorimetry for Uranium, gamma spectrometry for Uranium and Thorium, while Lead-210 has been measured at PTB Braunschweig by courtesy of Dr. W. Kolb).

The results obtained, toghether with available literature data on an other italian lignite mine fed power plant and on the imported coal, are shown in Table 1. The values found for the various types of coal show some variability, the higher figures being found in an italian lignite. Such result is of some concern owing to the high ash content of lignite (20 - 30%) to be compared with an average 10% for imported coal. Tipical italian coal-fired power plants are equipped with electrostatic precipitators (filters) allowing an ash retention of 95%.

The amounts of natural radionuclides yearly released into the environment via the stack have been evaluated, and are reported in Table 2, on the basis of both actual and future coal cosumption rate. The releases from a lignite fired power plant seem to be large if compared with data from other countries (4,5,6,7,8,9,10,11,12). The low heat content and high ash residue of lignite are almost responsible of such higher levels. Particular concern shall be given in future to the new power plant to be fired with high radioactivity content lignite from a mine located in Sardinia. For that concern the other plants to be fired with imported coal from Poland and Australia, literature data (13) show that for the normalized power of 1000MW the yearly releases are within the range of the values given in other studies, taken into account of possible concentration variability (13).

DISCUSSION

The importance of such releases from the population health viewpoint, can not immediately be deduced from the data presented. Various authors have been investigating both environmental impact and doses to individuals due to natural radioactivity releases from coal fired power plants (4,5,6, 7,9,10,11,14,15) getting to conclusions not always homogeneous. Any au-

thors infact stress the importance of individual doses received by people living in the vicinity of coal fired power plant (5,11), others on the other side evaluated the radiological impact of coal fuel cycle to be far less than for the nuclear fuel cycle when the best present technology is assumed for both (6,7,10). Other authors besides, have studied health effects of fossil fuel power plants and concluded that coal fuel cycle is far the most hazardous either from the radiological point of view, than from the general health point of view (4,5,9,11,14). Many of the quoted studies, however, evidentiate that technical improvement may largely reduce airborne particulate emission, giving place to exposure smaller by a factor of about 100 (10).

In conclusion the primary importance of evaluating population impact of coal fuel cycle is to provide fundamental guidelines on which promote and shape technological studies and developments that may lead to a safer management of coal produced energy.

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Table 1

Natural radionuclides concentration in coal and ash
(pCi/g)

	^{238}U	^{226}Ra	^{210}Pb	^{232}Th
Lignite(Centr. Italy)	0.4 -0.7	0.1 - 0.4	0.7 - 1.4	2 - 3
Lignite ash (centr. Italy)	2.1 - 2.8	1.0 - 2.0	1.2 - 8.9	9
Lignite (Sardinia)	6.8	-	-	-
Lignite ash (Sardinia)	27	-	-	-
Polish coal (13)		0.9		
Polish coal ash (13)	-	0.6 - 4.1	-	-
Australian coal (13)	-	0.8 - 3.1	-	-
Australian coal ash (13)	-	14	-	-

Table 2

Estimates of radionuclide releases from coal fired power plants in Italy

Type of coal and installed power	Activity released - mCi/y ^{238}U	^{226}Ra	^{232}Th	^{210}Pb	^{222}Rn
Italian lignite 250 MW	30	30	120	100	300
Italian lignite 70 MW	34	34	50	50	-
Italian lignite 500 MW (to be started)	400-2000	400-2000	-	-	-
Imported coal 1000 MW (1979-1985 values)	-	40-160	-	-	-

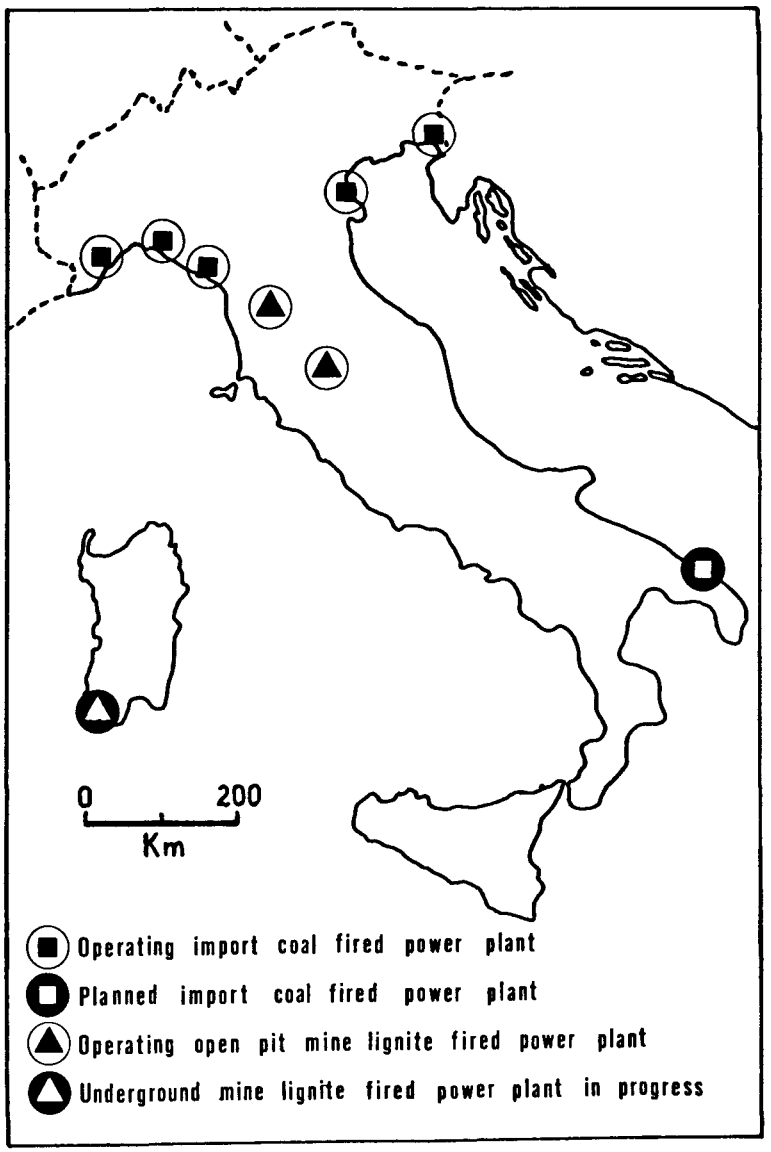

Fig. 1

Location of lignite mines, lignite fired power plants and coal fired power plants in Italy.

THE RADIOLOGICAL IMPACT OF GEOTHERMAL ENERGY

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SUMMARY. Geothermal energy is generally considered a very promising "alternative energy", even if its availability is restricted to certain areas characterized by very particular geological conditions and the power obtainable from it is very limited.
Despite other favourable features, geothermal energy has consistently proved to have an environmental impact which includes radiological aspects. Geothermal fluids when brought to the surface do in fact release to the environment very large amounts of Rn-222. In Italy, in an area of about fifty square kilometers about 3 kCi of Rn-222 are discharged yearly.
A variety of examples are reported to show in detail the radiological impact of the existing geothermal plants which have been in service for many years or are now coming into production in different regions of Italy.

RESUME. L'IMPACT RADIOLOGIQUE DE L'ENERGIE GEOTHERMIQUE. On considère généralement l'énergie géothermique comme une "alternative" très prometteuse même si elle n'est disponible qu'en des zones que caractérisent des conditions géologiques très particulières et si la puissance fournie est très limitée.
Malgré d'autres aspects favorables, l'énergie géothermique se révèle avoir généralement un impact sur l'environnement aussi sur le plan radiologique.
En effet, les fluides géothermiques qui parviennent en surface, libèrent dans l'environnement une très grande quantité de Rn-222. En Italie, dans une zone d'une cinquantaine de km^2, environ 3 kCi de Rn-222 sont rejetés chaque année.
Divers exemples exposés montrent amplement l'impact radiologique des centrales géothermiques en service depuis plusieurs années ou entrant maintenant en exploitation dans différentes régions d'Italie.

KURZFASSUNG. RADIOLOGISCHE AUSWIRKUNGEN DER NUTZUNG GEOTHERMISCHER ENERGIE.
Die geothermische Energie wird allgemein als eine vielversprechende "Alternativenergie" angesehen, auch wenn sie nur in bestimmten Gebieten mit ganz besonderen geologischen Bedingungen zur Verfügung steht und die aus ihr gewinnbare Leistung sehr begrenzt ist.
Trotz der Vorteile der geothermischen Energie weiss man aus Erfahrung, dass sie Auswirkungen auf die Umwelt hat, die auch radiologische Aspekte mit einschliessen.

Wenn geothermische Ströme an die Erdoberfläche gelangen, geben sie nämlich sehr grosse Mengen Rn-222 an die Umwelt ab. In Italien werden auf einem Gebiet von ungefähr 50 km^2 jährlich ca. 3 kCi Rn-222 abgeleitet. Anhand von mehreren Beispielen wird berichtet, wie sich die geothermischen Anlagen, die in verschiedenen Regionen Italiens seit mehreren Jahren in Betrieb sind bzw. deren Inbetriebnahme bevorsteht, aus radiologischer Sicht auswirken.

INTRODUCTION

Geothermal energy is an abundant resource, and its practical applications demonstrated economically very convenient. At present time only a small fraction of the world energy demand is produced by geothermal sources (3%), but amongst the alternative energies it is believed to be one of more realistic to be employed on a large scale on the short-term(1).

The potential resources of geothermal energy are believed to be very large and the technology rather well established; the environmental problems involved instead revealed to be much worse than initially outlined.

In Fig. 1 are showed the areas of exploration and exploitation of geothermal energy on a worldwide scale. As it can be seen Italy occupies a rather preminent place with at least three large geothermal fields, two of which operating power plants of 400 MW and 15 MW respectively.

In the framework of the program established to evaluate comparatively the environmental impact of the various energy sources in Italy, some work has been carried out also on the radioactive airborne waster disposed by geothermal plants.

GEOTHERMAL ENERGY AND NATURAL RADIOACTIVITY

Geothermal energy may be used for power generation through different technologies, depending on the thermodynamic conditions and composition of the endogenous fluids. The production of hot steam may be obtained either naturally or artificially following various mechanisms (see Fig.2). In all these mechanisms, mobilization of natural radionuclides is involved, especially of Radon-222 which is easily leached by water from the bedrock. Other natural radioactive elements (e.g. Radium-226) have been often found in geothermal fluids at levels higher than normal, while uranium and thorium always showed levels even lower than normal ones.

The three italian large geothermal fields exploited for energy production, are (see Fig.3):

a) Larderello in Tuscany;

b) Monte Amiata at the border between Tuscany and Latium;

c) Campi Flegrei near Naples.

The first field is well known since more than a century and hosts a 400 MW power plant since about 50 years. Recent work (2) reviewed the Radon-222 levels in geothermal fluids for all the wells of the field (Fig.4). From such results it has been calculated an emission into the atmosphere of about 3 KCi/y of Radon-222 from the stack of the power plants which are located at Larderello.

The second field is located around Monte Amiata and has installed two small power plants: a 3 MW one at Bagnore and a 15 MW at Piancastagnaio (see Fig. 5). They releases about 40 and 190 Ci/Y of Radon-222 (3).

The only relevant finding about the areas around these geothermal plants consists in the results of a survey on Lead-210 and Polonium-210 levels in the diet of the village of Abbadia-San Salvatore only 4 km away from Piancastagnaio plant (4). The levels found for Lead-210 and Polonium-210 in fact, are significantly higher than those found for other italian towns by a factor of more than two (see Table 1). No data are available for the other sites, except some preliminar results of a joint survey DOE--ENEL on Radon-222 levels in the area of Larderello, showing values very close to those encountered in normal background areas (5).

Monte Amiata is a normal background area, and the natural emission rate of Radon-222 from soil is estimated to be 100 aCi/cm^2 sec. The plant stack point emission of about 190 Ci/y of Radon-222 is therefore equivalent to the activity naturally emanated from soil in the same period and on an area of about 10 km^2. The activity released by the power plant, if deposited in the surroundings, might therefore double the background concentration of Radon-222 and its daughter products. The metheorological characteristics of the area, very frequent dew and long period of snow cover in winter as well as the interaction of the wind with the orography, may contribute to prevent diffusion of the power plant gaseous discharge and to originate a consistent fall-out. Moreover the area aound the power plant (see Fig.5) is extensively contivated and the products are almost in-loco consumed. In the past years, in fact, a socioeconomical study of the area has determined that the population of the village Abbadia-San Salvatore (about 8500 inhabitants) is supplied almost with locally produced foods (cereals, fruits, meat, milk etc.) (6). It seems thus reasonable to infer a possible correlation between the high Lead-210 and Polonium-210 levels found in the analyzed diets and the explotation of the geothermal power plant.

CONCLUSION

The data presented above are only a starting point to achieve a clear understanding of the transfer of Radon-222 and its daughter products from the stack of a geothermal power plant to the food-chain. The exposure of the population to such eventual fall-out seems to be not relevant, especially if compared with the environmental and health impact of toxic stable elements (e.g. As, Hg, etc.) frequently found at high concentration in geothermal fluids.

Nonetheless future programs include a thorough survey of Radon-222 levels in air and of Lead-210 and Polonium-210 in the food-chain all around the area of Monte Amiata, as well as the area of Larderello, even if, in this last case, the first data do not reveal any particular result (5).

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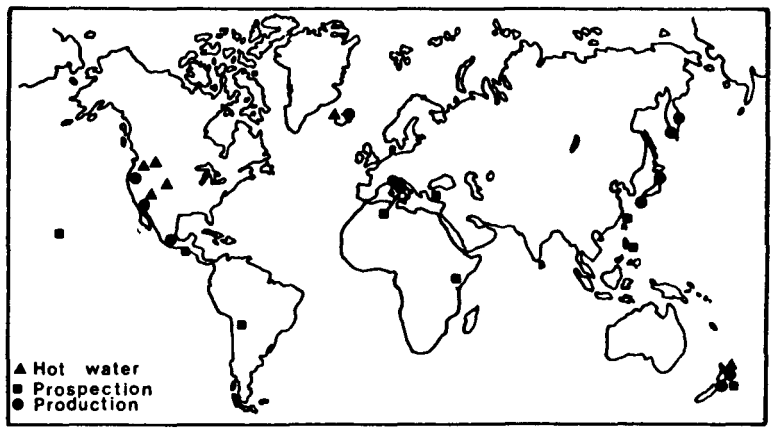

Fig. 1

Worldwide location of geothermal fields for prospection and energy production.

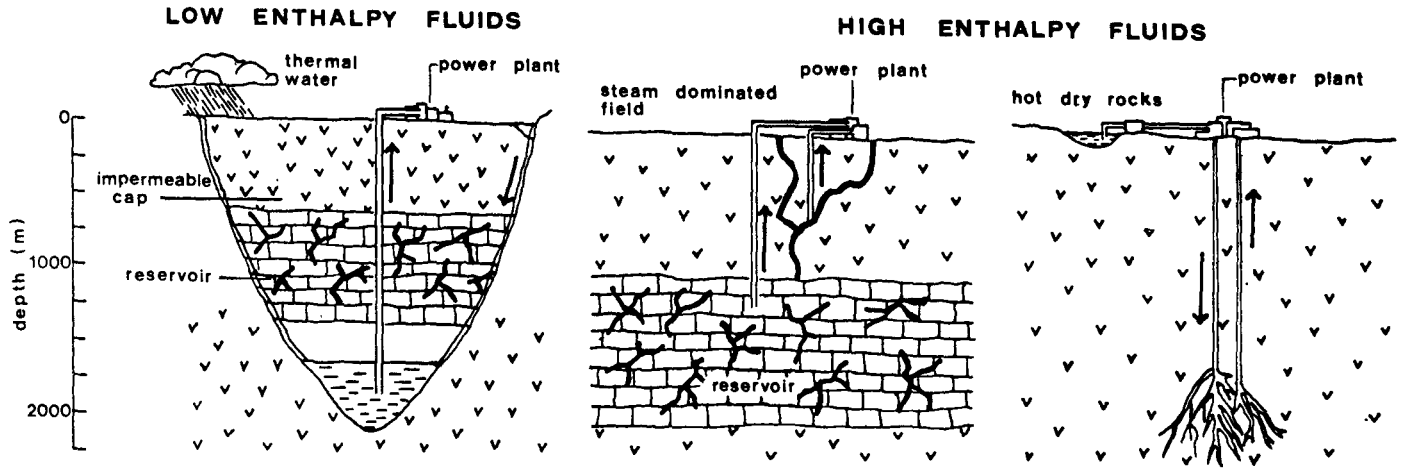

Fig.2

The three main geothermal mechanisms for power production.

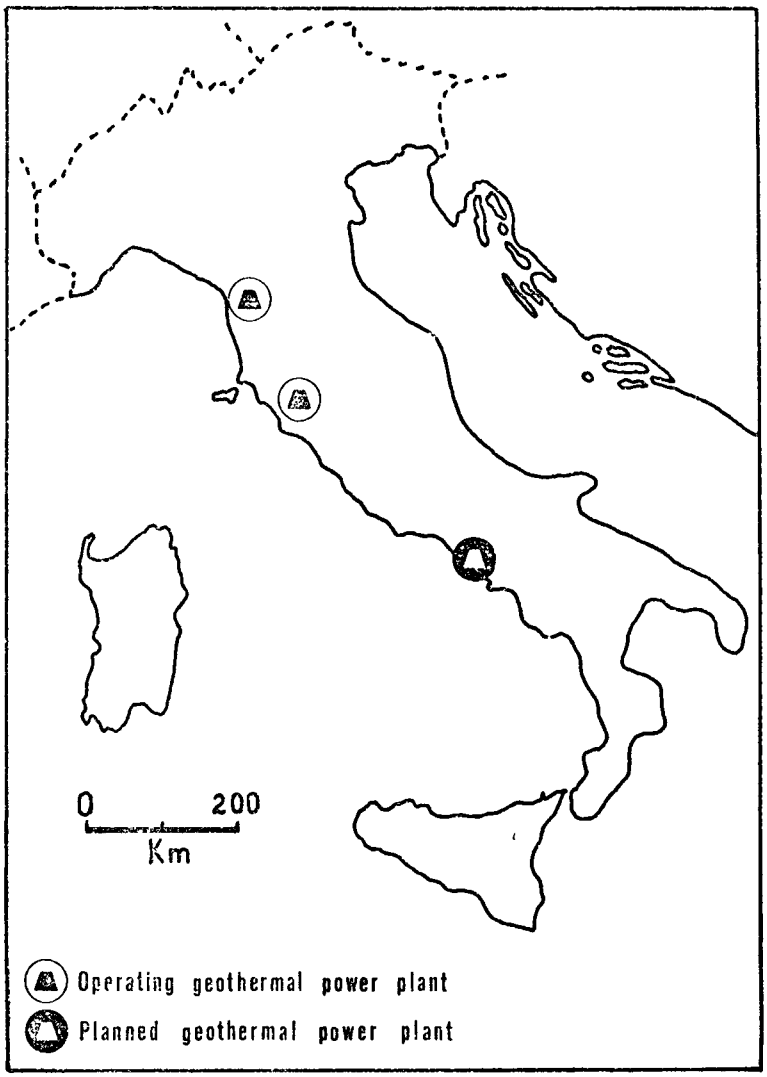

Fig. 3

Location of the three largest geothermal fields in Italy.

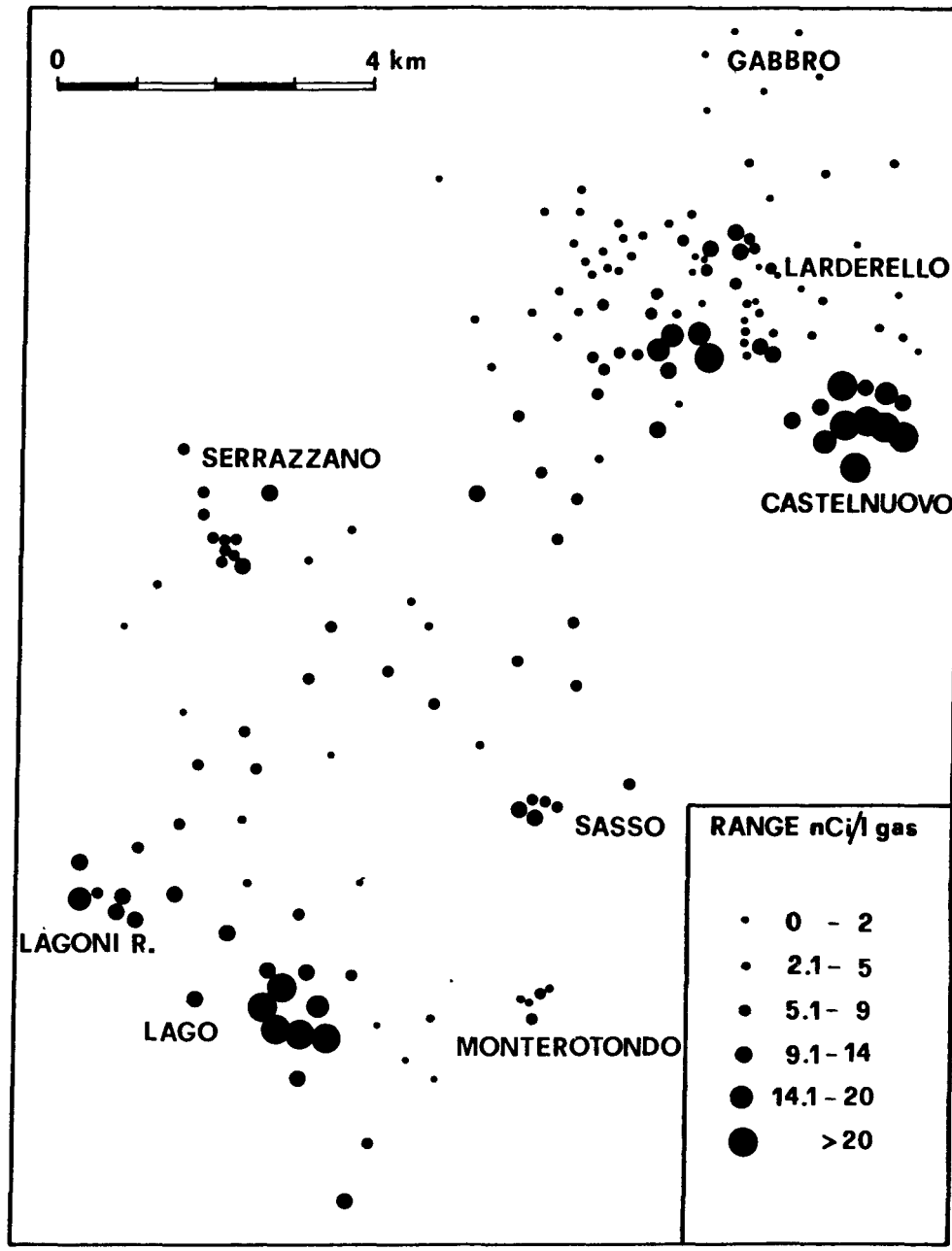

Fig. 4

Location and Radon-222 concentration in the fluid of the Larderello geothermal field wells.

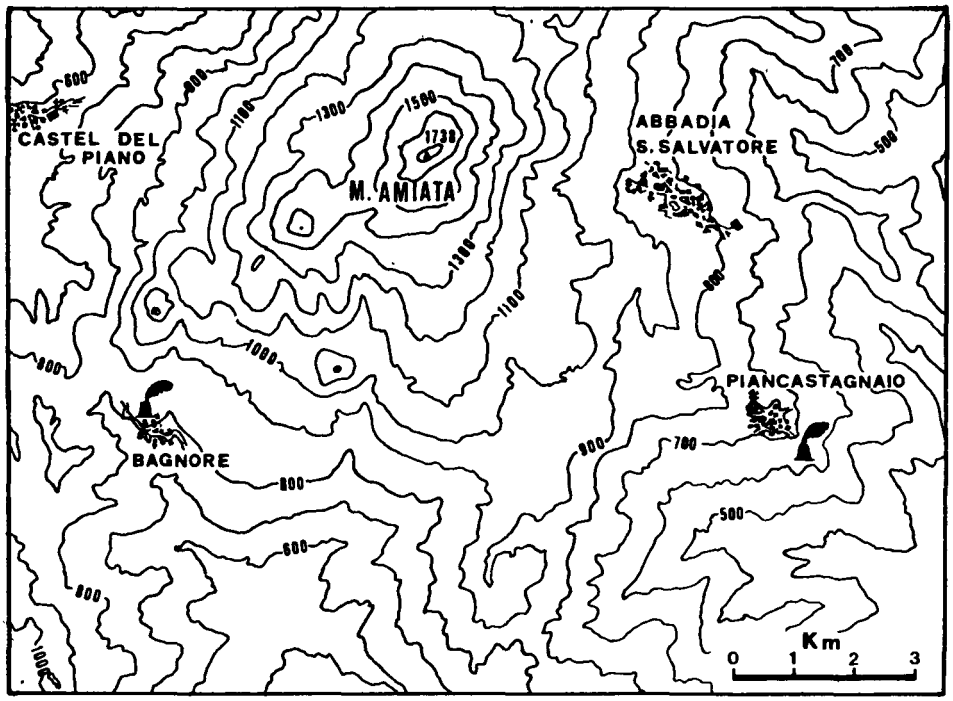

Fig.5

Map of the Monte Amiata geothermal field.

EXTERNAL RADIATION EXPOSURE DUE TO NATURAL RADIONUCLIDES IN PHOSPHATE FERTILIZERS IN THE FEDERAL REPUBLIC OF GERMANY

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SUMMARY. In the F.R. of Germany the annual consumption of mineral phosphate fertilizers amounts to about 1 million tons P_2O_5. Most of these fertilizers contain the natural radionuclides of the uranium series and potassium-40 in specific activities appreciably exceeding the average specific activities found in common soils, thus possibly contributing to the technologically enhanced exposure to natural radiation. The purpose of our study presented here was to estimate the contribuation of this radioactivity to the external radiation exposure of members of the public and of persons occupationally involved in handling rock phosphates or phosphate fertilizers. The natural radionuclide content of about 70% of the mineral phosphate fertilizers used in the F.R. of Germany has been determined gamma-spectrometrically. From the results, and allowing for the intensity of use of fertilizers and for residence times in agricultural or horticultural areas, an average annual exposure of 0.001 mrem/y (whole body) may be estimated for members of the public, with local maximum values of about 0.1 mrem/y. This implies a contribution of 186 man-rem/y to the population dose in the F.R. of Germany.

Appreciably higher exposure values have been obtained for the small group of persons occupationally involved in handling rock phosphates or phosphate fertilizers. By direct measurements or by calculations from specific activities in production, transport, loading and storage of rock phosphates and phosphate fertilizers and in agriculture, we have estimated mean annual doses to individuals of from 0.05 mrem/y (agriculture) to 20 mrem/y (production) with maximum values of up to 45 mrem/y. By taking into account the number of persons employed and annual occupation times or the average annual work output in the various occupations, an average contribution to the population dose of 174 man-rem/y (whole body) resulted.

In total, the external exposure due to natural radionuclides in phosphate fertilizers contributes only 360 man-rem/y to the population dose in the F.R. of Germany, which is negligibly small compared to the mean value due to terrestrial radiation of about 3.10^6 man-rem/y. For a small group of persons, however, occupational exposure may be incurred equivalent to the mean terrestrial exposure.

RESUME. EXPOSITION EXTERNE AUX RAYONNEMENTS EMIS PAR LES RADIOISOTOPES NATURELS DES ENGRAIS PHOSPHATES UTILISES DANS LA REPUBLIQUE FEDERALE D'ALLEMAGNE.
En R.F. d'Allemagne, la consommation annuelle d'engrais phosphatés minéraux est d'environ 1 million de tonnes de P_2O_5. Comme la plupart de ces engrais ont des activités spécifiques en radionucléides de la série de l'uranium et en potassium-40 bien supérieures à celles des sols, on peut s'interroger sur leur contribution à un accroissement de la dose d'irradiation naturelle lié à la technologie. Le but de la présente étude est d'évaluer cette contribution à l'irradiation externe de la population dans son ensemble et des personnes manipulant de ces engrais dans leur métier. On a mesuré par spectrométrie gamma les activités spécifiques d'environ 70% des engrais phosphatés minéraux utilisés en R.F. D'Allemagne. Des quantités moyennes employées de ces engrais et des estimations de la durée de séjour en zones rurales, on en a déduit une exposition annuelle moyenne de la population de 0,001 mrem/a (dose globale) avec, par endroits, des maxima de 0,1 mrem/a. Il en résulte une contribution à la dose population de 186 homme-rem/a.
Le groupe restreint des personnes manipulant des phosphates bruts ou des engrais phosphatés dans leur métier reçoit des doses d'irradiation externe nettement plus élevées. En tenant compte des doses locales supplémentaires - tantôt mesurées directement, tantôt calculées sur la base des activités spécifiques - reçues par les travailleurs des secteurs production, transport, chargement et stockage de phosphates bruts et d'engrais phosphatés ainsi que du secteur agricole, on a obtenu des moyennes annuelles d'exposition aux rayonnements comprises entre 0,05 mrem/a (agriculture) et 20 mrem/a (production), avec des pointes de 45 mrem/a. Compte tenu du nombre des salariés et du temps de travail annuel ou du rendement annuel moyen dans les différents secteurs, la contribution moyenne à la dose population s'élève à 174 homme-rem/a (dose globale).
Au total en R.F. d'Allemagne l'exposition de la population au rayonnement externe émis par les radionucléides naturels des engrais phosphatés n'est que de 360 homme-rem/a ce qui, comparé à la charge radiologique moyenne due au rayonnement terrestre qui est d'environ 3.10^6 rem/a, est négligeable. Cependant, un nombre restreint de personnes professionnellement exposées sont susceptibles de recevoir des doses correspondant à celle due au rayonnement terrestre.

KURZFASSUNG. AUSSERE STRAHLENEXPOSITION DURCH NATÜRLICHE RADIONUKLIDE IN PHOSPHATDÜNGEMITTELN IN DER BUNDESREPUBLIK DEUTSCHLAND. In der Bundesrepublik Deutschland beträgt der jährliche Verbrauch an mineralischen Phosphatdüngemitteln etwa 1 Million P_2O_5. Da die meisten dieser Düngemittel spezifische Aktivitäten der Radionuklide der Uranreihe und Kalium-40 aufweisen, die weit höher sind als die Werte in den Böden, muss ein Beitrag zur technologisch erhöhten Exposition durch natürliche Strahlung in Betracht gezogen werden. Das Ziel unserer vorliegenden Untersuchung war es daher, diesen Beitrag zur externen Strahlenexposition von Mitgliedern der Bevölkerung und von Personen, die beruflich mit Phosphatdüngemitteln umgehen, abzuschätzen. Aus den gammaspektrometrisch bestimmten spezifischen Aktivitäten von etwa 70% der in der Bundesrepublik verwendeten mineralischen Phosphatdünger, dem durchschnittlichen Phosphatdüngeraufwand und abgeschätzten Aufenthaltszeiten in landwirtschaftlichen Bereichen ergab sich eine durchschnittliche Strahlenexposition für Mitglieder der Bevölkerung von 0,001 mrem/a (Ganzkörper) mit lokalen Maximalwerten von bis zu 0,1 mrem/a. Daraus resultiert ein Beitrag zur Bevölkerungsdosis von 186 Mann-rem/a. Deutlich höhere externe Strahlenexpositionen wurden für die kleine Gruppe von Personen erhalten, die beruflich mit Rohphosphaten oder Phosphatdüngern umgeht. Aus den zusätzlichen Ortsdosisleistungswerten - direkt gemessen oder aus spezifischen Aktivitäten berechnet - in den Arbeitsbereichen Produktion, Transport, Verladung und Lagerung von Rohphosphaten und Phosphatdüngemitteln sowie Landwirtschaft ergaben sich mittlere jährliche Strahlenexpositionen von 0,05 mrem/a (Landwirtschaft) bis 20 mrem/a (Produktion) bei Maximalwerten bis zu 45 mrem/a. Berücksichtigt man die Anzahl der Beschäftigten und die jährliche Arbeitszeit oder die mittlere jährliche Arbeitsleistung in den verschiedenen Arbeitsbereichen, so erhält man einen durchschnittlichen Beitrag zur Bevölkerungsdosis von 174 Mann-rem/a (Ganzkörper).
Insgesamt trägt somit die externe Strahlenexposition durch die natürlichen Radionuklide in Phosphatdüngern nur 360 Mann-rem/a zur Bevölkerungsdosis in der Bundesrepublik bei. Dieser Wert ist vernachlässigbar klein - verglichen mit der mittleren Bevölkerungsdosis durch die terrestrische Strahlung von etwa 3.10^6 Mann-rem/a. Für eine kleine Personengruppe können jedoch beruflich bedingte Strahlenexpositionen in Höhe des Wertes der terrestrischen Strahlenexposition auftreten.

1. Introduction

Most of the rock phosphate processed to phosphate fertilizers in the FRG is imported from the large sedimentary phosphorite deposits in North Africa and in Florida / USA (1). These deposits of marine origine had been formed in the Tertiary Period by physico-chemical and, to the greatest extent, by biological concentration and deposition of phosphate which had been leached from igneous rocks and washed to the sea (2). In the course of deposition, uranium dissolved in the form of uranyl complexes in the sea water concentrated in the phosphate sediments (3). Therefore, sedimentary rock phosphate may contain appreciable amounts of uranium and its decay products (4, 5, 6, 7). Through the various stages of production uranium with its decay products may reach the commercial fertilizers used in agriculture. Furthermore, radium as a decay product of uranium from rock phosphates enters the technical gypsum which is precipitated during the sulfuric acid treatment of rock phosphate (sc. (5)). In view of the large amounts of phosphate fertilizers annually produced and distributed over the agricultural area of the FRG - that is 55 % of the entire area - radioactive materials in rock phosphates and phosphate fertilizers may contribute to the radiation exposure of persons occupationally handling phosphates and of members of the public. Besides uranium and thorium with their decay products, potassium-40 which is contained in mixed phosphate fertilizers has to be considered. For example, about 60 % of the annual potassium fertilizer consumption in 1977 / 78 was due to mixed phosphate fertilizers (8).

In our study which has been supported by the 'Bundesministerium des Innern' of the FRG, we estimated the external radiation exposure due to radioactive materials in mineral phosphates of members of the public and of the small group of persons working in production, transport, storage and application of rock phosphates and phosphate fertilizers and the respective contributions of the population dose in the FRG.

2. Radionuclide content of phosphate fertilizers

The activities of natural uranium, radium-226, natural thorium and potassium-40 have been determined gammaspectrometrically by use of a Ge(Li) detector (Harshaw ACO 33) in connection with a 2000 channel analyzing system ELVIRA 4-16E (Frieseke & Hoepfner). Our measurements were made on 86 commercial phosphate fertilizers comprising about 70 % of the mineral phosphate

fertilizer species used and authorized in the FRG, if allowance is made for special nutrient mixed fertilizers of the same manufacturer (9). For comparison, some samples of rock phosphates of various origines have been included. Experimental methods and results have been published in detail elsewhere (10).

Altogether, our results have shown, that the specific activities of uranium and radium are relatively high in most phosphate fertilizers as compared to the values in the most common geological formations and soils (7, 11), whereas the thorium contents are generally lower. As maximum values we found 62 nCi/kg for natural uranium and 23 nCi/kg for ^{226}Ra. The specific U_{nat} activities reflected the influence of the different phosphate types used in production. Fertilizers showing low uranium content are based on basic slag or, entirely or partly, on Kola apatite.

High uranium contents indicate the use of sedimentary rock phosphates. The uranium from the rock phosphates is probably nearly completely retained in the fertilizers. In contrast, the specific radium activity is significantly lower than that of uranium indicating that this nuclide is no longer in radioactive equilibrium with its parent ^{238}U in contrast to our findings in the rock phosphates. This reflects the influences of the diverse chemical rock phosphate processing methods. (For details see (10)).

Our results are summarized according to the six main phosphate fertilizer groups by the mean values of the phosphate content related specific activities given in Table I.

The delivery proportions for agricultural consumption in 1977 / 78 (8) has been used as a measure of the relative application frequency of the phosphate fertilizer groups. Last line of this table gives the over-all mean values of the specific activities of phosphate fertilizers weighted by their relative contribution to the agricultural phosphate consumption.

3. External exposure of members of the public due to radionuclides in phosphate fertilizers

3.1 Contribution of radionuclides contained in phosphate fertilizers to the natural radioactivity in agricultural areas

From the mean values of the specific activities of natural radionuclides in phosphate fertilizers, from the agricultural consumption of 0.873 million

9)

tons P_2O_5 (8) and from the average fertilizing intensity of 66.1 kg P_2O_5 per ha (= 10^4 m^2) in 1977 / 78 (8), one can derive, that about 61 Ci of natural uranium, 41 Ci of radium-226, 2 Ci of natural thorium, and 550 Ci of potassium-40 are distributed over the entire agricultural area of the FRG in 1977 / 78 due to phosphate fertilizing, or, per ha, 4.6 µCi of natural uranium, 3.1 µCi of radium-226, 0.2 µCi of natural thorium, and 41.6 µCi of potassium-40 on the average. With a maximum fertilizing intensity which has been reported to be 1525 kg P_2O_5 per ha in 1973 / 74 in the region of Würzburg (12) one arrives at 106.0, 71.4, 3.4, and 961 µCi/ha for U_{nat}, ^{226}Ra, Th_{nat} or ^{40}K respectively.

It must be considered, however, that radium may be bound to the soil matrix to a relatively high extent over many years, as it is known for the other earth alkali ions (13). This would result in an accumulating of radium from phosphate fertilizers in the soil due to the annual fertilizing. With appropriate assumptions (10), we estimated the whole radium activity, deposited in the soil with phosphate fertilizers, to 120 µCi / ha on the average.

Table II summarizes the estimated values of the activities per unit area of agricultural land due to phosphate fertilizing.

The natural uranium and radium content of unsupported soils in the FRG is in the range of 0.1 to 1.0 nCi / kg, except for some regions with unusually high uranium content. The calculated mean specific activities distributed per unit area with phosphate fertilizers, yielded an increase of the specific activities in the soil of about 1 pCi / kg with a maximum of about 26 pCi / kg in the region of the highest fertilizing intensity by assuming homogeneous distribution of the fertilizers in the soil down to the mean ploughing depth of 20 cm. If complete retention in the soil is assumed over the last 80 years, this increase would be about 44 pCi / kg on the average. This means, that the specific activities of uranium and radium in the soils should be increased by a few percent only on the average and by ca. 50 % at the utmost.

In limited agricultural areas, however, essentially higher values may occur. For example, if one considers a fertilizing intensity of 2000 kg P_2O_5 per ha, which is supported as stock fertilizing to vineyards occasionally (14), and the maximum specific activity of uranium and radium of 120 nCi / kg P_2O_5 in the fertilizer, one arrives at an increase of the specific soil activity of 100 pCi / kg.

3.2 External exposure

The additional exposure rate above phosphate fertilized soils was calculated by assuming that the activity of the natural radionuclides contained in phosphate fertilizers were homogeneously distributed in the soil down to the mean ploughing depth of 20 cm. Wash-out effects, elimination by plant metabolism or emanation of radon from the soil were neglected and radioactive equilibrium of radium and thorium with their gamma emitting daughter products was assumed in the soil. Attenuation of the gamma radiation in soil and air as well as build-up effects on the photon flux densities in the soil were taken into account. Then, the exposure rate in 1 m height above the soil is given by the following equation:

$$\dot{J} = \frac{10^{-3} m}{2 \cdot 10^{9} \cdot \rho_{eff}} \sum_{i} a_i \Gamma_i \qquad \text{in } \mu R/h, \qquad (1)$$

where

m is the amount of phosphate fertilizer applied, in kilograms P_2O_5 per hectare;

a_i the specific activity of the radionuclide i in the phosphate fertilizers, in nCi / kg P_2O_5;

Γ_i the "specific gamma-ray constant" of the radionuclide i in 1 m height above soil for the geometry given here (infinitely extended radiating disc of 20 cm thickness), in R / h per Ci / g;

ρ_{eff} the effective density of the soil, in g / cm^3.

The Γ_i-values were calculated for the geometry assumed by applying the appropriate values of the mass attenuation coefficients of soil and air, of the mass energy absorption coefficients of air and of approximated build-up factors of soil at the various photon energies. These data were derived from the chemical composition of a typical sand soil with an effective density of 1.35 g / cm^3 and a mean water content of 5 %. The calculations, performed at a PDP 8/e computer, yielded the following Γ_i-values:

$1.93 \cdot 10^6$ R · g/Ci · h for ^{226}Ra in equilibrium with decay products,

$2.52 \cdot 10^6$ R · g/Ci · h for ^{232}Th in equilibrium with decay products,

$1.63 \cdot 10^5$ R · g/Ci · h for ^{40}K.

Together with the mean specific activities of the phosphate fertilizers of 46.8 nCi ^{226}Ra, 2.2 nCi Th_{nat} and 630.0 nCi ^{40}K per kg P_2O_5 (see Table I) and a mean amount of fertilizers applied of 66.1 kg P_2O_5 / ha during 1977 / 78 in the FRG (8) one obtains an additional exposure rate of $4.9 \cdot 10^{-3}$ μR / h from eq.(1).

By admitting an eventual accumulation of radium in the soil by mineral phosphate fertilizing during the last 80 years one obtains a mean additional exposure rate of $8.8 \cdot 10^{-2}$ μR/h. Essentially higher values may appear locally in areas with especially high amounts of phosphate fertilizers applied. Assuming a maximum value of 2000 kg P_2O_5/ha and specific activities as high as 120 nCi ^{226}Ra / kg P_2O_5 and 1000 nCi ^{40}K / kg P_2O_5 results in a maximum local additional exposure rate of 0.3 μR / h.

Allowing for a mean residence time of members of the public of 1 hour a day in agricultural areas and taking into account a dose conversion factor of 0.561 rd/R for whole body (15) and a quality factor of 1 one obtains a value of 0.001 mrem/a as an average annual exposure. In regions with maximum fertilizing intensity this value would be raised to 0.06 mrem/a. With the assumption of complete accumulation in the soil we calculated an average exposure of 0.018 mrem/a.

4. Occupational external exposure due to phosphate fertilizers

4.1 Exposure rates

For the working fields production, transport, storage and application of rock phosphates and phosphate fertilizers the exposure rates have been determined by measurements and, where this was impossible, by estimates from measured values at comparable locations or by calculations based on adequate models and measured specific activities.

The direct measurements of exposure rates have been performed by use of the portable scintillation dose rate meters H 7201 (Eberhard Halle, Braunschweig, FRG) and MAB 601 (Münchener Apparatebau Kimmel KG., Ottobrunn, FRG). For the measurement of the exposure and of the personal dose LiF-dosimeters have been used.

Direct measurements were done at 9 phosphate production plants, at two representative large agricultural storehouses, on a barge loaded with Marokko

phosphate, in a freighter loaded with 20000 tons of Florida phosphate and at railway and truck loading stations. Exposure rates above agricultural areas have been calculated according to section 3.2 . From this, the average additional exposure rates at the remaining locations and working fields were estimated on the basis of appropriate assumptions (for details see (16)).

Our results, as expressed as mean and maximum additional exposure rates, are listed in Table III according to the various fields of activity. These values exceed the mean terrestrial exposure rate in the FRG by a factor up to 4 to 6. At some locations, exposure rates up to 12 times the mean terrestrial exposure rate can arise.

4.2 Number of occupied persons, annual occupation times and annual work output in phosphate fertilizer working fields

In order to obtain mean and annual exposures of individuals due to occupationally working with or handling rock phosphates and phosphate fertilizers and their contributions to the mean population dose, data for the number of persons occupied in the respective working fields and for their mean and maximum occupation times must be known besides of the mean and maximum exposure rates given above. For the working fields production plants, agricultural storehouses and agriculture, these data could be obtained separately from available statistics (17), by questioning of the respective companies, organisations or authorities (18, 19) and by appropriate estimates of the occupation time per year.

As to transport of phosphates, however, only the total work output per year (in man · hours per year) and maximum individual occupation times per year could be estimated. Our approximations started from the detailed freight and transport statistics (20) for each transport branch from which the annual transport outputs (in tons · kilometers) are calculable. By use of appropriate estimates of freight quantities and occupied persons per transport unit and of mean transport velocities the annual transport outputs in tons · kilometers per year could be transformed to transport work outputs per year in man · hours per year.

The corresponding data for loading and unloading were calculated from the total quantities in each transport branch and from assumptions on freight quantities and work expense (in man · hours) needed for loading and unloading per transport unit.

Finally, from average transport velocities and maximum transport distances, maximal annual exposure times in the various transport branches were estimated.

Table IV summarizes the estimated values for mean annual work output and for mean and maximum annual occupation times according to the various working fields.

4.3 Occupational exposure of individuals and occupational collective dose

The annual occupational exposures of individuals due to phosphate fertilizers and the related occupational annual collective doses were obtained from the exposure rates and from the annual exposure times and the annual work output respectively as estimated above. Dose conversion factors of 0.561 rd/R for whole body and of 0.695 rd/R for male gonads according to O'Brien and Sanna (15) and a quality factor of 1 have been applied.

For the working fields production, storehouses and agriculture the values of the mean and maximum exposure of individuals can be evaluated from the data of Tables III and IV. For transport and loading, only maximum values could be calculated. The results for whole body are listed in column 2 of Table V. The corresponding values for male gonads are 24 % higher.

The highest occupational exposure of individuals due to rock phosphates and phosphate fertilizers are to be expected in fertilizer production plants and in agricultural storehouses, resulting in about doubling of the mean terrestrial exposure in the FRG. Relatively high values may occur at long-distance road transport of rock phosphates, whereas the maximum exposure of persons occupied in agriculture is within the variation of the terrestrial exposure.

The annual occupational collective doses due to rock phosphates and phosphate fertilizers were calculated from the exposure rates and the annual work output in the respective phosphate working fields. The results are given in column 3 of Table V. The values in brackets are obtained with the assumption of total accumulation of radium from phosphate fertilizers in the soils during the last 80 years (10, 16). As can be seen from Table V, the working fields agriculture, production, storage and local road transport contribute nearly equally to the occupational collective dose due to phosphate fertilizers, whereas the contribution of long-distance transport by

ship or via roads is very small. The total contribution to the population dose caused by occupationally working with or handling rock phosphates and phosphate fertilizers is 174 man · rem / y and 215 man · rem / y related to whole body and male gonads respectively. This is about three times the population dose due to exposure of the whole population to the gamma radiation from phosphate fertilizers applied in the agricultural areas of the FRG (62 man · rem / y related to whole body), which can be estimated from the results in section 3.2 .

5. Conclusions

Our results show that natural radionuclides contained in phosphate fertilizers contribute very little to the external radiation exposure of members of the public, even if complete retention of radium deposited with phosphate fertilizers in the soil during the last 80 years is assumed.

From the estimates given above one may conclude that the contribution of natural radionuclides contained in phosphate fertilizers to the internal radiation exposure of members of the public should also be very low compared to that from other natural radiation sources.

In contrast, the occupational external exposure of individuals due to phosphate fertilizers may reach values up to about 50 mrem / y. This means a doubling of the mean terrestrial radiation exposure and a 50 % increase of the natural radiation exposure. Furthermore, the internal radiation exposure of persons working in agriculture, in fertilizer storehouses or in production plants has to be considered. The inhalation of radon and of phosphate fertilizer dust may increase the internal exposure remarkably.

The mean population dose due to terrestrial radiation in the FRG is about $3 \cdot 10^6$ man · rem / y. The value of 240 man · rem / y due to natural radionuclides in mineral phosphates is very small compared to this.

The "man · rem concept" used in this discussion does not lend itself strictly to an evaluation of an additional radiation risk of the population. Following Jacobi (21) this would be possible only for comparison of risks for components of the civilisatory radiation exposure whose contributions to the total dose are small compared to the natural radiation exposure, or for a risk comparison of collectives with the same relative dose distribution. None of these two assumptions holds here. Nevertheless, one can con-

clude from our estimates that the additional radiation risk of the total population due to natural radionuclides in phosphate fertilizers should be negligibly small. For some individuals, especially in fertilizer production plants or agricultural storehouses, however, an increase of the radiation risk may not be excluded.

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Table I: Mean specific activities in phosphate fertilizers.

Type of fertilizer	Agricultural consumption in 1977/78 %	Mean specific activity in nCi / kg P_2O_5			
		U_{nat}	^{226}Ra	Th_{nat}	^{40}K
Superphosphat	7.3	77.7	78.1	2.4	20.4
Basic slag	15.2	(1.0)	1.0	0.3	0.6
calcined, ground soft, partly converted rock phosphate etc.	3.8	64.2	46.4	2.5	10.9
PK - Fertilizers	25.9	67.7	61.7	2.2	1011.3
NP - Fertilizers	12.8	88.7	30.4	2.7	4.0
NPK - Fertilizers	35.0	92.4	55.1	2.9	1044.4
Phosphate fertilizers total	100.0	69.5	46.8	2.2	630.0

Table II: Activity per unit area due to radionuclides contained in phosphate fertilizers, in μCi / ha.

Radionuclide	Annual increase		Accumulated values (on the average)
	average	maximum	
U_{nat}	4.6	106.0	(174)
^{226}Ra	3.1	71.4	119
Th_{nat}	0.2	3.4	-
^{40}K	41.6	961.0	-

Table III: Additional exposure rates from rock phosphates and phosphate fertilizers.

Working field		mean µR/h	maximum µR/h
Production		18	40 (90)*
Transport and loading			
Sea transport			
Rock phosphates:	transport	3	(90)
	loading	10	(90)
Phosphate fertilizers:	transport	2	(60)
	loading	6	(60)
Inland ship transport			
Rock phosphates:	transport	10	(90)
	loading	26	(90)
Phosphate fertilizers:	transport	6	(60)
	loading	18	(60)
Railway transport			
Rock phosphates		26	(90)
Phosphate fertilizers		18	(60)
Transport via roads			
Rock phosphates:	transport	16	-
	loading	26	(90)
Phosphate fertilizers:	transport	6	-
	loading	10	(60)
Agricultural storehouses		10	40
Agriculture			
"outdoors"		$4.9 \cdot 10^{-3}$ ($8.8 \cdot 10^{-2}$) **	0.3
at storing, Fertilizing etc.		10	(60)

* values in brackets: local maximum which is not used in the further calculations, since the residence times are small at these locations

** estimated assuming total radium accumulation in soil

Table IV: Mean annual work expense and annual occupation times in phosphate fertilizer working fields

Working field		Mean annual work expense in man . hours / year	Annual occupation times in hours / year	
			mean	maximum
Production		$4 \cdot 10^6$	2000	2000
Transport and loading				
Sea transport				
Rock phosphates:	transport	$1.3 \cdot 10^5$	-	2900
	loading	$5.6 \cdot 10^4$	-	800
Phosphate fertilizers:	transport	$9.8 \cdot 10^3$	-	2900
	loading	$1.4 \cdot 10^4$	-	800
Inland ship transport				
Rock phosphates:	transport	$6.7 \cdot 10^5$	-	2700
	loading	$6.5 \cdot 10^4$	-	220
Phosphate fertilizers:	transport	$1.5 \cdot 10^5$	-	2700
	loading	$2.9 \cdot 10^4$	-	220
Railway transport				
Rock phosphates:		$1.3 \cdot 10^4$	-	2000
Phosphate fertilizers:		$1.3 \cdot 10^5$	-	2000

Table IV (continued)

Long-distance transport via roads				
Rock phosphates:	transport	$1.9 \cdot 10^4$	-	3000
	loading	$6.8 \cdot 10^3$	-	380
Phosphate fertilizers:	transport	$8.4 \cdot 10^5$	-	3000
	loading	$4.3 \cdot 10^5$	-	380
Local transport via roads				
Rock phosphates:	transport	$5.3 \cdot 10^4$	-	500
	loading	$5.3 \cdot 10^4$	-	500
Phosphate fertilizers:	transport	$3.3 \cdot 10^6$	-	500
	loading	$3.3 \cdot 10^6$	-	500
Agricultural storehouses		$5.6 \cdot 10^7$	2000	2000
Agriculture				
"Outdoors"		$7.7 \cdot 10^8$	310	1000
at storing, fertilizing etc.		$1.0 \cdot 10^7$	8	40

Table V: Occupational radiation exposure due to phosphate fertilizers

Working field		Annual dose to individuals (whole body) in mrem/y		Annual collective dose (whole body) in man · rem / y
		mean	maximum	
Production		20	45	40.4
Transport and loading: total		-	-	43.8
including:	Sea transport			
	- rock phosphates	-	9	0.5
	- phosphate fertilizers	-	6	0.1
	Inland ship transport			
	- rock phosphates	-	18	4.7
	- phosphate fertilizers	-	11	0.8
	Railway transport			
	- rock phosphates	-	29	0.2
	- phosphate fertilizers	-	20	1.3
	Long-distance transport via roads			
	- rock phosphates	-	33	0.3
	- phosphate fertilizers	-	12	5.2

Table V (continued)

Local transport via roads			
- rockphosphates	-	12	1.3
- phosphate fertilizers	-	5	29.4
Agricultural storehouses	11	45	31.4
Agriculture: total	0.05 (0.06)*	0.39	58.2 (94.1)
including: "outdoors"	0.001 (0.02)	0.17	2.1 (38.0)
at storing, fertilizing etc.	0.045	0.22	56.1
Total	-	-	173.8 (209.7)

* Values in brackets: calculated by assuming total radium accumulation in soils

RECHERCHES SUR LES EFFETS GENETIQUES DE LA RADIOACTIVITE NATURELLE ANORMALE OBSERVEE LOCALEMENT DANS LE SUD-OUEST DE LA FRANCE

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RESUME. Des débits de dose de 10 à 15 mrad/h de rayonnements d'origine naturelle se rencontrent localement en France ce qui dépasse considérablement ce qui a été observé dans la plupart des autres régions à haute radioactivité naturelle du monde. La capacité d'une telle radioactivité naturelle à produire des anomalies chromosomiques dans les cellules somatiques a été étudiée dans des lymphocytes de lapins maintenus, pendant 28 mois, sur le sol d'une hutte construite dans un endroit où le débit de dose atteint 8 mrad/h. Pour étudier les effets de la radioactivité naturelle sur la fertilité des mammifères nous avons utilisé des souris de race BALB/c qui ont été placées dans cette hutte durant l'été (4 mois).
Nos observations démontrent qu'une radioactivité naturelle élevée peut théoriquement produire des anomalies chromosomiques dans les cellules somatiques et qu'elle perturbe la fertilité des animaux directement exposés. Compte tenu de la superficie réduite des points à radioactivité élevée, la probabilité d'observer de tels effets chez l'homme vivant dans ces régions est nulle.
Ce travail a été réalisé dans le cadre d'un contrat de recherches Euratom-CEN no 274-79-1 BIO B.

KURZFASSUNG. UNTERSUCHUNGEN ÜBER DIE GENETISCHEN AUSWIRKUNGEN HÖHERER LOKALER PEGEL NATÜRLICHER RADIOAKTIVITÄT IN SÜDWESTFRANKREICH. Dosisleistungen von 10 bis 15 mrad/h zufolge natürlicher Strahlung treten örtlich in Frankreich auf und liegen damit erheblich über den in den meisten anderen Regionen der Welt mit erhöhter natürlicher Radioaktivität festgestellten Werten. Die Fähigkeit eines derart hohen Pegels der natürlichen Radioaktivität, in den somatischen Zellen Chromosomenaberrationen hervorzurufen, wurde an Lymphozyten von Kaninchen untersucht, die 28 Monate lang auf dem Boden einer Hütte an einem Ort gehalten wurden, an dem die Dosisleistung

8 mrad/h erreicht. Die Auswirkungen der natürlichen Radioaktivität auf die Fruchtbarkeit von Säugetieren haben wir an Mäusen der Rasse BALB/c untersucht, die den Sommer über (4 Monate lang) in dieser Hütte gehalten wurden.

Unsere Beobachtungen zeigen, dass eine hohe natürliche Radioaktivität Chromosomenanomalien in den somatischen Zellen hervorrufen kann und dass sie die Fruchtbarkeit der Tiere beeinträchtigt. Angesichts der begrenzten Ausdehnung von Flächen, wo eine erhöhte Radioaktivität vorliegt, besteht kaum eine Möglichkeit, analoge Beobachtungen bei der Bevölkerung dieser Region zu machen.

Diese Arbeit wurde im Rahmen des Forschungsvertrages Euratom-CEN Nr. 274-79-1 BIO B durchgeführt.

SUMMARY. RESEARCH INTO GENETIC EFFECTS FROM ABNORMAL NATURAL RADIOACTIVITY OCCURRING LOCALLY IN SOUTH WEST FRANCE. Dose-rates of 10-15 mrad/h from natural radiation occur locally in France, thus considerably exceeding values reported for the majority of other areas of high natural radioactivity in the world. The ability of such high natural radioactivity to produce chromosome aberrations in mammalian somatic cells was studied in lymphocytes from laboratory rabbits maintained, for a 28 month period, on the floor of a hut built at a site where the dose rate amounts to about 8 mrad/h. To study the effects of high natural radioactivity on mammalian germ cells, male and female mice of the BALB/c strain were placed in the hut during the summer period (4 months).

The results suggest that exposure to high natural radioactivity could theoretically, induce chromosomal aberrations in somatic cells and affect the fertility of animals directly exposed. Taking into account the minor extent of areas with high radiation levels, the probability of observing such effects in man is vanishingly small.

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1. INTRODUCTION

Les recherches réalisées jusqu'à présent, en Inde et au Brésil, sur les effets génétiques et cytogénétiques d'une radioactivité naturelle élevée ont donné des résultats plutôt disparates et même parfois contradictoires.

En 1964, Grüneberg [1] a analysé les caractéristiques génétiques d'une population de rats (*Rattus rattus L.*) vivant sur une bande étroite de terrain, de quelque 20 km de long, située sur la côte indienne de Malabar. Durant plus de 600 ans, cette bande de terrain était, en fait, une île sur laquelle se sont succédé de 800 à 1000 générations de rats. On peut donc considérer qu'il s'agissait d'une population en équilibre. La radioactivité de cette bande de terrain était environ 7,5 fois plus élevée que celle des îles voisines dont la population a servi de témoin. Chaque génération de rats a accumulé environ 1 rad de rayonnement γ durant sa vie reproductive, ce qui correspond à 1000 rad pour les 1000 générations. Durant cette même période, les témoins n'ont accumulé que 134 rad. Pour comparer les caractéristiques génétiques des deux populations de rats, Grüneberg a utilisé un total de 47 paramètres (6 dentaires et 41 squelettiques); il a également tenu compte de la mortalité embryonnaire. Il n'a cependant pu mettre en évidence aucune différence systématiques entre les populations.

Gopal-Ayengar et ses collaborateurs [2, 3] ont étudié des populations humaines vivant dans les régions à haute radioactivité naturelle de la côte de Kerala, au sud-ouest de l'Inde. Les populations de cette région reçoivent une dose annuelle moyenne de 397 mrad, ce qui correspond à quatre fois la dose normale, mais certains individus reçoivent jusqu'à 2 rad, c'est-à-dire 20 fois la dose normale. L'analyse des données démographiques ne leur a pas permis de mettre en évidence l'existence, entre les populations témoins et les populations exposées, de différences significatives dans la fertilité, les proportions des sexes, la mortalité périnatale, les avortements et les malformations importantes. Selon Kochupillai *et al.* [4], cependant, la fréquence de trisomie 21 et d'anomalies du même type serait plus élevée que la normale dans la population d'une région voisine où la radioactivité naturelle atteint de 1500 à 3000 mrad par an. L'échantillon de population étudié est trop petit et la fréquence des déficiences génétiques dans la population témoin trop faible pour justifier les conclusions de ces auteurs [5].

Etant donné les difficultés présentées par les études épidémiologiques

sur l'homme, certains auteurs ont réalisés des examens cytogénétiques systématiques chez les personnes vivant dans les régions à haute radioactivité naturelle. Une première étude de ce type, effectuée au Brésil par Barcinski *et al.* [6], a donné des résultats négatifs mais, dans des recherches ultérieures, ces mêmes auteurs [7] ont observé un accroissement du taux d'anomalies chromosomiques dans les populations de la région de Guarapari. Des résultats négatifs ont également été publiés par Georges *et al.* [8] pour les populations de la région côtière de Kerala. Selon Pohl-Rüling *et al.* [9], le taux d'anomalies chromosomiques serait néanmoins plus élevé dans les lymphocytes de personnes vivant dans la région de Badgastein et exposées à des doses de 1500 mrad par an; des observations similaires ont été faites par Kochupillai *et al.* [4] en Inde. Beaucoup plus convaincants, cependant, sont les résultats positifs de Takahasi [10] sur un scorpion (*Tityus bahiensis*) vivant dans l'état de Minas Gerais (Brésil) où les doses varient de 7 à 13 rad par an. Les chromosomes des scorpions n'ont pas de centromère localisé. Ceci permet la cassure des chromosomes sans perte de matériel chromosomique étant donné que chaque fragment a la propriété de se diriger correctement à l'anaphase. Cette caractéristique, ajoutée au fait que ces animaux vivent de 5 à 10 ans, est évidemment for intéressante pour étudier les effets cytogénétiques d'une exposition chronique. Chez les scorpions récoltés aux environs des points où la radioactivité atteignait 28 rad par an, Takahashi a observé une augmentation statistiquement significative du nombre de spermatocytes porteurs d'anomalies chromosomiques.

Dans la plupart des études que nous venons de résumer, il a été difficile, ou même impossible, d'estimer de façon exacte la dose de radiation reçue par les personnes ou les animaux étudiés. Si l'on y ajoute la difficulté, déjà évoquée, de trouver des témoins valables, il était pratiquement impossible de conclure si, oui ou non, une radioactivité naturelle élevée pouvait avoir des effets génétiques ou cytogénétiques sur les populations humaines ou animales vivant dans ces régions.

Dans plusieurs régions de la France (Figure 1), la radioactivité naturelle est localement élevée. Afin d'éviter les difficultés liées à l'étude des populations naturelles, des animaux, et des espèces végétales porteuses de marqueurs génétiques [11, 12] ont été importés. Leur élevage, ou leur culture, dans des conditions bien connues a permis, grâce à une dosimétrie rigoureuse de connaître les doses effectivement reçues et, par conséquent, d'établir des relations dose-effet beaucoup plus strictes.

2. LA RADIOACTIVITE NATURELLE DANS LE SUD-OUEST DE LA FRANCE

Nos observations ont été réalisées sur des animaux maintenus sur un site radioactif des environs de Lodève. A 50 km au nord de la Méditerranée, 45 km au Nord-Nord-Est de Béziers, 45 km à l'Ouest-Nord-Ouest de Montpellier, (fig. 2), le Bassin de Lodève est une région de transition entre les plaines du Languedoc héraultais au Sud et les plateaux des Causses calcaires (représentant à cette longitude, la marche la plus méridionale du Massif Central), au Nord. C'est un ensemble de collines de basse altitude (200 à 400 mètres) disposé de part et d'autre de la vallée de la Lergue, rivière principale de la région drainant du Nord au Sud, vers l'Hérault et la Méditerranée, les précipitations d'un climat méditerranéen dégradé par l'altitude et la continentalité : hiver relativement doux et pluvieux, période de sècheresse estivale de un mois et demi environ, pluviosité annuelle moyenne de 975 mm, température annuelle moyenne égale à 13°C.

Cette géomorphologie collinéenne est sculptée dans de puissantes séries détritiques permiennes dont la structure monoclinale basculée vers le Sud-Sud-Est conditionne par ses pendages de 15° la dissymétrie générale des versants (Nord abrupts, Sud à pente plus douce). Les conglomérats mais surtout les grès, caractérisés souvent par une granulométrie très fine (pélites et silts) sont les éléments lithologiques dominants de cette séquence sédimentaire affectée par trois systèmes de failles tardihercyniennes, pyrénéennes et alpines.

Ces accidents structuraux ont joué le rôle de pièges pour l'uranium surtout dans l'Autunien moyen (de couleur grise) et dans les niveaux gris de l'Autunien supérieur (dominé par les faciès rouges). L'histoire sédimentologique de ce radionuclide naturel comprend quatre étapes principales:

- lessivage des bords du Bassin et venues volcaniques contribuant à la mise en place de sédiments à faibles teneurs;
- mobilisation de l'uranium par les tectoniques hercynienne et pyrénéo-alpine (le broyage des sédiments favorise cette mobilisation;
- transport par les eaux et les hydrocarbures à travers les terrains;
- accumulation dans les failles et les fissures avec d'autres éléments associés comme le zinc, le cuivre ou le plomb.

Les minéralisations sont de deux types :

- stratiforme et dans ce cas sans expression minéralogique, l'uranium étant lié aux bitumes, aux hydrocarbures ou à la matière orga-

nique. Les teneurs sont faibles ou très faibles : de l'ordre de 0.1°/₀₀, elles dépassent rarement 1°/₀₀;

- fissurales : liées aux failles de direction Est-Nord-Est, elles sont très riches et exploitables.

Par le jeu de l'érosion, certaines de ces minéralisations sont actuellement en surface. Deux d'entre elles (fig. 2) constituent des indices de première importance compte-tenu des activités radioactives qu'elles permettent d'y déceler :

- la première est dans la petite vallée qui, affluente de la vallée de la Lergue, conduit au hameau du Mas d'Alary;

- la seconde est au Sud du village de Saint-Martin du Bosc, non loin et au Sud de la route départemantale 144 E, au lieu-dit Riviéral.

C'est à ce dernier endroit que l'expérience de radiobiologie dont les résultats sont rapportés ci-dessous a été réalisée. La minéralisation uranifère fixée sur l'avant dernier niveau fertile gris de l'Autunien supérieur rouge, au voisinage d'une fissure, correspond à des accumulations de carburanes et d'un peu de pechblende accompagnés en surface par des produits résultant de leur altération comme l'uranotile et les arséniates d'uranium. Dans la partie la plus riche de l'indice, les teneurs sont exceptionnellement élevées puisqu'elles sont de l'ordre de 1% certains échantillons atteignant les valeurs de 7 à 8%.

La présence de ces fortes concentrations en uranium est à l'origine de la très forte radioactivité naturelle caractérisant ces indices de surface. Les rayonnements ionisants émanent non seulement des deux isotopes naturels de l'uranium (99.3% d'U 238, 0.7% d'U 235) mais aussi de leurs 28 descendants radioactifs parmi lesquels certains, caractérisés par une période courte ou très courte, contribuent d'une manière active à l'irradiation des êtres vivants évoluant en cas lieux.

L'uranium et le radium 226 des roches, des sols et des végétaux ont été dosés respectivement par fluorimétrie et spectrométrie gamma. Parallèlement, un effort important a été consacré à la dosimétrie afin d'évaluer avec le plus de précision possible les débits de dose caractérisant ces stations exceptionnellement radioactives. Fournis par le Laboratoire de Dosimétrie Physique du Centre d'Etudes Nucléaires de Fontenay aux Roses (C.E.A.), des dosimètres radiothermoluminescents au $CaSO_4$ ont été placés au-dessus des deux. Au Mas d'Alary, un débit de 10 mrad/h a été trouvé sur le point le plus radioactif de l'indice. A Riviéral, des valeurs comprises entre 2,7 et 14,7 mrad/h ont été mesurées dans la zone la plus riche en uranium [11].

3. OBSERVATIONS REALISEES SUR LES ANIMAUX

a. Matériel et Méthodes

Un abri a été construit dans un endroit où le débit de dose atteint 8 mrad/h. Sur le sol, nous avons placé des cages contenant chacune un lapin muni d'un dosimètre individuel au fluorure de lithium. Pour éviter toute interférence avec une contamination interne résultant de l'ingestion de nourriture radioactive, ces animaux ont reçu une nourriture commerciale. Tous les quatre mois, un échantillon de sang a été prélevé sur chaque animal et, après une culture de 48 heures, les chromosomes des lymphocytes ont été examinés afin de déceler la présence éventuelle d'anomalies induites. La teneur de l'air en radon (^{222}Rn) a été déterminée au printemps et en automne : elle atteignait en moyenne 0,27 WL (Working Level). Le "Working Level Unit" est une unité de concentration se définissant comme toute combinaison dans un litre d'air des descendants à vie courte du ^{222}Rn, telle que l'énergie alpha totale pour aboutir à la décroissance jusqu'au ^{210}Pb soit de $1,3 \times 10^5$ MeV. Cette valeur correspond à l'énergie alpha libérée par la décroissance des descendants à vie courte en équilibre avec 100 pCi de ^{222}Rn. Le "Working Level Month" (WLM) est une unité d'exposition résultant de l'exposition à WL pendant un mois. Pour etudier la part éventuellement prise par le radon dans les effets observés, nous avons fait inhaler, en laboratoire, à des lapins une dose cumulée comparable à celle qu'ils ont reçue pendant un an à Lodève soit 0,27 WL x 4 x 12 = 12 WLM.

Pour étudier les effets de la radioactivité naturelle sur la fertilité des mammifères et pour détecter la production éventuelle de dommages génétiques dans les cellules reproductrices, nous avons placé des souris mâles et femelles dans le même abri. Etant donné la sensibilité de ces animaux aux conditions climatiques, nous n'avons laissé les souris à Lodève que durant les mois d'été. Par après, elles ont été croisées, durant 6 mois, en laboratoire avec des souris témoins de même race.

Des lapins et des souris de même race ont été maintenus dans les mêmes conditions à proximité du site radioactif afin de servir de témoins.

b. Résultats [13, 14]

La dose de rayons γ reçue par les lapins durant les 28 premiers mois de cette étude variait selon les animaux de 43 à 163 rad alors que, pour les témoins, elle ne dépassait pas 0,3 rad (Tableau I). La fréquence des

anomalies chromosomiques instables a augmenté jusqu'au 8ème mois mais s'est révélée très variable par la suite (Tableau II). Il faut noter que, chez les lapins ayant séjourné plus d'un an sur le site radioactif, la réponse des lymphocytes au mitogène utilisé (phytohémagglutinine) a fortement décru, ainsi que le montre le petit nombre de mitoses analysables que nous avons trouvé dans les cultures faites à ce moment. Nous n'avons observé aucun accroissement significatif des anomalies chromosomiques dans les lymphocytes des lapins ayant inhalé du ^{222}Rn en laboratoire. Ces résultats démontrent qu'une radioactivité naturelle élevée peut entraîner l'apparition d'anomalies chromosomiques dans les cellules somatiques des mammifères. Ils démontrent également que le radon ne semble pas contribuer à la production de ces anomalies, un effet synergique radon-rayons γ, quoique fort peu probable, étant cependant toujours possible. Ceci concorde d'ailleurs avec les observations de Kilibarda *et al.* [15] sur des travailleurs yougoslaves de mines d'uranium exposés à des concentrations de $3,7 \times 10^{-10}$ à $2,61 \times 10^{-9}$ Ci/litre et avec celles de Brandom *et al.* [16, 17] sur des mineurs exposés à moins de 100 WLM. Pour expliquer la grande variabilité dans les taux d'anomalies observées chez les lapins, nous avons procédé, en laboratoire, à diverses expériences qui nous ont permis de démontrer que, dans cette espèce, la longueur moyenne de vie des lymphocytes porteurs d'anomalies chromosomiques est voisine de 50 jours seulement alors qu'elle est de plus de 1000 jours pour les lymphocytes humains [18]. Il est probable, dès lors, que le manque de relation entre la dose de rayons gamma reçue et le taux d'anomalies observé s'explique en bonne partie par la durée de vie réduite des lymphocytes de cette espèce qui ne représente donc pas un modèle expérimental idéal pour ce genre d'étude. Des résultats récents de Evans *et al.* [19] sur des travailleurs occupés au remplissage des réacteurs et ayant reçu par an, des doses inférieures aux 5 rem autorisés démontrent que, chez l'homme, on peut observer une relation entre le taux d'anomalies et la dose cumulée reçue.

Une première étude faite en 1976 suggérait que la fertilité des souris mâles ayant séjourné trois mois sur le site radioactif était légèrement plus élevée que celle des mâles témoins maintenus à proximité. En 1977, nous avons répété cette expérience mais en l'étendant cette fois également aux femelles. Ces animaux étaient âgés de 4 mois au moment ou l'exposition a débuté et avaient 8 mois lorsque nous les avons

repris pour les croiser. Ainsi que le montre le tableau III les mâles irradiés, qui ont reçu une dose plus élevée (15 rad) que dans la première expérience (9 rad), ont produit plus de nuchées que les témoins alors que l'inverse s'observe pour les femelles. Les différences entre animaux témoins et animaux exposés ne sont cependant significatives que pour les jeunes produits ($P < 0,05$) et les jeunes sevrés ($P < 0,01$). Si on compare mâles et femelles, on se rend compte que, pour ces dernières, c'est surtout l'âge qui a joué, des difficultés lors de la mise bas ayant entrainé la mort d'un nombre élevé de jeunes. Les oocytes jeunes de la souris sont extrêmement radiosensibles, leur DL50 ne dépassant pas 50 R [50]. nos résultats démontrent que des débits de dose extrêmement faibles entraînent la destruction d'une bonne partie de ces oocytes jeunes. Nos observations sur les souris mâles sont, par contre, beaucoup plus inattendues mais concordent cependant avec des résultats publiés récemment par Newcombe et Mc Gregor [21, 22] : ces auteurs signalent, en effet, que des doses aussi faibles que 25 à 50 R administrées aux spermatozoïdes de la truite arc-en-ciel augmentent de façon statistiquement significative le pourcentage d'embryons viables.

CONCLUSIONS.

1. Nos résultats démontrent que des débits de dose anormalement élevés (8 mrad/heure) peuvent produire des anomalies chromosomiques dans les cellules somatiques des mammifères. Ceci concorde avec les résultats positifs obtenus chez les végétaux par Delpoux *et al.* [12].
2. De tels débits sont également suffisants pour diminuer significativement la fertilité des souris femelles alors qu'ils accroissent le nombre de jeunes vivants produits par les mâles.
3. Les effets observés sont essentiellement dus aux rayonnements gamma et ne sont, probablement, pas provoqués par le radon inhalé.
4. Si l'on tient compte du fait que les affleurements à radioactivité aussi élevée ne mesurent jamais plus de quelques mètres carrés et que les animaux mis en expérience se trouvaient dans des cages placées à même le sol la probabilité d'observer des effets de ce genre dans les populations humaines vivant dans ces régions est pratiquement nulle. Pour les populations humaines vivant sur des sites radioactifs plus étendus il reste théoriquement le danger de contamination via la chaine alimentaire entrainant des transferts à partir des sites radioactifs et produisant des irradiations internes. Compte tenu de la radioactivité naturelle faible généralement rencontrée en France on peut également considérer cette possibilité comme inexistante pour ce pays.

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TABLEAU I - Doses (mrad) de rayons gamma reçues par les lapins maintenus à Lodève

Lapin N°	4 mois	8 mois	12 mois	16 mois	20 mois	28 mois
1	26,500	52,750	70,750	101,750	130,750	163,250
2	22,300	44,300	57,300	76,800	95,800	124,300
3	22,500	43,250	56,000	73,500	91,500	126,000
4	13,000	26,500	38,700	51,200	*	13,800
5	7,800	14,800	20,500	28,500	36,300	43,500
6	65	102	152	207	254	314
7	40	95	140	195	240	330
8	50	110	158	205	235	315

*Le lapin 4 est mort après 19 mois.

TABLEAU II - Observations cytologiques réalisées sur les lapins maintenus à Lodève.

Lapin N°	0 Jour		4 Mois		8 Mois		12 Mois		16 Mois		20 Mois		28 Mois	
	Cellules analysées	Anomalies	Cellules analysées	Anomalies	Cellules analysées	Anomalies	Cellules analysées	Anomalies	Cellules analysées	Anomalies	Cellules analysées	Anomalies	Cellules analysées	Anomalies
1	200	0	400	4 fragments	200	4 dicentriques	200	4 fragments 2 dicentriques	150	3 fragments	100	0	50	1 fragment 1 dicentrique
2	200	2 fragments	300	2 fragments	100	2 dicentriques	200	4 fragments	100	2 fragments	100	0	250	1 fragment 3 dicentriques
3	200	1 fragment	-		200	4 fragments 1 dicentrique	200	3 fragments 1 dicentrique	200	2 fragments	100	0	200	4 fragments 1 translocation
4	200	0	350	4 fragments 1 dicentrique	200	1 dicentrique	200	1 fragment 1 dicentrique	150	1 fragment	*		100	0
5	200	0	-		100	1 fragment	200		100	1 dicentrique	100	0	100	1 anneau
6	200	0	400	0	400	1 fragment	200	0	150	0	200	1 fragment	250	1 fragment
7	200	0	400	1 fragment	400	0	200	0	200	0	200	0	200	0
8	200	0	400	0	400	0	200	0	200	0	200	0	200	0

*Le lapin 4 est mort après 19 mois

TABLEAU III - Fertilité des souris témoins et des souris exposées.

Sexe	Traitement		Nichées produites		Jeunes produits		Jeunes sevrés	
	Groupe	Dose reçue (mrad)	Total	Moyenne	Total	Moyenne	Total	Moyenne
Mâles	Témoins	68	63	3,15	365	18,25	281	14,05
	Exposés	15.000	71	3,55	448	22,40	388	19,40
Femelles	Témoins	68	40	2,00	155	7,75	78	3,90
	Exposés	15.000	28	1,40	102	5,01	58	2,90

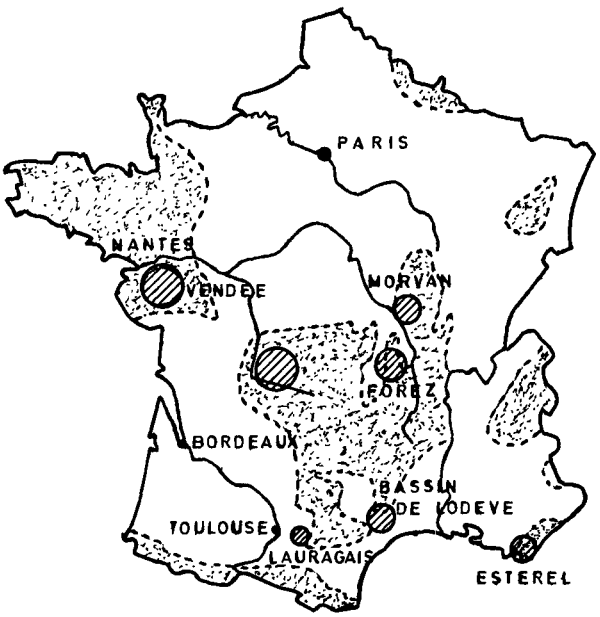

Fig. 1. Régions de France présentant localement une radioactivité élevée

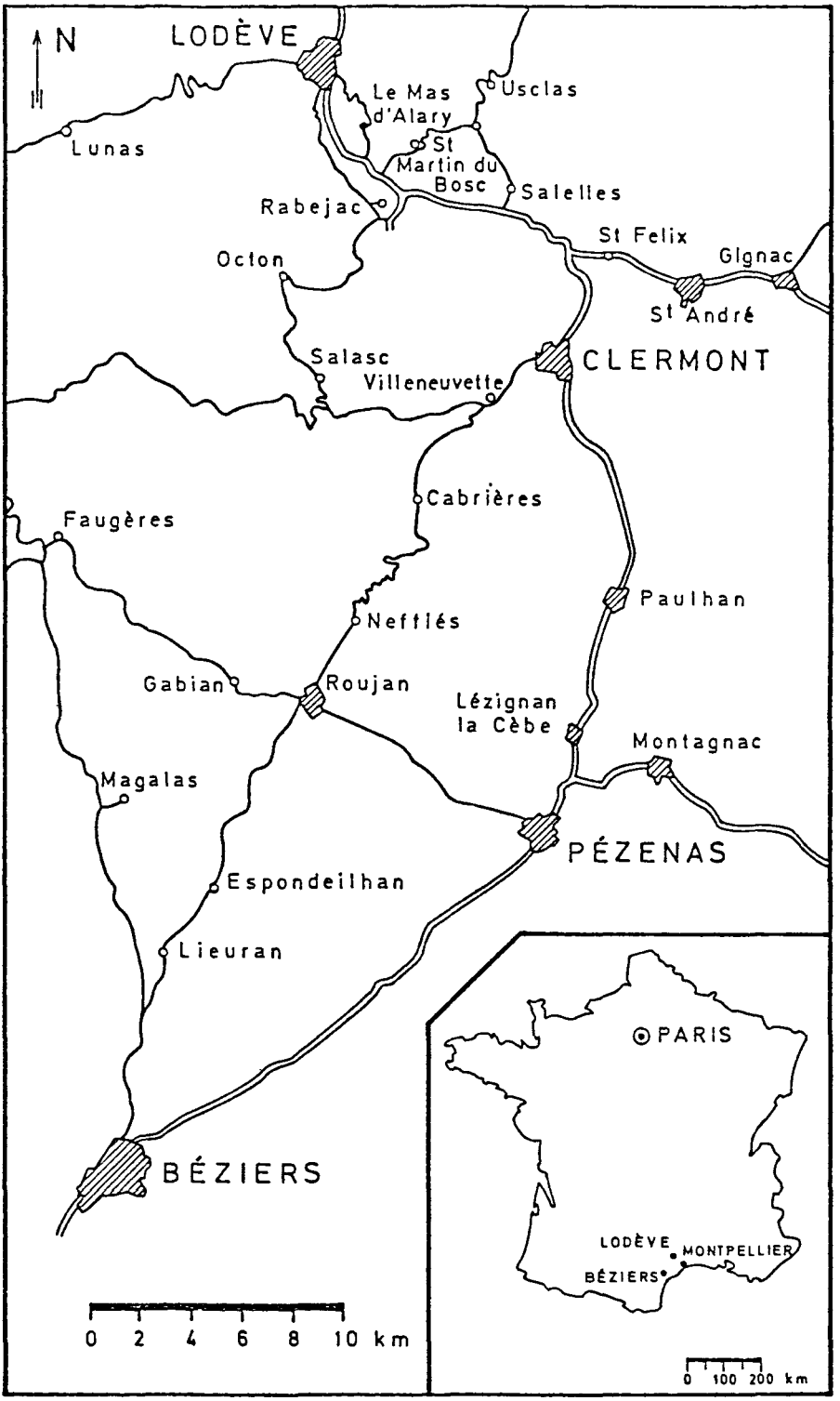

Fig. 2. Situation géographique de la région de Lodève.

PANEL SESSION

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CONCLUSIONS OF THE SEMINAR - CONCLUSIONS DU SEMINAIRE
ERGEBNISSE DES SEMINARS

Summaries of sessions, presented by the Chairmen
Résumés des sessions, présentés par les Présidents respectifs
Zusammenfassende Referate der einzelnen Sitzungspräsidenten

Introductory address by Sir Edward POCHIN:

I am very much aware that there have been good discussions during most of our sessions, and that the discussions stopped really because of the end of the time rather than because of the end of the questions. But I think that we have had an impressive display of information and that we should think about what we get from it. I believe that we get two things from it: firstly we have had valuable perspectives of the relative importance of different types of background or near-background radiation exposure; we have had ideas on actions that we might propose to ourselves or to the Commission, on things that could usefully be done.

We have been discussing essentially the physics of measurements and we cannot go far, either now or in the discussion, into the biological risk assessments because this is a large separate subject-I say "separate", although I believe that it is important to have a good interface between the measurements of the amount of radiation and the size of populations exposed, and the biological implications of these exposures. But as to perspectives, I think we have obtained useful ideas on how large are the variations in normal radiation exposure that groups of people have always experienced for thousands of years. We can see, as we knew before, of course, that these variations from one part of a country to another are large relative to any additions due to human actions, and very large relative to non-medical additions of any type that have been made.

We have seen background quite commonly being twice the ordinary average value world-wide, less commonly three times that, and only quite rarely and very locally, in small areas, five or ten or more times the world average. We have had a perspective on the source of the largest variations in natural radiation. We can assume that the internal radiation component

from potassium-40 is constant except for the small changes according to sex and age which do not affect dose within the cell mass but which are due simply to variations in the fat content of the body, as has repeatedly been shown. So we have a fairly constant internal radiation level for most tissues and a fairly constant cosmic radiation level for most of the population within the European area that we are considering particularly. We have seen the variations in the terrestrial radiation level for whole body exposure, and we have seen the size and the importance of natural variations due to radionuclide content of water or of food or of the air—I think it has emerged clearly that the source that is of greatest magnitude in causing these variations appears to be that associated with the condition of housing; and particularly in small houses near the ground floor, and presumably in cold countries where you don't have windows wide open during the winter and fast air changes.

We have also, I think, several bases for actions. One importance of obtaining measurements of radiation levels in different positions throughout a country is as a base-line with which to compare radiation levels subsequently, if there were believed to have been a contamination of a particular area owing to radioactive releases. I rather suspect that the measurements locally round a reactor should be made in advance by the management of that reactor, rather than purely as a Community project. When, therefore, we are looking at the Europe-wide measurements, we are needing only a widely based general survey to exclude changes attributed to releases when in fact those variations had always been present. The local problem of discharges into water is somewhat different, since rivers lead to larger rivers and this may become an international problem. So there may not be so much an action in extending in very much detail the baseline of measurements on land, but there may be importance in measurements in waterways.

It is very clear also from several of the papers that there is need for careful study of food chains and distribution pathways; and the Community has recently published an important review on the types of model to be used in estimating population doses or local doses in relation to such pathways. This is a publication jointly by the NRPB and CEA, undertaken

on behalf of the Commission*). This is a study of a general procedure and I believe that some of the papers during the last three days provide valuable numerical data for particular areas that can be fed into these models to study the distribution of radioactivity derived from local sources.

Finally, may I return to a point with which I started in my comments three days ago regarding the chances of effective epidemiological studies. Here I feel that authors of papers should wish to look at their dose levels in relation to the size of populations over which those doses are distributed, because it is essentially the product of population size and elevation of dose, or more exactly of population size and the square of elevation of dose, which gives some criterion as to the prospects from population epidemiology. A study of the chances of useful epidemiology in populations exposed, more or less "whole-body", to external radiation from background is an important one to make, but I haven't seen the populations of several millions exposed to several times natural total background which might give results which could be statistically reliable, for cancer incidence or mortality as a whole, within limited periods of five or ten years. I think that we individually as authors of papers, and the Community collectively, should wish to look carefully also at the possibilities in different areas of ascertaining the frequency of disease. I believe that this is important and I would return again to the importance that we attach in my own country to the value of being able to study the causes of death recorded on death certificates - not for people identified individually by name but for groups of people identified to a central office by name, of which the distribution of causes of death may be returned to the investigator without names associated with each individual death. This I believe is a subject which causes difficulty in some countries; but the solution of it, nationally, might do a great deal to help in getting a better prospect of valid estimates of the harm from radiation at low doses, and you know as well as I do how many non-valid estimates of the harm of radiation at low doses are being put out on very poor statistical grounds.

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*) "Methodology for evaluating the radiological consequences of radioactive efluents released in normal operations" C.E.C., July 1979.

It is very important that good statistical studies should be made, both on general populations, if they are large enough and exposed at high enough dose, and in addition on exposed workers. And finally, as a biological point, I believe it is important that we should look for different types of harm that may be caused by radiation - cancer death, cancer occurrence, occurrence of particular types of cancer, occurrence of particular types of cancer at particular ages in either sex - to see how far we can focus down on an index which is going to be much more sensitive as a measure of low dose effects than the difficult one of studying total cancer incidence in the whole population. So I believe that there are a number of points which have been emphasized in our discussions which may call for useful action by individual scientists, individual countries, or the Community. I hope that we may be able to review points arising from the different Sessions, and in the different context of papers in these Sessions, and be able to make a list of possible useful agenda arising from this extremely valuable meeting.

J.P. MORONI

La première session a été sans doute celle qui a posé le moins de problèmes scientifiques car la mesure de l'exposition au rayonnement naturel à l'extérieur semble actuellement sinon résolue tout au moins en bonne voie. Dans cette session nous avons eu des contributions concernant un certain nombre de pays de la Communauté, l'Allemagne, les Pays-Bas, l'Irlande, le Denmark, le Groenland où, nous l'avons vu, se trouvent des zones d'assez haut niveau, la France pour laquelle les cartes de l'exposition sont en cours d'achèvement, sans oublier les contributions qui nous ont été données pour les pays invités par Mme SWEDJEMARK, MM. PORETTI et TSCHIRF respectivement sur la Suède, la Suisse et l'Autriche. Du point de vue général de l'obtention d'une carte de l'irradiation de l'Europe, préalable indispensable pour pouvoir aborder les étapes suivantes,on peut dire que les difficultés ont été surmontées. Les difficultés étaient d'abord d'ordre technique concernant les appareillages. Il fallait trouver un ou plusieurs appareils, une ou plusieurs méthodes qui soient indépendants de l'énergie, et suffisamment fiables. Je ne m'étendrai pas ici sur cette recherche; je crois que cela est actuellement pratiquement résolu. D'ailleurs j'ai pu remarquer, comme vous avez dû le faire également, que dans les discussions il y a eu beacoup moins d'interventions que nous en avions pour les questions d'étalonnage des appareils et, d'une manière générale, sur la technique.

Un autre piège classique est celui des erreurs systématiques qui peuvent apparaître dans l'élaboration de ces cartes. Là aussi je crois que nous sommes en bonne voie. Nous avons maintenant de longue date reconnu qu'il ne fallait pas faire des mesures uniquement le long des routes, qu'il fallait se garder de la contribution du radon à l'extérieur les jours où la météorologie est défavorable à cause d'inversions, etc.: il s'agit là de faits désormais bien connus.

Une question cependant demeure, qui n'a pas encore reçu de solution définitive: la définition des zones de mesures, des mailles du réseau de mesures effectuées sur le terrain. Là il y a encore un certain nombre de perfectionnements à apporter. Une fois ces difficultés levées il reste, je ne dirai pas à faire les mesures, car elles sont faites dans de nombreux pays et en cours de l'être dans de

nombreux autres, mais il reste à compléter cette tâche. Je crois que là aussi cela est en bonne voie et nous espérons sortir une carte de l'exposition dans les pays de la Communauté avant la fin de 1980. Je renouvelle à ce sujet les appels que j'avais déjà faits pour que les personnes ici présentes ou leurs collaborateurs nous transmettent toutes données qu'ils auraient sur ce point.

Par ailleurs, il est certain, comme de nombreux participants l'ont rappelé au cours de cette première séance, qu'il convient de considérer le but que nous devons atteindre sous un angle réaliste et demeurer modeste dans les prétentions que nous pouvons avoir quant à la signification des cartes.

Ainsi que vous l'avez dit, Monsieur le Président, nous savons d'ores et déjà quelle est l'amplitude des variations que nous pouvons attendre. Nous savons également, cela a été confirmé par de nombreux intervenants lors de cette première session, qu'il y a une étroite relation entre la géologie et le champ de rayonnement. Par conséquent, à grande échelle les résultats sont déjà prévisibles. Par contre, si l'on essaie d'effectuer des mesures dans le détail, là aussi tout le monde est d'accord pour dire qu'on trouve des variations considérables à l'échelle locale, à l'échelle de quelques mètres même; il s'agit là de détails qui de toute façon n'ont aucune signification en ce qui concerne l'exposition de populations. Donc j'insiste sur cette nécessité de considérer ces résultats avec un point de vue réaliste, ce qui n'empêche pas d'ailleurs que des études soient faites sur des points de détail, mais avec une optique tout autre, comme c'est le cas dans la très intéressante communication qui a été présentée par M. BONKA sur la haute activité trouvée dans certaines zones de sable.

J.P. MORONI

The first session was certainly the one which raised fewest scientific problems, since measurement of external exposure to natural radiation seems to be, if not solved, at least well on the way to a solution. During this session, we had contributions concerning the Community which related to Germany, The Netherlands, Ireland and Denmark, to Greenland where, as we have seen, there are areas with a relatively high radiation level and to France for which exposure maps are currently being prepared. From the invited countries we had the contributions of Mrs SWEDJEMARK, Mr. PORETTI and Mr. TSCHIRF regarding Sweden, Switzerland and Austria respectively.

The difficulties relating to the production of an exposure map of Europe, a prerequisite for further studies, may be said to have been overcome. Initially there were technical problems concerning the equipment. One or more sufficiently reliable, energy-independent instruments and procedures had to be found. I will not dwell further on this work; I believe it has been largely accomplished. Moreover, it was noticeable to me, and no doubt to the other participants, that during the discussions there were far fewer contributions than in the past on the calibration of instruments and procedures in general.

Another classic trap lies in the systematic errors which may occur during the preparation of such maps and , here again, I believe we are working along the right lines. We have long since realized that measurements must not be made only along roads and that one must beware of the contribution from radon outdoors when weather conditions are unfavourable because of temperature inversions etc.. These traps are now well known.

A final solution does not yet appear to have been found to the problem of defining the measurement zones, i.e. the measurement grid. A number of final adjustments still remain to be introduced. Once these difficulties have been removed, the main task will not actually be to take the measurements, since this has already been done in many countries and is in hand in many others, but to complete the work overall. This is now well under way and we hope to produce a map of radiation exposure in Community countries by the end of 1980. I once again ask all those

present and their associates to send us all their available data on this subject.

Nevertheless, we must, as was several times pointed out during this first session, consider our objective from a realistic point of view and not exaggerate the significance of the maps.

Thus, as you have remarked, Mr Chairman, we now know the degree of variation which can be expected. We also know, as confirmed by a number of comments during the first session, that there is a close link between the geology and the radiation field. In broad terms, therefore, the results are already predictable. On the other hand, everyone agrees that, if one tries to take detailed measurements, substantial local variations are found even over distances of a few metres, but in any case this factor is of no importance to population exposure. I therefore stress the need to view the results realistically, but though this should not prevent very localized studies in the kind of detail presented, but with a quite different perspective, in the very interesting paper contributed by Mr BONKA on the high activity to be found in certain sand zones.

M.C. O'RIORDAN

The objectives set for the Seminar were "to amass existing information and determine priorities for further studies". Did the session on indorr-exposure achieve these objectives? I believe it did, despite the efforts of the Chairman.

My colleagues undoubtedly added to our store of information and promised us more data in the near future. Perhaps more importantly, they showed us how they were analyzing that data. By this I do not mean mere statistical analyses but rather how they were assessing the significance of the data in social terms.

Our social purpose is to protect individuals and the community from unnecessary radiation exposure. It seems to me immaterial whether the source of exposure is natural or unnatural - a rad is a rad, regardless of origin. The only difference is that we have come to grips with the one and shy away from the other.

What does ICRP advise? Let me paraphrase: Normal exposure is not subject to control, the Commission recommends, but increases are. It notes that increases can occur in some environments and because of some practices. Then it asserts that existing variations in exposure are not grounds for increases.

It seems to me that the central idea here is that of normality and that it must have two connotations a mathematical one and a social one. My colleagues in Session II also seemed to recognize this, either consciously or unconsciously. Almost invariably they described a distribution of radiation exposures or of related quantities and then directed our attention to the extremes of the distributions and in particular to the higher regions. None of them questioned the normality of living indoors but in concentrating on the upper values they betrayed some concern about abnormality.

In one case, as we heard, this concern is so acute that remedial action is being taken. I suggest however, that there is nothing unique in the Stockholm situation: it is merely a matter of degree. But having seen the difficulties that can be created through the inadvertent use of certain materials, not only in Sweden but elsewhere, we would be blame-worthy indeed if we did not assess any significantly abnormal indoor exposure, present or potential.

It follows therefore that futher studies might well be directed to establishing the distributions of indoor exposures in an adequate manner. This has not been achieved throughout the Community, particularly for radon daughters. By carefully considering the factors, such as geology and building methods, that affect exposure, it should be possible to generate sufficient data without excessive effort.

I shall not burden you again with my views on modelling but I believe that it has a degree of usefulness in the field of natural radiation just as it has in the field of artifical radiation.

Finally I want to turn to metrology. In most areas of radiation protection it is possible to be assured of the quality of the measurements by tracing them to a national laboratory which has in turn engaged in international intercomparisons. Alternatively it is possible to participate in direct intercomparison exercises organized by the IAEA and latterly by the ICRM, the International Committee on Radionuclide Metrology. Neither of these agencies have, to the best of my knowledge, sponsored an intercomparison of radon daughter measurements. I am not persuaded however that there is a pressing need for organized Community action here because the experts making these measurements seem to be in harmony already.

W. JACOBI

Um die Diskussion etwas anzuregen, möchte ich auch mit ein paar kritischen Bemerkungen beginnen. Zunächst einmal, wenn ich auf das Titelblatt unseres Programms schaue, so steht da "Strahlenbelastung der Bevölkerung" oder im Englischen "Burden", im Französischen "Charge". Nun, dieses Wort hat für mich nur einen negativen Aspekt - eine Belastung. In Wirklichkeit ist doch ein Teil dieser natürlichen Belastung, nämlich das was wir als normal bezeichnen, ein natürlicher Vorgang, unter dem sich auch unser menschliches Leben - vielleicht im negativen aber auch im positiven Sinne entwickelt haben kann. Ich finde es daher erstens einmal dringend notwendig, dass wir klar unterscheiden, was ist normale natürliche Strahlenexposition und was ist eine modifizierte, durch Zivilisation, durch Technologie bedingte Veränderung dieser Exposition?

Wenn wir diese Trennung beachten, ergeben sich auch sofort zwei Zielsetzungen. Die normale Exposition und ihre Schwankungsbreite ist meines Erachtens ein hervorragender Massstab zur Beurteilung der zivilisatorischen Strahlenexposition der Bevölkerung, insbesondere auch durch die Emissionen kerntechnischer Anlagen. Um die Frage der Akzeptanz dieser zusätzlichen Exposition beantworten zu können ist es notwendig, die Verteilungsfunktion dieser normalen natürlichen Strahlenexposition zu kennen. Wir haben im Laufe dieses Symposiums erfahren, dass wir für einige Beiträge schon eine ganz gute Abschätzung machen können. Ich denke dabei an die terrestrische Strahlung und die kosmische Strahlung. Aber wir sollten auf diesem Weg weiterfahren und auch für die anderen normalen natürlichen Strahlenquellen eine solche Verteilungsfunktion aufstellen, um die Strahlenabweichung dieser Verteilung als Kenngrösse für die Festlegung von Grenzwerten durch zusätzliche Belastung heranziehen zu können.

Bei dem anderen Teil der natürlichen Strahlenexposition, die wir als zivilisationsbedingte oder technisch bedingte Veränderung der normalen Exposition betrachten, kommt es darauf an zu erkennen, ob es Einflüsse gibt, die dazu geführt haben, dass Einzelpersonen oder einzelne Gruppen von Personen einer sehr hohen zivilisationsbedingten natürlichen Exposition ausgesetzt sind, so dass das damit möglicherweise verknüpfte Strahlenschadenrisiko nicht mehr akzeptierbar erscheint. In diesem Bereich kommt es daher darauf an, nicht nur die Verteilungsfunktion der Personenexposition zu erfassen, sondern gerade ihre Spitzenwerte festzustellen und Massnahmen zu ihrer Reduktion vorzuschlagen.

Ich möchte nun auf die speziellen Punkte aus unserer Sitzung zu sprechen kommen.

Wir haben gesehen, dass im Falle der Ingestion natürlicher Radionuklide zweifelsohne das K-40 die entscheidende Rolle spielt. Über das K-40 im Körper wissen wir sehr gut Bescheid und hier gibt es sehr wenig Probleme. Wir haben ferner aus den verschiedenen Ländern eine grosse Zahl von Daten über die Aktivität von Uran, Thorium, Radium, Blei und Pollunium in der Nahrung und im Trinkwasser vorgelegt bekommen. Diese Daten haben gezeigt, dass diese Aktivitäten ganz erheblich schwanken können aber auf der anderen Seite die resultierende Aktivität und Dosis im Körper relativ klein ist. Der Grund dafür ist klar: Wir haben zwar sehr grosse lokale Schwankungen der Aktivität im Boden und im Wasser, aber unsere Nahrungsmittel stammen aus einem sehr viel weiträumigeren Gebiet; wir sind im Essen sehr viel europäischer als im Denken und in der Politik. Wir müssen daher den gesamten Verteilungsweg der natürlichen Radionuklide in der Nahrung vom Erzeugungsort bis zum Verbraucher erfassen. Das heisst, wir sollten die vorliegenden Einzelmessungen, die gewissermassen Mosaiksteine bilden, zusammensetzen, um Aufschlüsse über die Transferwege und Transferfaktoren zum Menschen zu erhalten. Wir müssen auch hier daran denken, dass uns die natürlichen Radionuklide wichtige Anhaltspunkte über den Aktivitätstransfer zum Menschen liefern, die wir dann für künstliche Radionuklide heranziehen können. Ich möchte z.B. erinnern an die Analogien des Verhaltens von Thorium zu Plutonium und von Radium zu Strontium.

Am Ende dieser Transferkette steht der Mensch. Auch hier können uns die Erfahrungen über die Verteilung dieser natürlichen radioaktiven Stoffe im Menschen Aufschluss geben über den Stoffwechsel dieser Elemente und über ihre Aufnahme und biologische Halbwertszeiten in Organen, die wir dann im Strahlenschutz bei anderen ähnlichen Radionukliden sehr wichtig gebrauchen können.

Die Daten, die in dieser Sitzung gebracht wurden, zeigen die Basis auf, auf der wir weiterarbeiten sollen. Wir müssen dabei aber immer klar das Ziel vor Augen sehen: Erstens die Erfassung der Verteilungsfunktion der normalen Exposition als Massstab für die Festlegung von Grenzwerten, zweitens die Feststellung ihrer Veränderung durch den Menschen und seine Zivilisation mit dem Ziel, Extremwerte rechtzeitig zu erkennen und entsprechend den Grundsätzen des Strahlenschutzes zu reduzieren.

W. JACOBI

I should also like to begin by making a few critical remarks which I hope will simulate discussion. Firstly, looking at the title page of our programme I see the words "Radiation Burden of Man" - in German "Belastung" and in French "Charge". For me, the term "burden" has only negative connotations. In reality, however, part of this natural "burden", that is to say the part which we regard as normal, is a natural process in which human life has developed, in a negative sense perhaps but also in a positive sense. I therefore regard it as absolutely essential that we first draw a clear distinction between what is normal natural radiation exposure and what are the changes in exposure which have resulted from civilization and technological development.

If we respect this distinction, two objectives immediately emerge. In my view, normal exposure and fluctuations therein are an excellent yardstick for assessing the radiation burden on the population as a result of civilization, and in particular emissions from nuclear facilities. If we are to judge the acceptability of additional exposure, we need to know the distribution function of the everyday natural radiation. In the course of this symposium we have seen that a fairly good estimate can be made for contributions from certain sources; I am thinking here of terrestrial radiation and cosmic radiation. However, we must go further in this direction and determine a distribution function for the other natural sources of radiation so that the standard deviation of this distribution can provide a yardstick for maximum values for additional exposure.

As regards the second aspect of natural radiation exposure, that is to say changes in the normal exposure levels as caused by civilization and technology, we must determine whether there are any circumstances which have resulted in specific individuals or groups being subjected to such a high level of civilization-induced natural radiation that the risk of harm is no longer acceptable. In such a case, it is therefore not only a question of determining the distribution function of the exposure of members of the population but in particular of establishing the peak values and proposing ways of reducing them.

I should now like to cover certain specific points which arose during this session.

We have seen that in the case of ingestion of natural radionuclides it is K-40 which undoubtedly plays the decisive role. We have a very good knowledge of K-40 in the body and there are few problems in this area. In addition, we have received from the various countries a large amount of data on the uranium, thorium, radium, lead and pollonium levels in foodstuffs and drinking water. These data have shown that while there may be considerable variations in such levels the resulting activity and dose in the body remains relatively small. The reason for this is evident: local variations in activity in soil and water are indeed very wide, but our foodstuffs come from a much more extensive area; we are much more European in our eating habits than in our attitudes and politics. We therefore have to establish the exact distribution route of natural radionuclides in foodstuffs from the place of production to the user. This means that the available individual measurements, like pieces of a mosaic, must be put together to give a complete picture of the transfer pathways to man and the transfer factors. It should be remembered here that natural radionuclides provide us with important clues to the transfer of activity to man, which can in turn be applied to artifical radionuclides. I am referring, for example, to the analogies between the behaviour of thorium and plutonium or radium and strontium.

At the end of this chain is man. Here too, a knowledge of the distribution of natural radioactive materials in man can throw light on the metabolism of these elements, their assimilation and biological half-lives in organs, information which will be useful to us in radiological protection for other, similar, radionuclides.

The data contributed at this session has provided a basis for our future work. In this, we must always have the following objectives, clearly in mind: the first is to establish the distribution function of normal expsure as a yardstick for fixing maximum permissible values; and the second is to determine the changes brought about by man and his civilization with the aim of identifying in due course the peak exposure values and reducing them in accordance with the principles of radiological protection.

R. KIRCHMANN

Cette 4e session concernait, comme vous le savez, la modification de l'exposition due à la technologie, déjà évoquée par le Dr. JACOBI. Il y avait deux volets: le premier concernait les nuisances radiologiques associées à la production d'énergie et le second l'exposition résultant de l'utilisation de phosphates, comme engrais.

En ce qui concerne le premier point, les données présentées lors de la session constituent certainement une contribution appréciable en ce domaine. En ce qui concerne le charbon et les lignites on a eu connaissance de données chiffrées sur les rejets à partir de ces combustibles solides européens et aussi de compositions de charbon importé, ainsi qu'une évaluation de l'exposition de populations au voisinage, résultant de ces rejets. Il apparaît que le thorium est peut-être un élément critique de ces rejets. Il nous semble que ce genre d'études devrait se poursuivre dans le cadre de l'évaluation des risques pour la population en rapport avec les choix énergétiques. Cependant il ne faudrait pas limiter cette évaluation au seul aspect radiologique, mais on devrait l'élargir aux polluants conventionnels rejetés dans l'environnement en même temps, lors de la production d'énergie. Il faudrait peut-être aussi à l'avenir considérer l'impact de technologies avancées, telles que la conversion du charbon et notamment la gazéification souterraine du charbon qui est à l'étude dans certains pays européens. En ce qui concerne l'aspect géothermique, nous avons appris que les études faites en Italie, études fouillées, révèlent une contribution importante due au radon et la conséquence sur le niveau de contamination de la chaîne alimentaire par les descendants de ce radionuclide. Il nous semble souhaitable que ce type de recherche soit également poursuivi. On pourrait aussi procéder simultanément à l'indentification de voies de transfert pour des éléments chimiques qui pourraient être associés à la production de cette source d'énergie.

En ce qui concerne maintenant l'exposition résultant de l'utilisation des phosphates, il apparaît que le problème majeur ne se situe pas dans le transfert dans la chaîne alimentaire des radioéléments apportés par les engrais phosphatés, notamment le radium-226. Cette contribution ne représente en général que quelques pourcents par rapport au radium présent naturellement dans la chaîne alimentaire. Par ailleurs, l'étude

détaillée faite en République Fédérale d'Allemagne indique que l'exposition externe de la population résultant de l'utilisation des engrais est très faible, mais un problème pourrait se poser au niveau des travailleurs à la fois par irradiation externe et par l'irradiation interne consécutive à l'inhalation du radon émis. Ce type de risque mériterait de faire l'objet d'études plus détaillées à l'avenir.

R. KIRCHMANN

As you are aware, the fourth session dealt with technologically modified exposure, to which Dr. JACOBI has already referred. There were two aspects to the question: radiological pollution associated with energy production and exposure resulting from the use of phosphates as fertilizers.

The data on the first aspect presented during the session form a substantial contribution in this field. We were given statistical data on releases from coal and brown coal, based on solid fuels produced in Europe and on the composition of imported coal, together with an assessment of the resultant exposure of neighbouring populations. Thorium is perhaps a critical component of these releases. It seems that further studies of this type should continue within the framework of assessing the risks to the population which are associated with various modes of energy production. However, I do not feel that assessments should be restricted to the radiological aspect but should be extended to include the accompanying conventional pollutants discharged into the environment in the course of energy production. Future work should perhaps also consider the impact of advanced technology, such as coal conversion and, in particular, the underground gasification of coal currently being studied in some European countries.

Moving to the geothermal aspect, we have learned that the intensive studies carried out in Italy reveal substantial contamination by radon and the consequential effects of its daughters on food chain contamination levels. We believe that research in this field should also be continued, with concurrent work to identify the transfer pathways for chemical components which may be associated with geothermal energy production.

In the case of exposure resulting from the use of phosphates, the main problem does not seem to lie in the transfer through the food chain of radioisotopes, particularly radium-226, introduced in phosphate fertilizers. This source generally accounts for only a few percent of the radium present naturally in the food chain. On the other hand, the detailed study carried out in the Federal Republic of Germany suggests that, while the external exposure of the population resulting from the use of fertilizers is very low, a problem may arise in the case of workers as regards both external and internal exposure, the latter resulting from inhalation of the radon emitted. Such a risk warrants future study in greater detail.

ADDITIONAL ASPECTS RAISED FROM THE FLOOR

After the above summaries given by the session chairmen, the following particular points were discussed:

- MEASUREMENTS OF EXTERNAL RADIATION. Except for cosmic radiation there are few remaining problems. The main difficulty is defining a suitable measurement grid for external radiation. In practice, measurements can only be at discrete positions, given the physical impossibility of taking measurements over a whole country using transportable instruments such as, for example, might be carried by individuals, and the prohibitive cost of measurements using aircraft. However, there is still need to define the significance of field measurements with regard to a knowledge of the doses received by members of the population. Here, the wearing of an individual dosimeter is not a satisfactory solution in view of the other sources of radiation to which individuals are exposed (watches, television, etc), frequently without being aware of it.

- CONSTRUCTION MATERIALS. We need more information on the exposure caused by construction materials, since these often give the major contribution. At this stage, however, it is too early to conduct epidemiological studies - except perhaps in the United Kingdom - owing to the lack of comprehensive data both on exposure and on mortality and morbidity. In any case, one of the problems in such studies will be to take into account the other forms of pollution to which inhabitants are exposed and to weight the corresponding exposures.

- INSUFFICIENCY OF DATA ON UPTAKE. Attention must also be drawn to the inadequancy or, in some cases, total lack of data on the levels of lead-210, polonium-210 and thorium-230 and 232 in the food intake. In addition, not enough is known about the processes by which these radionuclides are transported in the environment or accumulate in the food chain.

- SIGNIFICANCE OF OTHER EXPOSURE PATHWAYS. More data are required on the doses resulting from inhalation of lead-210, polonium-220 and thorium from airborne combustion products and mining.

  At Community level, consideration is being given to a study to identify all the different technology-based sources of exposure of man. In the United Kingdom some data have already been obtained on various sources, tin ore in particular, and these data could be made available to the Commission.

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Finally, on various sides pleas have been expressed for the implementing a co-ordinated approach to work in the European Community on natural radiation. Thus future meetings of specialists in this field at regular intervals would be welcomed as a means of improving exchanges of information.

QUELQUES ASPECTS DE LA DISCUSSION

Après l'exposé de chacun des présidents de session, les participants ont abordé en outre les aspects marquants suivants:

- LA MESURE DU RAYONNEMENT NATUREL EXTERNE. Excepté celle du rayonnement cosmique, celle-ci ne pose plus guère de problème. La difficulté majeure est de définir un maillage de mesure adéquat du rayonnement externe. En pratique, les mesures ne peuvent être que discontinues vu l'impossibilité matérielle de couvrir tout un pays par des mesure faites à l'aide d'appareils transportables, par exemple à dos d'homme, ou vu le coût prohibitif des mesures faites par avion. Il reste de toutes façons à définir la significativité des mesures faites sur le terrain en regard de la connaissance de la dose reçue par les personnes. A cet égard, le port d'un dosimètre individuel n'est pas une solution satisfaisante du fait des autres sources d'exposition auxquelles les individus se trouvent soumis souvent à leur insu (montres, télévisions, etc).

- MATERIAUX DE CONSTRUCTION. Il y a lieu de mieux connaître l'exposition en résultant car leur contribution à l'exposition de l'homme est souvent prédominante. Il est cependant trop tôt pour entreprendre des études épidémiologiques - sauf peut-être au Royaume-Uni - sur les bases trop fragmentaires dont on dispose actuellement tant sur cette exposition que la mortalité et la morbidité. En tout cas, une des difficultés de ces études sera de tenir compte des autres polluants auxquels sont soumis les habitants et de pondérer les expositions correspondantes.

- LACUNE DES CONNAISSANCES SUR L'INCORPORATION. Il est à signaler le manque total ou l'insuffisance de données sur les teneurs en plomb-210, polonium-210, thorium-230 et -232 dans le régime alimentaire. On manque en outre de connaissances sur les mécanismes de transport dans le milieu ambiant et sur ceux d'accumulation dans la chaîne alimentaire de ces radionucléides.

- SIGNIFICATIVITE D'AUTRES VOIES D'EXPOSITION. On manque de données sur les doses reçues par inhaltion de plomb-210, polonium-210 et thorium venant des cendres volantes ou de l'exploitation minière.

  Au niveau de la Communauté on songe à entamer un travail d'identification des différentes sources d'exposition de l'homme liées à la technologie. On dispose à ce sujet au Royaume-Uni de données sur les différentes sources. Parmi celles-ci il y a lieu de signaler le minerai d'étain. Ces données pourront être mises à la disposition de la Commission.

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Enfin, de plusieurs côtés des voeux sont formulés pour annoncer une coordination des travaux sur la radioactivité naturelle menés dans la Communauté européenne. Il est aussi souhaité que désormais des séminaires réunissent à intervalle régulier les spécialistes du domaine afin d'améliorer l'information mutuelle.

EINIGE NEUE GESICHTSPUNKTE, DIE SICH IM VERLAUF DER DISKUSSIONEN ERGEBEN HABEN

Im Anschluss an die vorstehenden Übersichtsberichte der Sitzungsleiter gingen die Teilnehmer noch auf folgende Aspekte ein:

- MESSUNG DES NATÜRLICHEN EXTERNEN STRAHLUNGSPEGELS. Die Messung dieser Komponente stellt - abgesehen von der kosmischen Strahlung - eigentlich kein Problem mehr dar. Die grösste Schwierigkeit besteht darin, ein zweckmässiges Messnetz für die Erfassung der externen Strahlungskomponente einzurichten. In der Praxis sind kontinuierliche Messungen nicht durchführbar, weil es nicht möglich ist, Messungen mit transportablen, z.B. tragbaren Apparaten in einem ganzen Land durchzuführen und für Messungen vom Flugzeug aus die Kosten viel zu hoch sind. Auf jeden Fall bleibt aber herauszufinden, welche praktische Bedeutung die Ergebnisse von Feldmessungen in bezug auf die vom Menschen tatsächlich aufgenommene Dosis haben. Hierfür ist nämlich das Tragen eines Personendosimeters allein keine befriedigende Lösung, da auch andere Expositionsquellen, denen die jeweilige Person ausgesetzt sein könnte, häufig unwissentlich mit erfasst werden (Uhren, Fernsehen usw.).

- BAUSTOFFE. Erforderlich sind genauere Kenntnisse über die Exposition aus Baustoffen, da ihr Beitrag zur Strahlenbelastung des Menschen häufig einen Grossteil ausmacht. Abgesehen möglicherweise von Grossbritannien scheint es aber noch zu früh zu sein, epidemiologische Studien in diesem Rahmen durchzuführen, denn die gegenwärtig verfügbaren Grundlagen sowohl im Hinblick auf diese Exposition als auch auf Mortalität und Morbidität sind noch allzu unvollständig. Ein Problem wird bei diesen Studien allemal darin liegen, dass man auch den anderen Noxen, denen Hausbewohner unterworfen sein können, Rechnung tragen und die Belastungen ihrer jeweiligen Bedeutung entsprechend berücksichtigen muss.

- INFORMATIONSLÜCKEN HINSICHTLICH DER INKORPORIERUNG. Hingewiesen werden muss auf das völlige Fehlen bzw. die Unzulänglichkeit der Angaben zum Gehalt an Blei-210, Polonium-210 und Thorium-230 und -232 in Nahrungsmitteln. Ausserdem sind die Transportmechanismen in der Umwelt und die Anreicherung dieser Radionuklide längs der Nahrungskette nur unzureichend bekannt.

- ANDERE EXPOSITIONSWEGE. Im Rahmen der Gemeinschaft bestehen Pläne für ein Projekt zur Identifizierung der verschiedenen den Menschen belastenden Expositionsquellen, die auf die Anwendung neuerer Techniken zurückzuführen sind. Im Vereinigten Königreich liegen bereits derartige Angaben über die verschiedensten Quellen vor, von denen vor allem auch das Zinnerz zu nennen ist. Diese Daten können der Kommission zur Verfügung gestellt werden.

  Es fehlen Angaben über die infolge Inhalation von Blei-210, Polonium-210 und Thorium aus Flugasche oder im Bergbau aufgenommenen Dosen.

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Schliesslich ist zu vermerken, dass mehrfach der Wunsch zu einer Koordinierung der Untersuchungen auf dem Gebiet der natürlichen Radioaktivität innerhalb der Gemeinschaft vorgebracht wurde. Als nützlich auf diesem Wege werden periodische Treffen der auf dem Gebiet tätigen Spezialisten angesehen, die sich so wechselseitig über Fortschritte in ihren Arbeiten informieren könnten.

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