



**REPORT OF THE
UNITED NATIONS
SCIENTIFIC COMMITTEE
ON THE
EFFECTS OF ATOMIC RADIATION**

GENERAL ASSEMBLY

OFFICIAL RECORDS : TWENTY-FIRST SESSION

SUPPLEMENT No. 14 (A/6314)

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Annex A

RADIATION FROM NATURAL SOURCES

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I. Introduction

1. Radiation from natural sources was examined both in the 1958¹ and in the 1962² reports of the Committee. Its importance lies in the fact that the human species has always been exposed to natural radiation at relatively stable average levels and that doses from natural radiation are being used by the Committee as a standard of comparison with those received from other, man-made, sources.

2. The present review is essentially an updating of the earlier ones and should be read in conjunction with them. The relative emphasis here given to individual topics—as, for instance, neutron doses and radio-activity in air—thus reflects the amount of new information which has become available since 1962 rather than their actual importance in the over-all study of natural radio-activity. Much of the new information has been the result of improvements in instrumentation which have been stimulated by the study of environmental contamination.

3. Natural radiation arises from two sources: cosmic rays, entering the atmosphere from outer space, and radio-active materials in the earth's crust. These materials were already present when the earth was formed,

or, in the case of shorter-lived radio-isotopes, they are continually being produced by radio-active decay or nuclear reactions. Some radio-isotopes are produced by the interactions of secondary cosmic rays, mainly neutrons, with atmospheric gases and, to a small extent, by the interactions of the cosmic rays that reach the earth's surface.

4. Owing to the varying content of natural radio-active elements in the soil and the underlying rock, the intensity of external radiation and the levels of radio-active intake by man vary from place to place. Because of limited geographical representation and of the obvious limitation of sampling, the arithmetic means calculated from the most frequent values were accepted, but they may not be strictly representative for the whole world population.

5. It is obvious that arbitrary criteria are used to separate typical situations from those where background radiation is considered elevated either because of local abundance of radio-active material or because of special food chain mechanisms. In the present review, only those situations will be described as non-typical in which at least one of the factors contributing to the natural irradiation of man is higher than typical by one order of magnitude or more.

II. Cosmic rays

INTRODUCTION

6. Cosmic radiation consists of primary radiation entering the atmosphere from outer space and of secondary cosmic radiation produced by interactions of the primary radiation with nuclei in the atmosphere. Most measurements of the primary component have been carried out at atmospheric depths of 15 g/cm² or more (up to about 30 km), but extrapolations to outer space as well as direct measurements indicate that the primary component consists of highly energetic, positively charged nuclei, with protons constituting 83-89 per cent of the primary radiation, alpha particles 10-15 per cent, leaving 1-2 per cent for nuclei having $Z \geq 3$ and for some energetic electrons. About one positively charged particle/cm² × second with an average energy of 2×10^8 MeV arrives at the top of the atmosphere.³⁻¹²

7. This primary radiation is incident from all directions. The major portion of the primary cosmic radiation is of galactic origin. Low energy particles of solar origin in the 10 MeV range may reach the earth during large sunflares. The contribution of solar particles to the total cosmic-ray intensity in the lower atmosphere is negligible, however, if averaged over long periods. Slight annual modulations of cosmic-ray flux densities, as well as variations during magnetic solar storms, have been observed.¹³⁻¹⁷

8. The secondary cosmic-ray component comprises many types of radiation produced by nuclear collisions of the primary particles with nitrogen, oxygen or argon nuclei mainly in the upper atmosphere. At about sea level, this secondary radiation is usually divided into three separate groups: the muon, the nucleon, and the electron component. Some properties of the main secondary particles in cosmic radiation are summarized in table I.

9. *The muon component.* Muons (previously called μ -mesons) are the daughters of short-lived pions. The pions result from the interaction of high energy protons with atmospheric nuclei. The maximum muon flux density occurs at an atmospheric depth of about 150 g/cm² (usually at a height of about 12 km). Ionization due to cosmic rays detected at low altitudes is mainly due to the penetrating muons (about 70 per cent).^{18, 19} The muon component still contributes about 50 per cent to the ionization from cosmic rays at an altitude of 3 km, and its relative share continues to decrease with altitude.

10. *The nucleon component.* The nucleon component consists of nuclear fragments, mainly neutrons and protons.^{7, 12, 20, 21} It multiplies by cascade processes involving nuclear spallation in the atmosphere, whereby the secondary fragments knocked out of atomic nuclei possess high enough energy to produce still further fragments in subsequent nuclear interactions. As nitrogen and oxygen consist of an equal number of protons and neutrons, the initial nucleonic cascade in air consists of approximately equal numbers of protons and neutrons with high energies. Below about 500 MeV, however, ionization losses of protons in the air start competing with nuclear absorption: protons are therefore progressively removed from the cascade which, below a few hundred MeV, consists mainly of neutrons.

11. As neutron emission is the most probable de-excitation reaction when nitrogen and oxygen are excited to energies of about 8 MeV, neutrons in the range of a few MeV will be evaporated isotropically during the terminal process in nucleonic cascades.^{21, 22}

About 20 per cent of the energy from the incident primary radiation is transferred to nucleonic cascades. Most of it, however, is absorbed before reaching the earth's surface. Slow protons are stopped by ionizations, and slow neutrons are captured mainly in nitrogen, forming C¹⁴.

12. *The electron-photon component.* The electron-photon component includes electrons, positrons and photons. They originate mainly from the electron-positron-photon cascade resulting from the decay of the extremely short-lived neutral π^0 meson produced when high energy primary radiation interacts with atmospheric nuclei. The spontaneous decay of muons also contributes some high energy electrons to this component.

13. When cosmic rays are measured, the neutrons are usually dealt with separately from the other components. The latter, including gamma rays, are further subdivided according to their penetration power. The "soft" component consists of the radiations that are completely absorbed in about 15 cm of lead. At low altitudes, this component includes the bulk of the electrons, gammas, and protons, as well as a few slow muons. The "hard" component, some of which is only slightly attenuated by 15 cm of lead, may be able to penetrate much thicker layers. The "hard" component at sea level consists mainly of muons and of high energy protons.

GEOMAGNETIC EFFECTS

14. An approximation to the external geomagnetic field is obtained by assuming a magnetic dipole located 340 km off the centre of the earth, with poles pointing towards 80.1°N, 82.7°W, and 76.3°S, 121.2°E. This terrestrial magnetic field acts as a momentum selector for the primary charged particles. Consequently, the cosmic ray flux entering the upper atmosphere is latitude dependent as well as directional.⁷⁻⁹

15. The net result of these effects is that particles with the lowest energies reach the earth only in the vicinity of the geomagnetic poles, whereas those with energies in excess of about 6×10^4 MeV can reach the earth anywhere.²³⁻²⁸

16. The minimum momentum that an incident charged particle must possess in order to reach the earth's atmosphere despite the deflecting influence of its magnetic field is called the threshold rigidity. It varies with geomagnetic latitude as $\text{Cos}^4\phi$ and depends on the angle of incidence of the charged particle.

17. The threshold rigidity for vertically incident charged primaries at the geomagnetic equator is about 5.7×10^7 gauss × cm corresponding to a proton energy of 1.7×10^4 MeV.^{7, 27} However, the intensity of the ionization due to cosmic rays measured at sea level is only about 10 per cent higher near the geomagnetic poles than the ionization recorded at the equator. Thus, about 90 per cent of the ionization detected at sea level results from primaries which had enough momentum to arrive at the earth's geomagnetic equator. The primaries of lower momentum are relatively ineffective in producing penetrating muons.

18. The latitude variations are more pronounced in the case of the nucleonic component, as shown in figure 1. This latitude effect of neutrons (and of secondary protons) demonstrates that the nucleonic component is mainly produced by low energy primaries that are prevented by the earth's magnetic field from reaching the earth between 50°N and 50°S but that

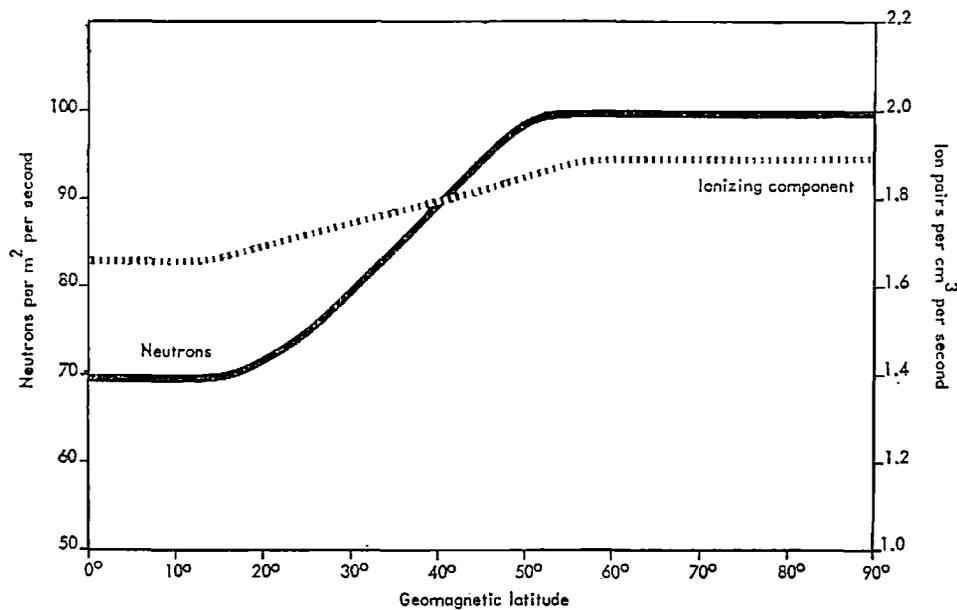


Figure 1. Geomagnetic latitude dependence of the ionizing component and neutrons at sea level^{9, 27, 28}

do reach the polar regions. Pion production, on the other hand, predominates at higher energies (above a few 10^4 MeV) and therefore is much less affected by the geomagnetic field.

19. The apparent plateau region for the secondary components at high geomagnetic latitudes has been extensively studied.^{4, 11, 13-17} Atmospheric attenuation due to ionization losses, and insufficient sensitivity of some experimental equipment, might partially account for it. However, there seems to be a real decline in the flux density of primary particles of galactic origin for energies less than 2×10^4 MeV per nucleon. Low energy primaries reaching the upper atmosphere are mostly of solar origin.

20. As the geomagnetic poles do not coincide with the earth poles and as the magnetic dipole representation of the geomagnetic field itself is a rather crude approximation which needs correction terms (higher magnetic moments) to account for the actual energy cut-off of primary cosmic rays, it is easy to explain the slight longitudinal effect observed in the relative intensities (up to 15 per cent) of cosmic radiation. Temporal distortions of the geomagnetic field due to solar activity are also observed.

21. It should be emphasized that the geomagnetic latitude effect revealed by the secondary cosmic radiations should be wholly attributed to the primary component. Owing to their comparatively short trajectory from the point of production in the atmosphere to ground level, no appreciable geomagnetic deviation is expected for the secondary charged particles.

ENERGY SPECTRUM OF PRIMARY RADIATION

22. Figure 2 represents the integral energy spectrum of primary protons.^{6-12, 20, 29, 30} The integral energy spectrum per nucleon of the total primary radiation is similar within the experimental accuracy. The latitude-sensitive portion of cosmic radiation up to about 6×10^4 MeV—the threshold rigidity for positively charged protons from the east at the magnetic equator—is often referred to as “low energy radiation”. As low energy particles are much more abundant, most of the cosmic-ray energy will reach polar regions where,

therefore, most of the secondary rays will be produced. The term, “very low energy radiation”, is reserved for primaries below 1.7×10^8 MeV, whose abundance undergoes seasonal changes correlated with solar events.

COSMIC RAYS IN THE ATMOSPHERE

23. Figure 3 shows the altitude variations of the main components of cosmic radiation in the atmosphere.^{7-9, 26, 29} Production of the secondary particles

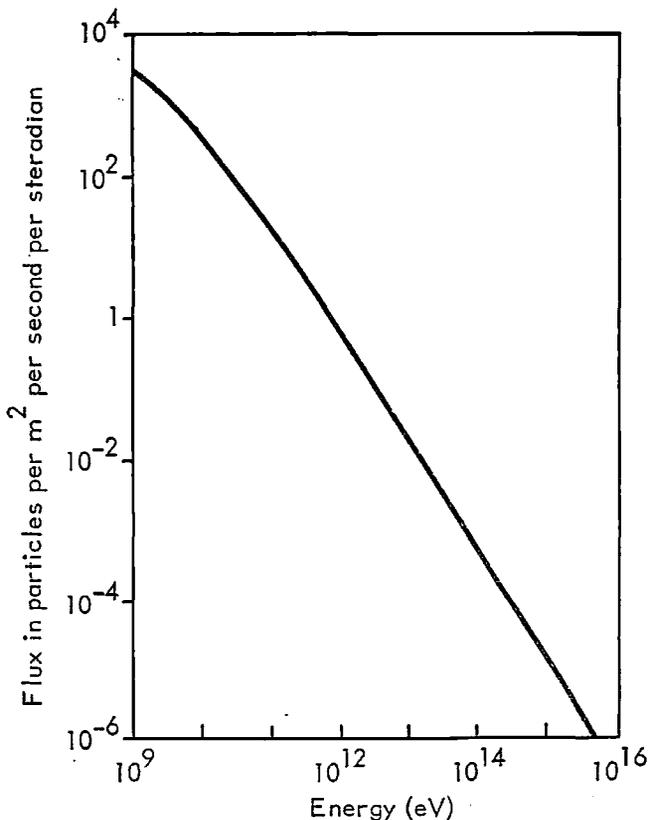


Figure 2. Integral energy spectrum of the primary cosmic-ray protons^{9, 10}

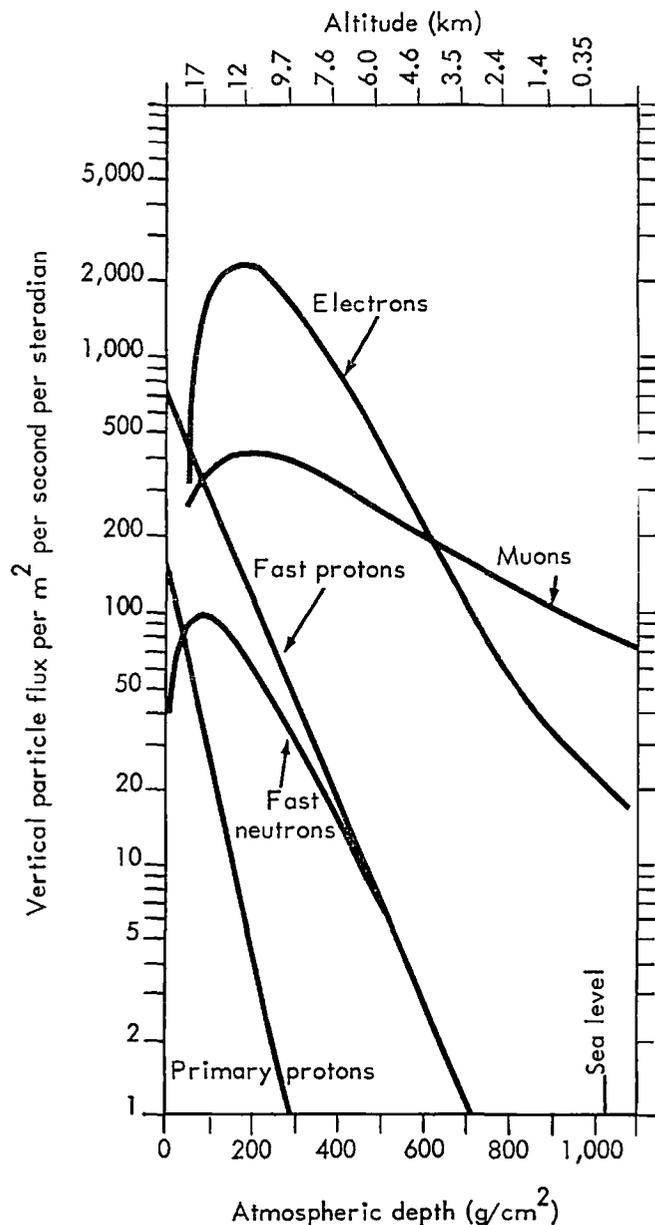


Figure 3. The vertical flux densities for the main components of cosmic radiation as a function of atmospheric depth (at geomagnetic latitude of 45°N)⁷

increases with atmospheric depth up to about 150 g/cm² (down to about 12 km) where the maximum flux density of cosmic rays is found. Below this altitude, particle loss through capture, ionization and muon decay predominates over production, and the various secondary cosmic-ray components decrease exponentially with decreasing altitude. The primary radiation carries almost all the energy above 25 km, while the muon component predominates below 3 km.

24. Atmospheric effects depending on barometric pressure and temperature are known to affect somewhat the cosmic-ray intensity at sea level, especially the muon component. The muon flux density reaching sea level depends on the thickness of the atmosphere below the point of production, and, because some muons decay in flight, will also depend on their path length in the atmosphere. These local changes are of the order of a few per cent and will not be discussed any further.

25. The vertical component of cosmic radiation is of relatively large significance at sea level, as atmos-

pheric absorption of secondary radiation and muon decay are minimal for vertical incidence. The east-west effect being small compared to the absorption effect at various angles, the lower threshold rigidity of particles coming from the west does not compensate for the additional attenuation of the secondary radiation in the atmosphere. For radiation from the east, the higher rigidity threshold and atmospheric attenuation both operate to diminish the secondary flux arriving at sea level.

Radio-nuclides produced by cosmic rays

26. Some of the nuclear fragments resulting from spallation of atmospheric nuclei during the nucleonic cascade process are radio-active.³¹⁻³⁸ About 1.7 spallations/cm² × second is the global average induced by cosmic rays. They occur in atmospheric gases in proportion to their relative abundance (nitrogen : oxygen : argon ~ 76.5 : 22.5 : 1). The distribution of nuclide production rates within the atmosphere, taking into account the energy spectrum of the nucleon component of cosmic radiation, was calculated by Lal.^{32, 39} Most radio-nuclides are produced in the stratosphere by low energy neutrons, their rate of production being approximately proportional to the neutron flux density and subject to its pronounced latitude dependence.

27. The main radio-nuclides produced by cosmic rays are listed in table II with some of their nuclear properties, calculated production rates and activity concentrations in the lower troposphere. C¹⁴ is mainly formed by the N¹⁴(n, p)C¹⁴ capture reaction with atmospheric nitrogen. About two-thirds of the neutrons produced by cosmic rays are removed through this process from the atmosphere.

28. The production rates of radio-active spallation fragments from oxygen and nitrogen (H³, Be⁷ and Be¹⁰) exceed by far the production rate of fragments due to argon spallation. Cosmic rays which reach the earth's surface can interact and produce radio-active nuclides, but these have extremely low activity and are unimportant in comparison with other nuclides of natural origin.

COSMIC-RAY NEUTRONS

29. As neutrons usually elude detection when ionization chambers are used, this nuclear component of cosmic radiation should be assessed separately. Cosmic-ray neutrons are produced by two kinds of reactions.^{5, 23, 28, 41-49} First, neutrons are knocked out of nuclei as a result of nuclear collision of high energy cosmic rays. These neutrons have energies from a few MeV up to more than 1,000 MeV. A larger source of neutrons in the atmosphere, however, is represented by evaporation neutrons which have an energy distribution peaked at about 1 MeV.^{12, 21, 23, 43, 50, 52} The evaporation process may account for about 80 per cent of the atmospheric neutron flux density according to estimates derived by Hess *et al.*⁴⁴

30. Neutrons produced in the atmosphere will eventually either leak out of the upper atmosphere into space or disappear through absorption. Atmospheric absorption occurs largely through N¹⁴(n, p)C¹⁴ capture reactions after the neutrons have been slowed down by elastic and inelastic collisions in some 150 g/cm² of air. A stationary condition in time is thus maintained where the neutron flux density in the atmosphere is proportional to the neutron production rate at a somewhat higher altitude.

31. Owing to the low flux density of neutrons produced by cosmic rays at sea level, most measurements concerning the neutron component were carried out at various heights in the atmosphere within the equilibrium region, below 150 g/cm² (up to 12 km) where the energy spectrum of neutrons is essentially constant.^{29, 41-58} From these measurements, estimates of neutron fluxes at sea level were obtained through extrapolations. It is, however, difficult in making such extrapolations to allow for the actual situation that obtains at sea level because of the presence of an interphase and of backscatter with a consequent breakdown of the atmospheric equilibrium conditions.

Flux densities and energy spectra

32. A major limitation in comparing the results of different experimenters is the interdependence between the energy spectrum, which is assumed, and the absolute neutron flux density that is derived from the measurements. Each detector is sensitive in a limited energy range only, and its sensitivity is often energy dependent.⁴¹ Thus, the interpretation of counting rates obtained through detectors of different kinds presupposes a detailed knowledge of the energy spectrum of cosmic-ray neutrons.

33. Figure 4 shows the cosmic-ray neutron energy spectrum in the equilibrium region as derived by Hess *et al.*^{44, 52} They used a set of detectors sensitive to different energy ranges from thermal energies up to about 10⁴ MeV. Their extrapolation to sea level yielded a flux density of 4 × 10⁻² n/cm² × second, of which 15 per cent were between 1 and 10 MeV, 75 per cent had lower energies, and about 10 per cent were above 10 MeV.

34. Miyake *et al.*⁴³ obtained an energy spectrum of cosmic-ray neutrons from 1 to 15 MeV by observing recoil protons by means of a high pressure cloud chamber filled with hydrogen. Their derived energy spectrum for 1 eV to 10⁴ MeV neutrons is in good agreement with figure 4, especially above 10 MeV and in the eV region.

35. Another calculation of the equilibrium spectrum of neutrons in the atmosphere up to energies of 20 MeV was performed by Newkirk.⁵¹ This spectrum also differs from Hess' spectrum mainly in the energy range of 0.1 to 4 MeV, and thus agrees better with other measurements.^{53, 55}

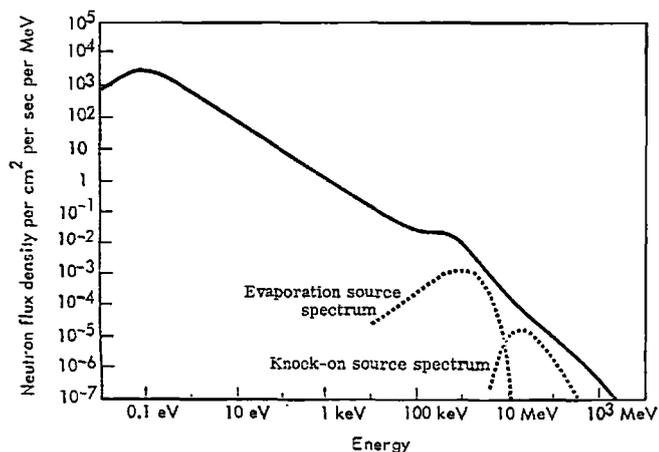


Figure 4. The equilibrium neutron flux density versus energy at sea level (44°N)⁴⁴ (the assumed spectra of the two neutron sources are also shown)

36. Subsequent workers arrived at lower estimates for the total neutron flux density.^{49, 55-59} The lower values might partly be due to the special care taken to minimize neutron production in the instruments themselves and in the surrounding materials. In particular, lightweight detection equipment flown with balloons was used instead of heavier aircraft-borne detectors.

37. Kastner *et al.*⁶⁰ using a liquid scintillator, reported a flux density of 4.5 × 10⁻³ n/cm² × second for neutrons with energies from 1 to 10 MeV as measured directly at an altitude of 180 metres. They suggested a total neutron flux density at sea level (41°N) of about 10⁻² n/cm² × second, 15-25 per cent of which are in the energy range 1 to 10 MeV, and about 70 per cent have lower energies.

38. It seems reasonable to follow the suggestion of Kastner *et al.*⁶⁰ and to assume the value of 10⁻² n/cm² × second as the total neutron flux density at sea level. A lower value of 0.54 × 10⁻² n/cm² × second at sea level as given by Haynes,⁵⁸ was recently used as a basis for dose estimates.⁶¹

COSMIC-RAY DOSE RATES^a

39. In the 1962 report,² the dose rate to soft tissue due to the ionizing component of cosmic rays at sea level at middle latitude was estimated as 28 mrad/year. Recent reported values generally agree with this figure.^{18, 19, 62-66} Table III lists the reported values in ion pairs/cm³ × second as well as in mrad/year.

40. Recent surveys¹⁸ confirmed the altitude dependence of the ionizing component as discussed in the 1962 report. Muons are the main contributors to absorbed dose at low altitudes (some 80 per cent of the

^a The "dose" concept. The interaction between radiation and matter results in a variety of processes that include absorption, scattering and the production of secondary radiations. Hence, when the human body is exposed to radiation, the distribution of absorbed energy is usually not uniform. Although such quantities as the surface dose, the maximum dose and the average dose can therefore differ appreciably depending on the nature, energy and direction of the incident radiation, it is frequently desirable to utilize a single numerical index to specify the degree of irradiation. This is usually termed the "dose" and expressed in the unit "rad" or its submultiples (mrad, etc.). The usual meaning is the one that will be adopted here when the term "dose" is employed without further specification. It corresponds to

- (a) The kinetic energy of the secondary charged particles produced per unit mass of soft tissue of approximate composition (H₄₀C₅O₈N)_n, in the case of indirectly ionizing radiations (neutrons, photons and other uncharged particles);
- (b) The energy deposited per unit mass in matter of the same composition as given in (a), in the case of directly ionizing radiations (all charged particles).

These quantities are obtained when the absorbed energy is computed from the appropriate interaction cross sections for the incident radiation spectrum. They will also be obtained within a few per cent, if the absorbed dose is determined in a small tissue-equivalent detector having a wall of sufficient thickness to establish secondary charged particle equilibrium. In most instances, the "dose" thus determined will differ by less than a factor of two from the maximum dose in a human phantom.

It is recognized that the above definitions do not conform with recently recommended usage.⁶⁹ In particular, (a) defines tissue kerma rather than the absorbed dose.⁶⁸ This departure from rigorous terminology is necessitated by the fact that most of the literature sources use this approach and by the absence of an acceptable term for (b).

The relative biological effectiveness (RBE) of any radiation depends not only on its type and quality but also on the effect under study and on other factors, such as dose, dose rate and the value of physiological parameters. This is because, even when equal amounts of different radiations are absorbed in any tissue, their effect is usually different as a result of differences in the microscopic distribution of the absorbed energy.⁶⁷

ionizing component according to Lillcrap,¹⁹ 70 per cent according to Lowder and Beck¹⁸).

Neutron dose rates

41. On the basis of Hess' energy spectrum^{44, 52} and the usual factor to convert flux densities to dose rates,^{61, 70, 71} $10^{-2}\text{n/cm}^2 \times \text{second}$ is assumed to correspond to a dose rate of 0.7 mrad/year. However, this value is considered to be an over-estimate, since the spectrum derived by Newkirk⁵¹ gives a lower dose rate for the same total flux density of neutrons. As most of the other experimental values for flux densities range from 0.4 to $1.5 \times 10^{-2}\text{n/cm}^2 \times \text{second}$,^{53, 55, 57, 58, 60} one obtains a range from 0.3 mrad/year to 1.1 mrad/year for the neutron dose rate at sea level.

42. Neutron flux densities, and to a lesser extent the energy spectrum, are latitude dependent. Thus, assuming 0.7 mrad/year at 41°N, slightly higher dose rates will be encountered near polar regions,^{57, 61} whereas at the equator the neutron dose rate might be about 30 per cent lower or approximately 0.5 mrad/year.

43. *Correction factors.* A number of factors need to be considered when estimating radiation dose rates to human tissue (outdoors or in buildings) from an established neutron flux density in air with a known energy distribution. Owing to the abundance of hydrogen atoms in the human body, neutrons produced in the atmosphere lose their energy faster in traversing 1 g/cm² of tissue than in traversing 1 g/cm² of air.⁷²⁻⁷⁶

44. Practically all neutrons entering the human body with energies below 5 MeV lose their energies by elastic collisions with hydrogen and are finally absorbed by $\text{H}(n, \gamma)\text{D}$ or $\text{N}^{14}(n, p)\text{C}^{14}$ reactions. The neutron dose decreases rapidly with increasing depth^{71, 75} and at 10 cm beneath the surface the dose is about one-third to one-tenth of that at the skin, depending on the energy and isotropy of the neutrons.

45. As neutrons lose on the average about half of their energy in the first collision with a hydrogen nucleus, neutrons of higher energy (above 5 MeV) lose significantly more of their energy in the human body than in the same mass of air (or other non-hydrogenous materials). However, neutrons above 5 MeV are relatively rare in the equilibrium spectrum of air, thus limiting this additional contribution to human tissue dose to 20 per cent at most of the neutron dose.

46. Neutron production in the human body must also be considered. One may assume that the cross sections per atom for the production of evaporation neutrons in the body (oxygen, carbon) are comparable with those in air (oxygen, nitrogen). The neutron component which is responsible for most of this terminal stage in the nuclear cascade leading to evaporation neutrons has a significantly shorter mean free path in the body than in air. More neutrons will therefore reach this final stage of evaporation interactions in the body than in an equivalent mass of air, thus tending to raise the average tissue dose received from neutrons.

47. As to the shielding by buildings and the computation of neutron dose indoors, tissue equivalent material might be as likely to add somewhat to the dose as to shield part of it, because of the scattering effects and the possibility of higher production rates in these materials as mentioned above. Heavy construction materials, such as lead and iron, tend to add to the neutron background.^{22, 77-79} Thus, it seems unjustified to allow for the shielding effect on neutron doses unless

detailed knowledge on the composition of the building materials is available.

CONCLUSIONS

48. The contribution of the ionizing component of cosmic rays at sea level was estimated in the 1962 report as 28 mrad/year at middle latitudes, which is also the best value agreed upon lately.^{18, 63, 65} This dose rate of ionizing radiation is mainly delivered by muons¹⁹ and is subject to the slight variations due to latitude effects which may lower the dose rate by about 10 per cent near the equator. The total dose rate approximately doubles for every 1,500-metre increase in altitude for the first few kilometres.^{2, 18, 64} The relative importance of the nucleonic component as compared to the muon component increases with altitude. These facts will be of importance in assessing doses received at high altitudes, including space flights and other high altitude flights.⁶¹

49. Revised data on cosmic-ray neutron flux densities indicate that, without allowing for shielding by building structures and for screening by body tissues, the corresponding absorbed dose is 0.7 mrad/year (range 0.3-1.1 mrad/year) in temperate and polar regions, while in equatorial regions it is likely to be around 0.5 mrad/year. This value may be compared with the dose rate of 2 mrad/year which can be inferred from the estimate given in the 1962 report.

III. Terrestrial radio-activity

SOIL

50. Practically all the natural environmental radiation of terrestrial origin is due to radio-nuclides of the uranium and thorium series and to K^{40} . Typical abundance figures for the accessible lithosphere are 2.8 parts per million of uranium and about eleven parts per million of thorium. The nuclides belonging to these two radio-active series and their more important properties are listed in tables IV and V. The U^{235} series is of lesser importance but is listed for completeness in table VI. A schematic representation of these three decay series is given in figure 5.

51. There are other primordial radio-nuclides in the earth's crust. Of these, the greatest contributor to terrestrial radiation is K^{40} , which makes up about 0.01 per cent of natural potassium. Certain properties of this nuclide are listed in table VII, along with the properties of selected primordial radio-nuclides which are of lesser importance because of low abundance, long half-life or weak radiations.

EXTERNAL RADIATION

52. External natural radiation is produced mainly by the gamma emitters of the natural radio-active series and by K^{40} in soils, rocks and construction materials, by cosmic radiation as discussed in part II, and to a small extent by atmospheric radio-activity. Outdoor and indoor terrestrial gamma dose rates measured before 1961 were summarized in tables IV, V and VI of annex E of the 1962 report.

53. Measurements of the total gamma dose obtained by means of high pressure argon or tissue equivalent ionization chambers,⁸⁰⁻⁸³ were recently supplemented by gamma spectroscopy in order to assess separately the various contributions to the external radiation dose.^{64, 81-89, 91, 92} Figure 6 shows a typical gamma spectrum on which the K^{40} photo-peak of 1.46 MeV and the main peaks of the uranium and thorium series are indicated.

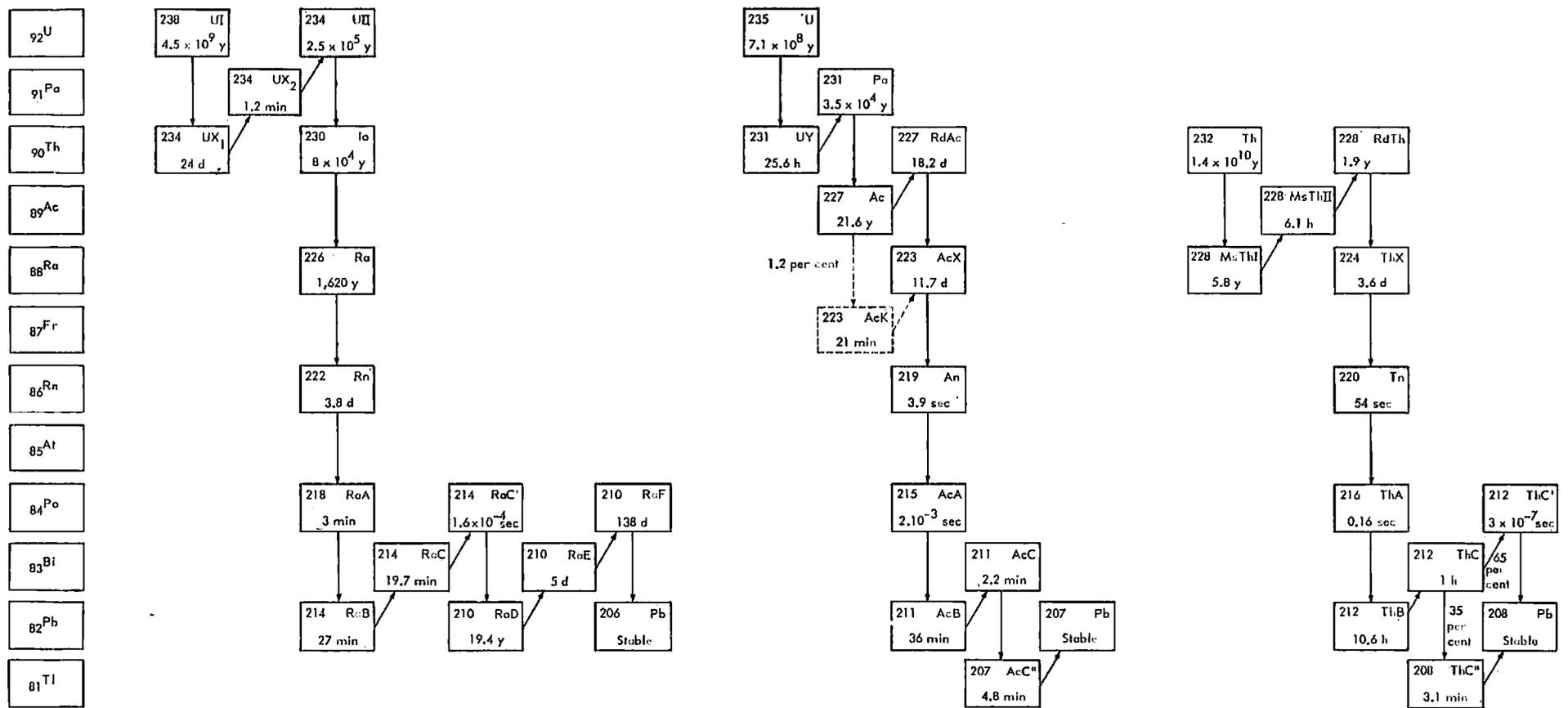


Figure 5. Decay schemes of the natural series^a (boxes show atomic weight, historical name and half-life)

^a Parallel decay branches of less than 1 per cent are not included.

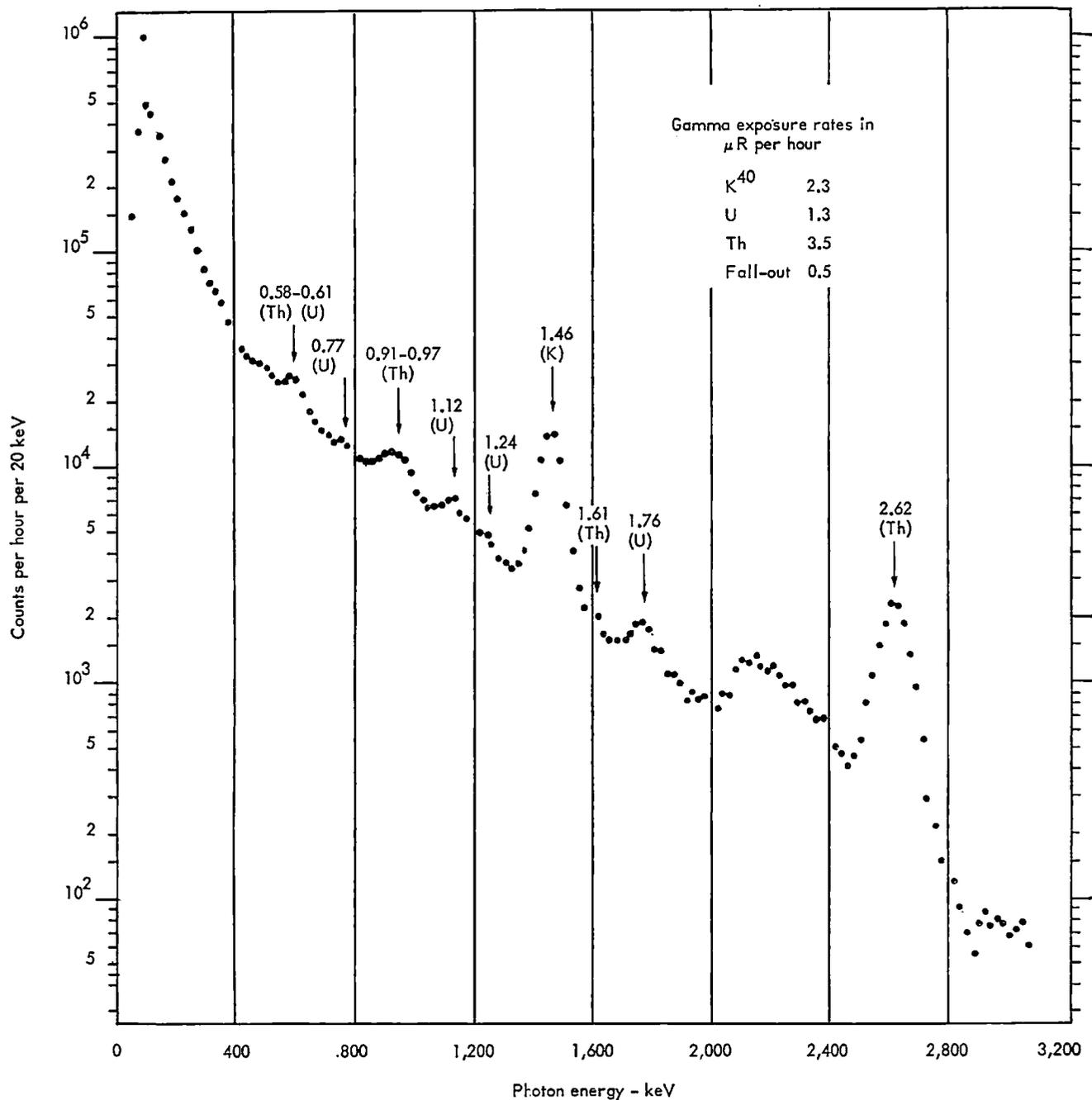


Figure 6. Environmental gamma radiation spectrum, 4 × 4" NaI(Tl)-scintillation detector, Denver, Colorado, 12 August 1965²⁴¹

54. The contribution of each component to the air dose, as derived from the evaluation of gamma spectra, using the 2.62 MeV photo-peak of Tl²⁰⁸ to represent the thorium series, the 1.46 MeV photo-peak for K⁴⁰ and the 1.76 MeV photo-peak of B²¹⁴ for the uranium series, were often compared with doses estimated on the basis of the measured abundance of Th²³², K⁴⁰ and U²³⁸ in underlying soils or rocks.⁸⁰⁻⁸⁸ In general, the two methods gave results consistent within 5-10 per cent for K⁴⁰ and Th²³², when soil density and moisture content were taken into account.

55. No such close correlation between the dose determined from Bi²¹⁴ and the U²³⁸ concentrations in underlying soils was found, however.^{84, 89, 93} Nor could it be expected, since the movement of free radon out of the upper layers of the soil into the atmosphere generally reduces the contribution of the uranium series

to the external dose at ground level. Dose rates can be correlated with the gamma-emitting daughters of the radon retained in soil. The concentrations of the daughters is only a fraction of the actual Ra²²⁶ concentration in upper soils.^{93, 94} Moreover, U²³⁸ is not likely to be in equilibrium with Ra²²⁶ because of the greater tendency of uranium to leach from upper soil layers during soil formation and weathering processes.

56. An increase in water content of soil will increase the density and consequently the attenuation of gamma rays from K⁴⁰ and the radio-active series and will thus cause temporal variations in the dose rate correlated to soil moisture. The dose rates from the uranium series may, however, be affected also in the opposite direction, since clogging of the pores in the soil impedes the escape of radon from soil air into the atmosphere, thus resulting in an increased dose rate

from radon daughters. Furthermore, during rainfall, the wash-out of short-lived gamma-emitting daughters of radon from the atmosphere will temporarily increase the external dose rate attributed to the uranium series.^{84, 85, 90, 94-97}

57. The measured average external radiation dose rates one metre above soil in populated areas fall mostly in the range 30-65 mrad/year with a representative value of about 50 mrad/year.^{84, 92, 97-100} Potassium and thorium each generally contribute more than one-third, while the contribution from the uranium series is usually somewhat less.

58. From the point of view of many human populations, exposure levels out of doors are more relevant than indoors. Indoor doses depend primarily on the radio-active content of construction materials and on the attenuation of outside radiation by roofs and walls. There seems to be no reason for changing the value of 50 mrad/year adopted in the 1962 report, as the few additional experimental results^{101, 102} fall within the ranges quoted earlier.

59. Unusually high natural radiation areas were discussed in the 1962 report and the five major inhabited areas with increased terrestrial radiation were tabulated in table XX of annex E of that report. Further studies concerning external dose rates to the populations in these areas supplement and confirm previous data.¹⁰³⁻¹⁰⁷

60. The comments made in the 1962 report concerning the insignificance of external beta radiation to gonad or bone marrow doses must still be accepted. The external radiation dose rate from natural airborne radio-activity is usually of the order of 2 mrad/year. Since it is small compared to the dose variations discussed in paragraph 56, atmospheric radio-activity as an external radiation source will not be dealt with separately.

Neutrons

61. Neutrons in the earth's crust may be produced by interactions of soil elements with cosmic rays, by spontaneous fission of U^{238} , by (α, n) and possibly by (γ, n) reactions. While of no significance as far as doses are concerned, these neutrons, some of which escape from soils and rocks, are of interest to geologists, and may sometimes be confused with cosmic-ray neutrons.

62. An indirect estimate for the production rate of neutrons due to the interactions of cosmic rays in upper soils may be obtained by dividing the average neutron flux density at sea level by the neutrons' mean free path in air, thus neglecting the differences in atomic composition of soil relative to air. A production rate of 7×10^{-5} n/second \times g soil at mid-latitudes (corresponding to about 2,000 n/year \times g soil) is consistent with the extrapolated cosmic ray neutron flux density of 10^{-2} n/cm² \times second at sea level and a mean free path of 150 g/cm² as discussed in paragraphs 29-38.

63. Spontaneous fission of U^{238} (half-life 8×10^{15} years, and 2.2 n/fission)¹⁰⁸ gives rise to a production rate of the order of only 1.4 n/year \times g soil (assuming three parts per million U/g soil).

64. Gorshkov *et al.* report the experimental yield of 0.107-0.014 n/ 10^6 alphas for the (α, n) reaction of Po^{210} alphas on SiO_2 , and a value of 0.238 n/ 10^6 alphas for granite.¹⁰⁹ The conversion factor from the Po^{210} alpha yield to the alpha yield from the uranium series in equilibrium should be at least eight and for the thorium series at least six. Upper limits were derived from Gurfinkel's¹¹⁰ values for the (α, n) reaction on O^{18} , taking into account the increase in yield with alpha

energies. These upper limits are twelve and fourteen for the uranium and thorium series, respectively.

65. Thus, taking three parts per million of uranium and eleven parts per million of thorium as representative of the upper earth's crust,¹¹¹ an average production rate of 13-24 n/year \times g soil and of 20-50 n/year \times g granite is obtained. Assuming sixty parts per million of uranium and 110 parts per million of thorium as possible upper limits of concentrations in granite, one would obtain a production rate of 450-800 n/year \times g granite from (α, n) reactions which might add a few per cent to the cosmic-ray neutrons observed at elevated altitudes. The 28 n/year \times g granite from spontaneous fission for this case would still remain insignificant.

NATURAL ACTIVITY IN WATERS

Oceans

66. The natural radio-activity of sea water^{112, 113} is mainly due to K^{40} (300 pCi/litre), Rb^{87} and the uranium series contribute about 3 pCi/litre each to the total activity of oceans. Natural H^3 in the upper oceanic layers might range from 0.6 to 3 pCi/litre, while all other radio-isotopes, including those of the thorium series, contribute less than 0.2 pCi/litre to sea water activity.

Fresh waters

67. Natural radio-activity in fresh waters is due to some activity transfer from soils and the atmosphere. Thus the activity concentrations found in waters depend on the concentrations encountered in the rocks with which the waters are in contact.^{36, 114} Members of a radio-active series in water are rarely in radio-active equilibrium with each other because of differences in chemical and physical properties such as solubility, sorbability, etc. K^{40} is often the main contributor to the beta activity in water.

68. As drinking water is one of the media by which natural radio-isotopes are transferred to man, the concentrations of specific nuclides, mainly belonging to the Ra^{226} chain, have been extensively measured for many years.^{104, 106, 115-128} Typical concentrations in continental waters, as well as usual concentrations of Pb^{210} and Po^{210} found in rain, are given in table VIII. Tables VII and VIII in annex E of the 1962 report list some earlier values of natural radio-activity in natural waters, springs and public water supplies for various countries. Recent information accumulated since 1962 is summarized in table IX.

69. Ra^{226} concentrations in drinking water vary by orders of magnitude, though most waters show values between less than 0.1 to about 1 pCi/litre. Surface waters (lakes, rivers) usually show less activity than those derived from deep wells, especially in areas where the concentration of natural radio-active minerals in the earth's crust is higher than usual.

70. Rn^{222} concentrations in fresh water may vary from less than 1 pCi/litre up to the order of 106 pCi/litre.^{2, 125-130} Levels less than 10 pCi/litre are found in lakes and rivers, and the highest concentrations observed have been reported for some spas and spring waters.^{123, 128} Concentrations of 10^2 - 10^4 pCi/litre have been found in ground waters and even higher concentrations in deep wells (table VIII).

71. Pb^{210} in water is mainly derived from the decay of radon in the air and the resulting deposition of this nuclide with rain. For Pb^{210} concentrations in surface water seem to exceed those in water from deep wells.¹¹⁷

Pb^{210} concentrations in water do not fluctuate as strongly as Ra^{226} , and there is no correlation between the two elements nor between Pb^{210} and the sulfate, calcium or fluoride content of water. Typical concentrations are between 0.05 and 0.2 pCi/litre.

72. Information on Ra^{228} or Th^{228} concentrations in water is extremely limited.^{36, 104, 119} Turner *et al.*¹²³ observed the presence of short-lived Th-series nuclides (Ra^{224}) in water sampled in the United Kingdom. Long-lived elements, such as Th^{228} and Ra^{228} , were present only in traces. Th^{228} constituting a very small fraction of the long-lived alpha activity in water.

THE NATURAL RADIO-ACTIVITY OF AIR

73. The natural radio-activity of the atmosphere is caused mainly by Rn^{222} , Rn^{220} and their radio-active decay products. The contribution of Rn^{210} or its radio-active daughters is negligible.^{54, 36, 37} Radio-active nuclides produced by cosmic rays (table II) are of minor importance as sources of atmospheric radio-activity; the radio-activity in dust particles blown from the soil by wind, or from K^{40} brought into the atmosphere by the evaporation of sea-water spray is also extremely low. These sources will therefore not be discussed any further.

74. A simple mathematical model to account for radon exhalation rates from soils, and to estimate concentration profiles of the radon isotopes in the troposphere was developed by Israel.¹³¹ His model, with some minor changes at times, has been widely followed by many authors.^{37, 132-134}

75. The spreading out of Rn^{222} and Rn^{220} in the atmosphere after their exhalation from the ground is caused by turbulent diffusion and convection. The decay products of the radon gases, being isotopes of heavy metals, become readily attached to aerosol particles. At ground level, more than 99 per cent of the Pb^{214} , Bi^{214} and Pb^{212} atoms, and about 75 per cent of the shorter-lived Po^{218} atoms are carried by aerosols.^{34, 36, 37, 131-137} The fraction of natural radio-activity not attached to aerosol particles increases with altitude. As concentrations of aerosols in the atmosphere are about 10^6 times higher than the concentrations of radon daughters, radio-active aerosol particles will usually contain a single radio-active atom each.

76. The effective radius of aerosols which contain the main portion of natural atmospheric radio-activity is between 50 μm and 80 μm .^{37, 135} As aerosols are unstable and tend to increase in size as a consequence of condensation and coagulation processes, reported size distributions of activity are only valid for short-lived radio-active decay products. Long-lived isotopes like Pb^{210} and daughters are therefore expected to be attached to larger aerosols.

77. The distribution of the radio-active decay products of radon in the atmosphere is consequently controlled not only by radio-active decay and diffusion, but also by sedimentation and wash-out related to the removal of aerosols.

78. Figure 7 gives a vertical distribution of concentrations for the radon isotopes and their radio-active decay products for "normal" conditions, assuming a half removal time of twenty days for all the radon daughters.¹³⁴⁻¹³⁶ Full radio-active equilibrium at ground level air for Rn^{222} and its short-lived radio-active daughters is not reached because of the continuous elimination of radio-active aerosols by downward diffusion and settling. However, equilibrium between

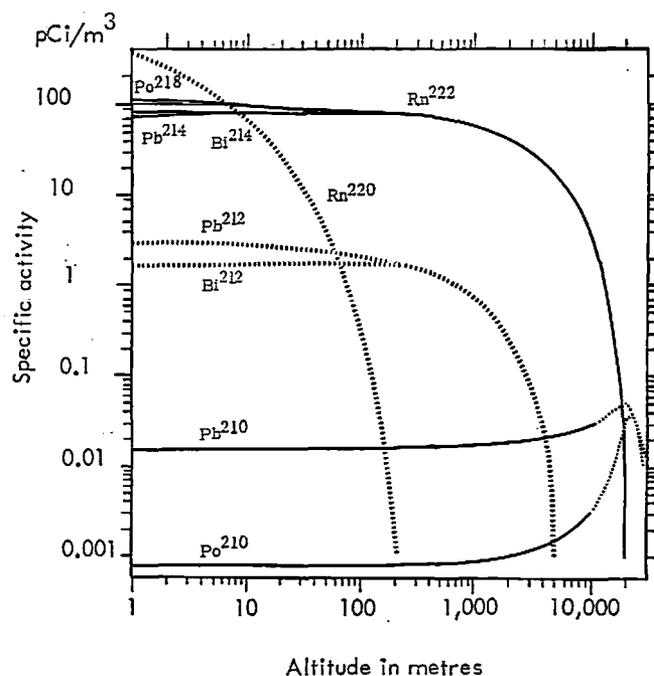


Figure 7. Vertical distribution of concentration of the radon isotopes and their radio-active decay products¹³⁴

Rn^{222} and its short-lived daughters is approached quite closely a few metres above ground.

79. Rn^{220} is virtually never in equilibrium with its radio-active decay products. The 10.6-hour half-life of Pb^{212} greatly exceeds that of Rn^{220} so that Pb^{212} atoms after their formation can diffuse to higher altitudes than Rn^{220} (or Po^{216}). A very low $\text{Pb}^{212}/\text{Rn}^{220}$ ratio, as expected, was measured by Fontan *et al.*¹³⁹ in ground level air. An excess of Pb^{212} over Rn^{220} is expected at somewhat higher altitudes. Owing to their short half-lives, the concentrations of Rn^{220} and of any of its decay products become insignificant, when compared to Rn^{222} concentrations, at some ten metres above ground.

80. The concentrations of Pb^{210} , the long-lived decay product of Rn^{222} , and its radio-active products Bi^{210} and Po^{210} must be extremely low in the troposphere owing to removal by precipitation. Their concentrations, however, increase gradually in the upper troposphere, and actually exceed those of their short-lived parents in the stratosphere. Concentration ratios of Po^{210} to Pb^{210} depend critically on the mean removal rate and increase rapidly above 1 km. Observed $\text{Bi}^{210}/\text{Pb}^{210}$ and $\text{Po}^{210}/\text{Pb}^{210}$ ratios in air and precipitation have therefore been used to determine the mean residence time of natural aerosols in the troposphere.^{37, 140-145}

Horizontal distribution and variability

81. Local concentrations of the radon isotopes and their radio-active daughter products depend not only on the emanation rate of radon gases from the soil but also on a variety of local meteorological conditions. Wind direction is of special importance in coastal sites, on the sea and in the polar regions, as radon is mainly carried by continental air masses.

82. Winds reduce Rn^{222} concentration in the upper soil layers, thus enhancing the exhalation rate. All air motions near the earth-atmosphere interphase are turbulent; they assist to some extent in the extraction of radon gases from the upper soil air. These air motions,

on the other hand, may enhance the removal rate of radio-active daughters formed in the lower atmosphere by increasing their impact on the ground. They are also removed by precipitation and dry settling of aerosols.^{37, 134-137, 145}

83. Diurnal, seasonal or more irregular variations of the radio-activity at a given site have been reported.^{135, 140-151} During night hours, a temperature inversion usually develops near the ground, which reduces the vertical mixing of the radon gases after their exhalation from the ground, thus producing a build-up of radio-activity which reaches its maximum concentrations in the early morning hours. The rising temperature during day-time will break this temperature inversion and facilitate turbulent mixing, thus reducing radio-activity concentrations near the ground. Minimum concentration values are usually found during the afternoon. Diurnal variations may cause concentrations to change by a factor of two to three on the average, but variations exceeding a factor of ten have been reported.

84. In some locations, the average radon concentrations in the air during winter and early spring are lower than those prevailing during summer.^{36, 97, 151, 152} This may be due to the known seasonal variations in escape of radon from the ground, the relatively low winter and spring values being attributable to the higher moisture content of the soil. Frozen ground reduces escape considerably, and thick layers of snow or ice stop it completely. In other locations, the maximum radon concentrations are observed during autumn and winter and minimum ones in spring and summer, in accordance with variations of atmospheric conditions, e.g. the rate of vertical turbulent exchange.^{139, 153, 154}

Measured concentrations of natural radio-activity

85. The release of Rn²²² and Rn²²⁰ is considerably greater from land than over water. Typical measured levels in ground level air over continents range from 30 to 300 pCi Rn²²²/m³ and 0.5 to 10 pCi Pb²¹²/m³;^{146-150, 162-157} over the oceans, the concentrations are between one and two orders of magnitude lower.^{37, 131, 158, 159} Concentrations of the order of 2 pCi/m³ for Rn²²² and less than 0.3 pCi/m³ for Pb²¹² were measured in Antarctica.^{160, 161}

86. Concentrations of Rn²²² and Rn²²⁰ in ground level air in various regions were summarized in table IX of annex E of the 1962 report, and in some recent publications.^{36, 37, 135} Only a few determinations for Rn²²⁰ have been reported, based on the beta decay of Pb²¹². The erroneous assumption of radio-active equilibrium of the Pb²¹² with the short-lived Rn²²⁰ in the air sampled is partly responsible for the low Rn²²⁰/Rn²²² ratios reported.^{37, 131, 162, 163}

87. Radon concentrations indoors are generally higher than outdoor concentrations, depending on the construction materials, the degree of ventilation and the escape from indoor water sources. Average values of Rn²²² concentrations indoors at various locations were reviewed in annex E of the 1962 report and summarized there in table XI.

88. On theoretical grounds, it can be expected that the concentration of radon in air in deep layers of undisturbed soils can be of the order of 10⁵ pCi/m³ air;^{37, 131} therefore, such radon concentrations are to be expected in the air of caves and unventilated underground mines. Measurements made in mines, particularly uranium or thorium mines, give results consistent with this expectation.¹⁷⁰⁻¹⁷³

Long-lived decay products of radon-222

89. The general features of the circulations of Pb²¹⁰ and Po²¹⁰ resemble the patterns of stratospheric fall-out of artificial nuclear debris. The concentrations of Pb²¹⁰ in ground level air are dependent on latitude and on the distribution of land masses and oceans. Lower values were found in tropical and polar regions than in temperate areas.¹⁶⁴ Concentrations in the southern hemisphere are lower than those in the northern hemisphere, because the land surfaces from which radon is exhaled are smaller in the southern hemisphere. The observed range of the average continental Pb²¹⁰ concentrations vary from 0.002 to 0.016 pCi/m³ air (excluding Antarctica). Data reported by other authors for selected locations were of the same order of magnitude.^{140, 142-144, 146, 165-169} Only a small fraction of the Po²¹⁰ in equilibrium with Pb²¹⁰ is present in the lower troposphere.

90. Yearly deposition rates of 1.7 mCi Pb²¹⁰/km² were measured for the United Kingdom,¹⁴⁰ and 2.4 mCi Pb²¹⁰/km² were reported from France.¹⁶⁶ Under equilibrium conditions, Pb²¹⁰ and Po²¹⁰ activity in upper soil layers could thus amount to some 60 mCi/km² each.

IV. The transfer of environmental radio-activity to man

INTRODUCTION

91. Naturally-occurring radio-active nuclides enter the body mainly through food and water, inhalation being of secondary importance. The information on K⁴⁰, C¹⁴ and tritium transport through the food chain has remained unaltered since the 1962 report was issued; this subject, therefore, will not be discussed again here. Rb⁸⁷ accompanies potassium in metabolic processes, and doses to man due to this element, which is commonly distributed in the biosphere, will be dealt with in paragraph 136.

92. As was pointed out earlier, the radio-active elements of the uranium series in soil are seldom in equilibrium because of their different chemical properties which determine the varying behaviour of these elements in the processes of weathering, soil formation, redistribution of minerals in the lithosphere, etc. Uranium is taken up by plants; however, because of its low specific activity and the low concentrations found in the biosphere, its radio-activity may be disregarded. Thorium is absorbed very poorly by the root systems.¹⁷⁴⁻¹⁷⁷ Therefore, only radium isotopes (Ra²²⁶, Ra²²⁸), and possibly lead (Pb²¹⁰) and polonium (Po²¹⁰), among the long-lived nuclides of both the uranium and thorium series are taken up by plants to a degree that may be important when further transfer of radio-activity to animals and to man is considered. On the other hand, significant amounts of Po²¹⁰ and Pb²¹⁰ are deposited on plants from the atmosphere.

93. Ra²²⁶ and Ra²²⁸ decay, directly or indirectly, to Rn²²² and Rn²²⁰, respectively. The latter are chemically inactive gases. They leave the soil and, together with their short- and long-lived daughter products, form the most significant part of the natural radio-activity that is inhaled by man.

94. *Areas of "normal" activity.* As reported in the 1962 report, the content of Ra²²⁶ in soil varies considerably with geological and geochemical conditions. The levels reported from the United States, which are possibly typical,¹⁷⁸ are of the order of 0.09 to 0.8 pCi/g soil. No extensive measurements of Ra²²⁶, or Th²³² have recently been reported. Because of the short half-

life of Ra²²⁸, it may be assumed that its concentration in soils is closely related to that of its parent, Th²³². From the data on the escape of radon from soil and on deposition of Pb²¹⁰ from the atmosphere it has been postulated that equilibrium levels of this nuclide in the few upper inches of soil should be about 60 mCi/km² (paragraph 90).

95. *Areas of high natural radio-activity.* Those areas in the State of Kerala in India, and the States of Rio de Janeiro and Espirito Santo in Brazil, where levels of radio-activity in soil are high because of the presence of monazite sands of high thorium content, were discussed in the 1962 report. Since then, additional data have been reported on the availability of radio-active elements from these soils to plants^{177, 179, 180} and will be discussed in paragraphs 110 and 111, together with the data on accessibility to plants of Ra²²⁶ and Ra²²⁸ in the high background area of volcanic intrusives in the State of Minas Gerais, Brazil.

INTAKE BY MAN

96. In addition to K⁴⁰, Rb⁸⁷ and C¹⁴, which are not considered in this section, radium (Ra²²⁶ and Ra²²⁸), radon (Rn²²²), radio-lead (Pb²¹⁰) and polonium (Po²¹⁰) contribute appreciable doses to man through food. When inhaled, radon (Rn²²² and Rn²²⁰) with their short-lived decay products, and possibly radio-lead (Pb²¹⁰), contribute to lung doses and must also be considered.

Water

97. The relative importance of drinking water as a source of natural radio-activity in human diet was studied by numerous investigators.^{117, 120, 123, 129, 165, 180-182}

98. Sanitary (usually chemical and physico-chemical) treatment of water, as commonly applied in a majority of urban centres in a number of countries, may remove large proportions—up to 90 per cent—of Ra²²⁶ and Pb²¹⁰.^{117, 122} Water-softening procedures seem to be especially effective in this respect. Chemical treatment (e.g chlorination and subsequent aeration) and storage may reduce the radon level in tap water, and boiling may further remove radon from the water which is actually consumed.

Typical levels of natural radio-nuclides in water

99. Water consumption contributes only a small percentage to the total intake of the nuclide (of the order of a few to 10 per cent—table X). As indicated in the 1962 report, the waters consumed by the majority of the world population usually contain less than 0.1 pCi/litre Ra²²⁶.

100. Typical concentrations of Rn²²² in waters derived from lakes, rivers and shallow wells, which are consumed by the majority of the world's population, are indicated in table VIII.

101. The contribution of drinking water to the total intake of Pb²¹⁰ and Po²¹⁰ has been estimated to be of the order of 1-10 per cent for the typical diet in the United States.¹⁶⁵

Areas with high concentrations of radium and radon in drinking water

102. In some areas, where Ra²²⁶ concentrations in water of several pCi/litre or even more are not uncommon, water may be a significant or even a dominant contributor to the total intake of Ra²²⁶. Thus, in rather

exceptional situations, involving populations relatively limited in size (less than 1 per cent of the population of the country as a whole in the cases investigated so far), the total intake of Ra²²⁶ may be twice as high as the average for the country (Cornwall in the United Kingdom),¹²³ or even higher by an order of magnitude (some regions of the midwestern States of the United States).^{122, 182, 183} However, in the monazite area in the State of Kerala, India, where elevated intakes of radium were noted, the relative role of water as a contributor of radium was limited to about 10 per cent, because of higher concentrations of Ra²²⁶ both in water and foodstuffs of plant origin from local sources.¹⁸⁰

103. Consumption of water with high concentrations of dissolved radon (deep wells in some areas, and spa waters which show concentrations of about 20 to 200 nCi/litre, with the highest known value of 5,000 nCi/litre)¹²⁰ is rather exceptional. In some areas, such as those found in the States of Maine and New Hampshire in the United States,¹⁸⁴ well waters supplying a population of the order of 100,000¹³⁰ contain radon at average concentrations of about 16 and 30 nCi/litre. Average concentrations of 1 to a few nCi/litre were found in central Sweden¹²⁹ (population of about 300,000), Cornwall¹²³ (population of about 300,000), the Federal Republic of Germany and elsewhere (see also 1962 report). Many factors discussed in paragraph 98 reduce the intake considerably. The possible significance of doses from radon taken in with water will be discussed in paragraph 148.

Foodstuffs

Areas of typical "normal" levels of intake

104. The total intake of Ra²²⁶ and the contribution of different food categories was studied in the United Kingdom,^{181, 185} the United States,¹⁸⁰⁻¹⁸⁰ and India.¹⁸⁰ The average estimates reported for areas with apparently normal concentrations of Ra²²⁶ in drinking water and foodstuffs vary between 0.7 and 5 pCi/day (table X), while smaller variations in the values of the Ra/Ca ratio have been reported.

105. In both the United States¹⁸⁷ and the United Kingdom^{181, 185} the main sources of radium in the diet are not the same as those from which the main proportion of calcium is derived (table X). In all the circumstances investigated so far, foodstuffs of plant origin (cereals and grain products, fruits and vegetables) form a major source of dietary Ra²²⁶. The relative availability for absorption in the gastro-intestinal tract of radium from water and different foodstuffs is unknown.

106. The daily intake in very low-income groups in Puerto Rico¹⁹⁰ is slightly less than 1 pCi/day. However, the calcium intake is also low, and thus the resulting Ra²²⁶/Ca ratio in the diet is similar to that in other regions of the United States. It could be inferred from the data of Chhabra¹⁸⁰ that a similar picture may obtain in India—at least for the area investigated—where the intakes of both radium and calcium seem to be low.²

107. The intake of Ra²²⁸ has not been studied on a scale comparable to that of Ra²²⁶, but the data derived from alpha-spectrometric measurements in different materials, including water, foodstuffs and human tissues,^{191, 192} indicate that the intake of Ra²²⁸ is, on the average, about two to four times less than that of Ra²²⁶.

108. The Pb²¹⁰ intake has not been specifically determined, and only some indirect estimates are avail-

able¹⁶⁵ which show that, on the average, the total intake per day should be of the order of 1-10 pCi. This seems to be typical for most regions, with the exceptions discussed in paragraph 113. The contributions of different foodstuffs are not known, but those of plant origin probably play the dominant role.

109. The estimated usual intake of Po^{210} is of the order of several pCi per day,^{193, 194} in agreement with the results of total dietary analyses.¹⁸⁹ The data are consistent with a faecal excretion of 1.8 pCi/day as determined by Holtzman¹⁶⁵ or of 1.7-6.4 pCi/day as found by Hill.¹⁹³ The data indicate that the main sources are fresh leafy vegetables and, in some circumstances, a few animal tissues (kidney, liver).

Special food chain mechanisms and areas of high natural radio-activity

110. Information on the intake of natural radio-elements in areas of high radio-activity in the soil is very limited. In Brazil, locally grown foodstuffs with higher than normal radio-activity are found in some localities near the towns of Araxa and Tapira, in the State of Minas Gerais.¹⁷⁰ In these areas, the concentrations of Ra^{226} and Ra^{228} in edible plants are about one to two orders of magnitude higher than normal. However, estimates of intake in this area are not yet available. Food from the monazite areas of Brazil contains only the normal amount of radio-activity from both the uranium and the thorium series. This seems to be due to the very low solubility of the minerals of which the monazite sands are composed.

111. In India, in the high activity area of Kerala State, recent studies¹⁸⁰ have revealed that the radium intake due to locally grown plant foodstuffs may be higher by a factor of four to five than in the Bombay area, and is still considered within the range of values typical of other countries.

112. Brazil nuts are known to accumulate the heavier alkaline earth elements, radium and barium, to an unusual extent. This effect has been observed in samples from Brazil, Malaya and Guyana;^{181, 195, 196} concentrations in kernels usually range from one to five pCi Ra^{226} /gramme. Thus the quantity of Ra^{226} in one gramme of Brazil nuts is comparable to the estimated average daily intake of the population shown in table X. It is not known whether the radium from Brazil nuts is absorbed in the gastro-intestinal tract of man to the same extent as radium in other foods.

113. In the 1964 report (annex A, paragraphs 118, 128), a special food chain mechanism, transferring unusually high activities of Cs^{137} from lichens through reindeer or caribou meat to man, was discussed. The same applies to Pb^{210} and Po^{210} of natural origin, and fragmentary data are available (table XV), showing high concentrations of both Pb^{210} and Po^{210} in lichens and in the meat of reindeer and caribou, which graze in arctic regions.^{193, 197} It was postulated that the dietary Pb^{210} and Po^{210} intakes of people subsisting on large amounts of caribou or reindeer meat may be elevated by an order of magnitude above the typical values observed elsewhere.¹⁹⁷ This is supported by still fragmentary evidence of Pb^{210} and Po^{210} in human tissues from northern Canada¹⁹⁸ and Finnish Lapland¹⁹⁹ (paragraph 133).

Air

114. *Normal concentrations.* The normal concentrations of radon and thoron in ground level atmospheric air have been discussed in paragraphs 79, 85 and 86.

The usual levels to which human populations are exposed, however, are heavily influenced by indoor concentrations. Indoor concentrations are normally higher than those found outside because of the accumulation of radon and thoron from building materials in closed or poorly ventilated premises. The problem was reviewed in the 1962 report, and no recent data are available which would warrant a change in the position adopted therein. It will be maintained, therefore, that the typical average effective concentration of Rn^{222} to which the world population is exposed is about 500 pCi/m³. Daughter products, from Po^{218} through Bi^{214} , are assumed to be present in concentrations between 100 and 500 pCi/m³. Corresponding average concentrations of thoron daughter products (Po^{216} , Pb^{212} and Po^{212}) are based on an estimate by Jacobi²⁰⁰ and have been assumed to equal about 600, 10 and 10 pCi/m³, respectively.

115. *High concentrations.* It needs to be mentioned here that, because of their occupation in underground mines, some groups of workers are exposed to air concentrations of Rn^{222} and daughter products higher than usual (paragraph 88). The fraction of the population subjected to this type of exposure, even in industrialized countries, does not exceed a fraction of 1 per cent. The importance of the problem and the health hazards involved were discussed in the 1964 report (annex B, paragraphs 152-174) and elsewhere.²⁰¹

116. The downward flow of cold air gives rise to increased concentrations of radon at the bottom of valleys. Rn^{222} values ten times higher than the night concentrations over flat country have been observed in valleys by Servant.²⁰²

LEVELS IN MAN

117. Our knowledge of concentrations of naturally-occurring radio-active potassium (K^{40}), carbon (C^{14}), tritium (H^3) and beryllium (Be^7) in human tissues and of their distribution in different organs has remained essentially unchanged since the 1962 report was issued, but substantial new information has become available on radium isotopes and nuclides of the Pb^{210} - Bi^{210} - Po^{210} chain.

Metabolic data on Ra^{226} and the Pb^{210} - Bi^{210} - Po^{210} chain

118. Several investigators have shown recently^{203, 204} that about 80-85 per cent of radium in the body is contained in the skeleton, the remaining fraction being distributed approximately uniformly in soft tissue as shown in table XIII. The concentration of radium per gramme of ash from soft tissues is similar to that in the skeleton, but is lower by a factor of about twenty when calculated on a fresh weight basis.²⁰³

119. The concentration of Ra^{226} per gramme of body ash of man is constant throughout the entire life span, starting from the earliest period of foetal life (four months) in which the determinations were possible.²⁰⁵ The rise of body calcium and radium in the process of growth is apparently parallel, and no significant discrimination between the two elements seems to take place at the placental barrier. The total body burden becomes constant in adult age and displays the organ distribution described in the foregoing paragraph.

120. Under steady-state conditions the ratio of radium to calcium in the body contrasts markedly with that in diet. The observed ratio (OR), defined as

$$\frac{\text{Ra/Ca in bone}}{\text{Ra/Ca in diet}}$$

for adults is about 0.016.^{186, 190} This compares with ratios of about 0.25 for strontium and 0.08 for barium.²⁰⁷ The consistency of the OR for radium under different conditions of dietary composition suggests that the content in bone is determined by the dietary ratio of the two elements.¹⁹⁰

121. The content of Pb^{210} in the human body has been the subject of several studies in recent years; the results of measurements are collected in tables XII and XIII. There is apparently little correlation of Pb^{210} with the content of Ra^{226} in bone from the same places.

122. Most of the total Pb^{210} present in the body is in the skeleton.^{165, 199} The distribution of Pb^{210} in the skeleton appears to be uniform within a factor of two, with slightly but significantly higher concentrations in trabecular than in compact bone. Concentrations in the bone tissue of males are somewhat (about one-third) higher than in that of females.¹⁶⁵

123. The contributions of different sources of Pb^{210} to the total body burden are not known in detail. However, inhaled air and dietary intake seem to be the main contributors. The decay of radon in the body (both the atmospheric radon and that formed *in situ* from the disintegration of Ra^{226}) and the intake of Pb^{210} with water seem to account for only a few per cent of the activity present in the human body.¹⁶⁵

124. The distribution of Pb^{210} in human soft tissues is not known in detail. The ratio of Po^{210} to Pb^{210} is less than unity in most of them with values close to equilibrium in the liver, and with some excess Po^{210} in the kidneys and ovaries.¹⁹³ The origin of this excess Po^{210} is still open to question. However, as pointed out by Holtzman,²⁰⁸ there are kinetical reasons for supposing that by far the largest fraction of Po^{210} in the body, including this so-called "unsupported Po^{210} ", comes mainly from the redistribution of that Po^{210} that is formed by radio-active decay of Pb^{210} in the body rather than from direct dietary intake.

125. Hunt *et al.*²⁰⁹ showed that, despite marked differences between individual teeth and observable variations between trabecular and compact bone in the same person, the average levels of Ra^{226} and Pb^{210} in bone were closely correlated with those in teeth. The average concentrations of radium in bone and teeth were equal, whereas those for Pb^{210} were twice as high in bone. It may be concluded, therefore, that teeth analyses for Ra^{226} may be used to study average population levels in the skeleton when bone material is not available and when no significant and prolonged changes in the rate of intake are expected. On the other hand, as the length of the physical half-life of Pb^{210} ($T_{1/2} = 19.4$ years) is comparable with the human life span, and the mineral turnover of the skeleton may significantly differ from that of the teeth, the Pb^{210} concentration in teeth as a measure of skeletal levels demands further study.

Normal levels of natural radio-activity in man

126. Ra^{226} has repeatedly been determined in human tissues by several investigators. The results published since 1962 are collected in tables XI and XIII.

127. The Ra^{226} values as reported in table XI indicate that bone levels in most areas of normal dietary intake of this nuclide are similar, the most frequent values clustering around $1-1.5 \times 10^{-2}$ pCi/g ash, corresponding to an average of about 30 to 40 pCi Ra^{226} in the skeleton of a standard man (assuming an average of 2.800 grammes of ash in the skeleton). This value is substantially lower than the value of 60 pCi accepted

in the 1962 report. It seems appropriate therefore to suggest, for the purpose of dose-rate calculation, a value of 30 pCi Ra^{226} as typical of the whole skeleton burden in areas of normal rates of intake.

128. Ra^{228} is the only important long-lived element of the thorium series that is absorbed to a significant degree from the gastro-intestinal tract. Ra^{228} decays through Ac^{228} to Th^{228} ($T_{1/2} = 1.9$ years), which in turn decays through a series of short-lived nuclides to the stable isotope Pb^{208} . Very small fractions of the amounts of radio-active decay products of Th^{228} found in bone leave the skeletal deposition site of Ra^{228} and Th^{228} .²¹⁰ The latter nuclide has recently been determined in bone, together with Ra^{226} . In two studies, the average ratio between activities of Th^{228} and Ra^{226} in bone ash varied between 0.25¹⁸³ and 0.4.²⁰³ These values are consistent with results reported earlier,²¹¹ and it seems appropriate to accept that the value of 0.3 to 0.4 is closer to the real situation than the figure of 0.7, which was assumed in the 1962 report. Thus, a natural skeletal burden of 10 pCi Th^{228} in a standard man seems typical.

129. From table XII it follows that the typical Pb^{210} burden in the skeleton of an adult is about 270 pCi. As only a small fraction of Po^{210} formed by the radio-active decay of Pb^{210} and Bi^{210} in bone is removed from its site of formation, the activities of all three nuclides in the skeleton are close to radio-active equilibrium.^{165, 203, 212-214} It seems that, in contrast to the 1962 report where a ratio of Po^{210}/Pb^{210} of 0.5 was assumed, a value of about 0.9 is closer to reality.

Areas of high natural radio-activity and special food chain mechanisms

130. In some areas of the midwestern States of the United States, where concentrations of Ra^{226} in drinking water of about 1 to 10 pCi/litre are not uncommon, Ra^{226} levels in bone are three to four times higher than those typical elsewhere (table XI). The population of the area concerned is about 1,000,000.¹²²

131. Preliminary data²¹⁵ from those areas of high natural radio-activity in Brazil where high activities of radium in plants of local origin were found (towns of Araxa and Tapira, State of Minas Gerais) show that the concentrations of Ra^{226} in teeth collected from the inhabitants are higher than in "normal" areas but only by a factor of two to three (table XIV). In view of the fact that a significant correlation exists between concentrations of Ra^{226} in bones and teeth²⁰⁹ (paragraph 125), these observations suggest that the skeletal levels of Ra^{226} in this area of Brazil might be elevated by a similar factor. The population living in the areas of Araxa and Tapira is about 15,000.

132. The data from the monazite area in Kerala State in India are even more scanty. As shown in table XI, in five samples of human bone analyzed so far,^{180, 216} the concentrations of radium were on the average higher than those typical for "normal" areas by one order of magnitude. The population residing in the monazite area of Kerala State amounts to about 80,000 people.

133. As discussed in paragraph 113, high concentrations of Pb^{210} and Po^{210} in lichens and in caribou and reindeer meat were found in arctic regions. Fragmentary data available on Pb^{210} and Po^{210} show that human beings subsisting to a significant degree on caribou meat display levels of both nuclides that differ widely from those usually found in the northern temperate zone. Thus, Hill¹⁹⁸ was able to demonstrate that

the average concentration of Po^{210} in placentae of Eskimo women from northern Canada who consumed large amounts of caribou, reindeer or moose meat amounted to 59 pCi/kg fresh weight, whereas in women subsisting on a diet normal for England and Canada 3.3 and 5.0 pCi/kg were found. Kauranen and Mietinen¹⁹⁹ reported levels of Po^{210} in the blood of reindeer-breeding Lapps which were eight times higher than those in the blood of southern Finns. A similar trend was observed for Pb^{210} in Eskimo bones, as can be inferred from table XII. The size of the populations living in the arctic regions and subsisting on reindeer or caribou meat as one of their main dietary items is difficult to evaluate but may be as high as 1,000,000.

DOSES FROM INTERNAL IRRADIATION BY NATURAL RADIO-ACTIVE NUCLIDES

134. Dose rates in millirads per year from internal sources of natural radiation to the gonads, to cells lining Haversian canals, to osteocytes and to the bone marrow contained in trabecular bone have been assembled in table XVI. The per cent contributions of alpha radiation to the total dose rates are also indicated.

135. It is realized that for the purpose of comparing risks, doses in rads ought to be weighted by appropriate RBE values. In the case of alpha emitters, however, the information on RBE is so uncertain and the values proposed by various authors so widely different that it is reasonable to consider them as still unknown, although it is possible that they may be higher than one.⁶⁷ If this were so, and if unweighted dose rates from natural sources as given in table XVI were used as a standard for obtaining comparative risks from other, man-made, sources, the comparative risks would thus be over-estimated depending on the RBE values applying to any given effect.

136. The estimates of doses from K^{40} , C^{14} and Rn^{222} and short-lived daughter products in soft tissues other than the respiratory tract are the same as those given in the 1962 report. Other values have been recalculated on the basis of different estimates of tissue concentrations and/or by introducing some changes in the method of dose calculation. Doses to gonads from Rb^{87} amount to about 0.3 mrad/year; they are certainly less in osteocytes and in bone marrow.

137. The doses of alpha radiation to osteocytes and Haversian canals from Ra^{226} and daughter products, Ra^{228} and daughter products, and Po^{210} have been calculated by the method of Spiers.²¹⁷ The assumed diameters of the cavities were $5\ \mu$ for the osteocytic lacunae and $50\ \mu$ for the Haversian canals. The values of the geometrical factors for Ra^{226} were those calculated by Charlton and Cormack,²¹⁸ assuming one-third retention of Rn^{222} and its short-lived daughter products (through Po^{214}), while those for Ra^{228} and daughters in equilibrium and for Po^{210} were those given by Stahlhofen.²⁰³ Doses to bone from beta emission of all nuclides of these series were ignored because the percentage of energy delivered from beta decay in bone constitutes, under the conditions of equilibrium assumed, only about 2, 4 and 7 per cent of the total energy of the Ra^{226} , Ra^{228} and Pb^{210} chains, respectively.^b The dose of beta radiation to bone marrow from

^b Recent evidence indicates that it is the irradiation of the endosteal cells lining the inner surface of bone, rather than that of osteocytes or the cells of Haversian canals, which is of relevance to the induction of bone tumours. The doses delivered to endosteal cells will normally be less than those quoted in this report for osteocytes and will often also be less than those given for cells of the Haversian canals.

these series was also ignored as it amounted to approximately 0.1 millirad per year.

138. The alpha dose rates in fresh bone from individual nuclides that result from these calculations are significantly higher than those obtained in 1962. The doses from Ra^{226} and Ra^{228} differ only slightly from previous estimates, whereas those from Po^{210} are definitely higher, both because higher concentrations have now been assumed and because a different method of dose computation has been used.

139. In the present review, the dose rate to the marrow contained in trabecular bone from Po^{210} has been assessed on the basis of the presumed concentration of the nuclide in soft tissues (~ 3 pCi/kg fresh weight) rather than in the mineralized bone itself as was done in the 1962 report.

140. If the total annual dose estimates from internal sources presented in table XVI are compared with the dose estimates given in the 1962 report expressed in millirads, very little difference is seen.

141. At the time when the 1962 report was adopted, it was realized that the irradiation of the respiratory tract from natural sources exceeded that of any other organ of the human body. On the other hand, only approximate calculations of average doses to different volumes of tissues in the respiratory tract were possible. Since then, however, significant progress has been made in better understanding the mechanisms by which daughter products of Rn^{222} and Rn^{220} are deposited and transported in the respiratory tract of man.

142. Thus, Altshuler *et al.*¹⁷⁰ have calculated the doses of alpha radiation from Po^{218} and Po^{214} . These doses account for almost the whole of the dose to alveolar, bronchiolar, bronchial and tracheal epithelia, and, in estimating them, allowance was made for the distribution of radio-activity (Po^{218} , Pb^{214} , Po^{214}) between free ions and natural aerosols of varying particle size, for the deposition of ions and particles in different parts of the respiratory tract, for the upward transport of radio-activity in the bronchial tree with the flow of mucus, and for the physical and anatomical factors involved in the penetration of alpha particles through the mucus and the bronchial epithelium.

143. A similar study has been made by Jacobi²⁰⁰ for both Rn^{222} and daughter products, and Rn^{220} and daughter products by applying somewhat different physical and anatomical criteria. In both studies, it was assumed that the critical irradiated tissue was the basal layer of cells of the bronchial epithelium.

144. Both studies yielded similar results, pointing to the fact that the dose rates from Po^{218} and Po^{214} alpha particles are highest in the epithelium of segmental and lobar bronchi. With average concentrations of Po^{218} , Po^{214} and Bi^{214} , each between 100 and 500 pCi/m³, the results of both studies indicate that the dose rate in these parts of the respiratory system is of the order of several hundreds of millirads per year, the doses to alveolar tissues and to bronchioli being lower by two and to the trachea by one order of magnitude. At average concentrations of Po^{216} , Pb^{212} and Bi^{212} of 600, 10 and 10 pCi/m³, respectively, as are assumed for the purpose of the present annex, the irradiation from these nuclides adds only another few per cent to the values given above for doses from Po^{218} and Po^{214} .²⁰⁰

145. The quantitative agreement between the results of the two studies may be somewhat fortuitous, and further studies are necessary to provide sounder infor-

mation on several of the critical parameters, especially those of a physiological and anatomical nature.^{219, 220} It must be mentioned that higher doses may be received if some uptake and retention of radio-activity by the tissues of the respiratory tract is assumed, but such a possibility is neglected at present for lack of relevant information.¹⁷⁰

146. On the other hand, rapid dissociation of Po²¹⁸, Pb²¹⁴ and Bi²¹⁴ atoms (or ions) from aerosol particles deposited in the respiratory tract with subsequent direct transfer to the blood stream and other organs could reduce the dose significantly. That such a possibility exists was demonstrated by Pohl.²²¹ Even so, however, it is still uncertain whether the demonstrated resorption of radon decay products takes place predominantly from the alveolar regions or from the bronchial tree, which would be more significant from the dosimetric point of view. In any case, the doses to the critical cells in some areas of bronchial epithelium thus obtained seem higher by at least one order of magnitude than those to the whole respiratory organ that were accepted in the 1962 report.

147. An additional exposure to natural polonium may result from cigarette smoking.^{193, 222-228} Average alpha-radiation doses to the respiratory tract of smokers from excess Po²¹⁰ deposition is not expected to exceed about 1 mrad/year. In some areas of bronchial epithelium where concentrations up to 0.3 pCi/gramme have been detected,²²⁸ the alpha dose due to Po²¹⁰ might reach the level of some tens of millirads per year. The biological significance of this irradiation is

unknown, but it appears most unlikely to be appreciable in view of the low doses involved.

148. The metabolism of radon ingested with water and the doses attributable to this source of radio-activity were studied by von Döbeln and Lindell¹²⁹ and by Hursh *et al.*¹³⁰ The dose per unit of activity ingested seems highest in the stomach (~20 mrad/ μ Ci Rn²²²), those to other organs being lower by two orders of magnitude. At normal or typical concentrations of radon in drinking water of surface origin, which are of the order of a few picocuries per litre,¹²³ the doses are negligible. In those areas where concentration of Rn²²² in drinking water is of the order of nanocuries per litre, the doses to the stomach could be correspondingly higher—of the order of a few millirads per year.

V. Recapitulation of dose rates

149. Estimates of dose rates to man from natural sources are summarized in table XVII. For comparison, the estimates given in the 1962 report but expressed in millirads per year are also included. The differences between the two sets of estimates are slight. The dose rates to cells lining Haversian canals, however, include a larger contribution from alpha particles, whereas those to the gonads and bone marrow include a smaller contribution than the dose rates obtained in 1962. Dose rates to the lung tissues are not given in the table since no exact estimate is available, but those to the epithelium of segmental and lobar bronchi are believed to be of the order of several hundreds of millirads per year (paragraph 144).

TABLE I. PROPERTIES OF THE MAIN SECONDARY PARTICLES IN COSMIC RADIATION

Particle	Electric charge	Rest mass		Mean lifetime seconds	Decay products
		MeV	Electron masses		
n	0	940	1,839	10 ⁸	e ⁻ + ν + p
p	+1	938	1,836	Stable	—
π^{\pm}	± 1	140	273	2.5 $\times 10^{-8}$	μ^{\pm} + ν
π^0	0	135	264	2 $\times 10^{-16}$	2 γ
μ^{\pm}	± 1	106	207	2.2 $\times 10^{-6}$	e ^{\pm} + 2 ν
e ^{\pm}	± 1	0.511	1	Stable*	—

* A positron (e⁺) is annihilated by combining with an electron, and two photons are emitted.

TABLE II. SOME RADIO-NUCLIDES PRODUCED BY COSMIC RAYS³¹⁻⁴⁰

Radio-nuclide	Half-life	Maximum energy of beta radiation in keV	Main mode of formation	Calculated atmospheric production rate (atoms/cm ² \times year)	Calculated concentration in the lower troposphere (pCi/m ³)
H-3	12.3y	18	Spallation of N ¹⁴ or O ¹⁶	8 $\times 10^6$	5 $\times 10^{-2}$
Be-7	53d	Electron capture	Spallation of N ¹⁴ or O ¹⁶	2.5 $\times 10^6$	0.5
Be-10	2.7 $\times 10^6$ y	550	Spallation of N ¹⁴ or O ¹⁶	1.4-2.6 $\times 10^6$	5 $\times 10^{-8}$
C-14	5,760y	165	N ¹⁴ (n, p) C ¹⁴	50-65 $\times 10^6$	1.3-1.6
Na-22	2.6y	540	Spallation of A ⁴⁰	1.8 $\times 10^3$	5 $\times 10^{-5}$
Si-32	700y	100	Spallation of A ⁴⁰	5-6 $\times 10^3$	8 $\times 10^{-7}$

TABLE II. SOME RADIO-NUCLIDES PRODUCED BY COSMIC RAYS³¹⁻⁴⁰ (continued)

Radio-nuclide	Half-life	Maximum energy of beta radiation in keV	Main mode of formation	Calculated atmospheric production rate (atoms/cm ² × year)	Calculated concentration in the lower troposphere (pCi/m ³)
P-32	14.3d	1,720	Spallation of A ⁴⁰	2.5 × 10 ⁴	1.1 × 10 ⁻²
P-33	25d	250	Spallation of A ⁴⁰	2.1 × 10 ⁴	6 × 10 ⁻³
S-35	87d	165	Spallation of A ⁴⁰	4.4 × 10 ⁴	6 × 10 ⁻³
Cl-36	3.1 × 10 ⁵ y	710	Spallation of A ⁴⁰	3.5 × 10 ⁴	1.2 × 10 ⁻⁸

TABLE III. THE IONIZING COMPONENT OF COSMIC RAYS AT SEA LEVEL

Author	Year	Ion pairs/cm ² × second (STP)	mrads/year
UNSCEAR ²	1962	1.90 - 1.96	28
Shamos and Liboff ⁶⁵	1965	2.2 ± 0.06	28.5 ± 0.8
Herbst ⁶⁴	1963		28
Kastner ⁶⁶	1965	2.2 ± 0.1	29 ± 1.3
Lowder and Beck ¹⁸	1965	2.1 ± 0.1	27.6 ± 1.3
Lillicrap ¹⁹	1965	*	26 ± 1.5

* Energy absorbed in a water Čerenkov detector.

TABLE IV. URANIUM SERIES^a

Isotope	Atomic number	Historical name	Half-life	Alpha and/or beta ray energies (MeV) ^b	Gamma ray energies (MeV) ^b
U-238	92	Uranium I	4.5 × 10 ⁹ y	α4.18(77), 4.13(23)	
Th-234	90	Uranium X ₁	24.1 d	β0.19(65), 0.10(35)	0.09(15), 0.06(7), 0.03(7)
Pa-234	91	Uranium X ₂	1.18 min	β2.31(93), 1.45(6), 0.55(1)	1.01(2), 0.77(1), 0.04(3)
U-234	92	Uranium II	2.50 × 10 ⁶ y	α4.77(72), 4.72(28)	0.05(28)
Th-230	90	Ionium	8.0 × 10 ⁴ y	α4.68(76), 4.62(24)	
Ra-226	88	Radium	1,622 y	α4.78(94), 4.59(6)	0.19(4)
Rn-222	86	Radon	3.82 d	α5.48(100)	
Po-218	84	Radium A ^c	3.05 min	α6.00(100)	
Pb-214	82	Radium B ^c	26.8 min	β1.03(6), 0.66(40), 0.46(50), 0.40(4)	0.35(44), 0.29(24), 0.24(11), 0.05(2)
Bi-214	83	Radium C ^c	19.7 min	β3.18(15), 2.56(4), 1.79(8), 1.33(33), 1.03(22), 0.74(20)	2.43(2), 2.20(6), 2.12(1), 1.85(3), 1.76(19), 1.73(2), 1.51(3), 1.42(4), 1.38(7), 1.28(2), 1.24(7), 1.16(2), 1.12(20), 0.94(5), 0.81(2), 0.77(7), 0.61(45)
Po-214	84	Radium C ^c	160 × 10 ⁻⁶ sec	α7.68(100)	
Pb-210	82	Radium D ^c	19.4 y	β0.06(17), 0.02(83)	0.05(4)
Bi-210	83	Radium E ^c	5.0 d	β1.16(100)	
Po-210	84	Radium F	138.4 d	α5.30(100)	
Pb-206	82	Radium G	Stable		

^a Based on reference 230.

^b Figures in parentheses indicate per cent yield per disintegration.

^c Parallel decay branches of less than 1 per cent are not listed.

TABLE V. THORIUM SERIES^a

Isotope	Atomic number	Historical name	Half-life	Alpha and/or beta ray energies (MeV) ^b	Gamma ray energies (MeV) ^b
Th-232 ...	90	Thorium	1.41×10^{10} y	α 4.01(76), 3.95(24)	0.06(24)
Ra-228 ...	88	Mesothorium I	5.8 y ^c	β 0.05(100)	
Ac-228 ...	89	Mesothorium II	6.13 h	β 2.18(10), 1.85(9), 1.72(7), 1.13(53), 0.64(8), 0.45(13)	1.64(13), 1.59(12), 1.10, 1.04, 0.97(18), 0.91(25), 0.46(3), 0.41(2), 0.34(11), 0.23, 0.18(3), 0.13(6), 0.11, 0.10, 0.08
Th-228 ..	90	Radio-thorium	1.91 y	α 5.42(72), 5.34(28)	0.08(2)
Ra-224 ...	88	Thorium X	3.64 d	α 5.68(95), 5.45(5)	0.24(5)
Rn-220 ...	86	Thoron	54.5 sec	α 6.28(99 +)	
Po-216 ...	84	Thorium A ^d	0.158 sec	α 6.78(100)	
Pb-212 ...	82	Thorium B	10.64 h	β 0.58(14), 0.34(80), 0.16(6)	0.30(5), 0.24(82), 0.18(1), 0.12(2)
Bi-212 ...	83	Thorium C	60.5 min	α (35%) 6.09(10), 6.04(25) β (65%) 2.25(56), 1.52(4), 0.74(1), 0.63(2)	(35%) 0.04(1), (65%) 2.20(2), 1.81(1), 1.61(3), 1.34(2), 1.04(2), 0.83(8), 0.73(10)
Po-212 ...	84	Thorium C'	0.30×10^{-6} sec	α 8.78(100)	
Tl-208 ...	81	Thorium C''	3.1 min	β 2.37(2), 1.79(47), 1.52, 1.25	2.62(100), 0.86(14), 0.76(2), 0.58(83), 0.51(25), 0.28(9), 0.25(2)
Pb-208 ...	82	Thorium D	Stable		

^a Based on reference 230, except where otherwise indicated.

^b Figures in parentheses indicate per cent yield per disintegration.

^c From reference 231.

^d Parallel decay branches of less than 1 per cent are not listed.

TABLE VI. ACTINIUM SERIES^a

Isotope	Atomic number	Historical name	Half-life	Alpha and/or beta ray energies (MeV) ^b	Gamma ray energies (MeV) ^b
U-235	92	Actinouranium	7.13×10^8 y	α 4.59(5), 4.55(4), 4.50(1), 4.41(4), 4.39(57), 4.36(18), 4.32(3), 4.21(6)	0.204(6), 0.185(54), 0.164(5), 0.143(11), 0.110(3)
Th-231 ...	90	Uranium Y	25.64 hr	β 0.30(45), 0.22(20), 0.13(20), 0.09(15)	0.095(2), 0.084(7), 0.026(12)
Pa-231 ...	91	Protoactinium	3.47×10^4 y	α 5.05(11), 5.02(23), 5.00(25), 4.97(2), 4.94(23), 4.84(1), 4.72(10), 4.68(3)	0.33(1), 0.30(2), 0.10(2), 0.06(13), 0.04(15), 0.029(90), 0.025(11), 0.02(4), 0.0165(20)
Ac-227 ...	89	Actinium	21.6 y	α (1.2%) 4.94(1), others (weak) β (98.8%) 0.046(100)	
Fr-223 ...	87	Actinium K	21 min	β 1.15(100)	0.08(24), 0.05(40)
Th-227 ...	90	Radio-actinium	18.17 d	α 6.04(23), 6.01(3), 5.98(24), 5.96(4), 5.87(3), 5.76(21), 5.70(19), 5.67(2)	0.33(7), 0.31(4), 0.30(5), 0.29(2), 0.28(2), 0.26(7), 0.24(13), 0.17(1), 0.11(4), 0.10(1), 0.08(5), 0.06(9), 0.05(16), 0.03(39)
Ra-223 ...	88	Actinium X	11.68 d	α 5.87(1), 5.75(10), 5.71(52), 5.61(25), 5.54(9), 5.50(1), 5.43(2)	0.45(1), 0.34(7), 0.27(13), 0.15(11)
Rn-219 ...	86	Actinon ^c	3.92 sec	α 6.81(80), 6.54(13), 6.42(7)	0.40(5), 0.27(9)

TABLE VI. ACTINIUM SERIES^a (continued)

Isotope	Atomic number	Historical name	Half-life	Alpha and/or beta ray energies (MeV) ^b	Gamma ray energies (MeV) ^b
Po-215 ...	84	Actinium A	1.83×10^{-8} sec	$\alpha 7.37(100)$	
Pb-211 ...	82	Actinium B	36.1 min	$\beta 1.36(92), 0.95(1), 0.53(6), 0.25(1)$	0.83(4), 0.70(1), 0.43(1), 0.40(4)
Bi-211 ...	83	Actinium C ^c	2.16 min	$\alpha 6.62(83), 6.27(17)$	0.35(14)
Tl-207 ...	81	Actinium C''	4.76 min	$\beta 1.47(100)$	0.87(1)
Pb-207 ...	82	Actinium D	Stable		

^a Compiled by W. M. Lowder from "Nuclear Data Sheets".

^b Figures in parentheses indicate per cent yield per disintegration.

^c Parallel decay branches of less than 1 per cent are not listed.

TABLE VII. SOME NON-SERIES PRIMORDIAL RADIO-ISOTOPES^{2, 111, 229}

Isotope	Abundance in the lithosphere (parts per million)	Half-life (years)	Alpha or beta ray energies (MeV) ^a	Gamma ray energies (MeV) ^a
K-40	3	1.3×10^9	$\beta 1.32(89)$	1.46(11)
V-50	0.2	5×10^{14}	Electron capture	0.71, 1.59
Rb-87	75	4.7×10^{10}	$\beta 0.27(100)$	
In-115	0.1	6×10^{14}	$\beta 0.6(100)$	
La-138	0.01	1.1×10^{11}	$\beta 0.20(30)$	0.81(30), 1.43(70)
Sm-147	1	1.2×10^{11}	$\alpha 2.24$	
Lu-176	0.01	2.1×10^{10}	$\beta 0.42(100)$	0.088(100), 0.202(100), 0.309(100)

^a Figures in parentheses indicate per cent yield per disintegration.

TABLE VIII. TYPICAL CONCENTRATIONS OF Ra²²⁶ AND DAUGHTERS IN CONTINENTAL WATERS (pCi/litre)³⁶

	Ra ²²⁶	Rn ²²²	Pb ²¹⁰	Po ²¹⁰
Spa waters and deep wells	1-10	10^4-10^5	< 0.1 ^a	~ 0.02
Ground water	0.1-1	10^2-10^3	< 0.1 ^a	~ 0.01
Surface water ...	< 1	10	< 0.5	—
Rain-water	—	10^3-10^5 b	0.5-3	~ 0.5

^a Below detection limits.

^b As determined through presence of short-lived Rn²²² daughters.

TABLE IX. RECENT DATA (REPORTED SINCE 1962) ON CONCENTRATION OF NATURAL RADIO-ACTIVE NUCLIDES IN WATER

(Concentration in pCi/litre)

Country or area	Source of water	Ra ²²⁶	Ra ²²⁸	Rn ²²²	Pb ²¹⁰	References
AUSTRALIA	Surface reservoirs	0.1-0.2				232
BRAZIL, high background area of Morro di Ferro (volcanic intrusives)	{ Different sources Tap, wells and springs	11.9 (0.12-107)				233 104
BRAZIL, monazite area	Tap and wells	0.3	0.2-1.6			104
INDIA, normal areas	{ Tap water (Bombay) Surface waters Springs and wells	0.02 0.05-0.6 ^a 0.16-0.5 ^a				180 106 106
INDIA, monazite area of Kerala State	Shallow wells	0.14				180
ISRAEL	{ Surface waters Springs, wells and boreholes Dead Sea (lake)	< 2 62		18-180 < 2-21, 300 20		128 128 128
JAPAN	Surface waters (rivers)	0.04-1.4				119
NEW ZEALAND	{ Artesian wells Surface waters			~ 0-1,000 0.3		234 234
SWEDEN	Tap water of varying origin (deep bored wells, inclusive)			~ 1,000 (100-33,000)		129
UNITED STATES						
Illinois	{ Ground waters Raw surface waters Treated surface waters Raw well water			30-300	0.127 0.019 0.05	117, 165 15, 81, 117, 165 15, 81, 117, 165 15, 81, 117, 165
Florida	Thermal and mineral springs	0.3-3.3				121
South Carolina	Wells	1.4-2.8				235
Utah, near Great Salt Lake	Artesian wells	0.1-2.0		400-1,800		

^a Including Ra²²³ and Ra²²⁴.

TABLE X. ESTIMATES OF TOTAL INTAKE OF Ra²²⁶ AND OF CONTRIBUTIONS FROM DIFFERENT FOODSTUFF CATEGORIES

Category of foods	UNITED STATES					UNITED KINGDOM ¹⁸⁵		INDIA ¹⁸⁶	
	New York, N.Y. ¹⁸⁷	Chicago, Ill. ¹⁸⁷	San Francisco, Cal. ¹⁸⁷	San Juan, P.R. ¹⁸⁶	Consumers' Union Five-city study ¹⁸⁸	Teenager twenty-two-city study ¹⁸⁹	Country-wide study	Bombay	Kerala State Monazite area
Cereals and grain products	0.56	0.76	0.51				0.17	0.41	1.48
Meat, fish, eggs	0.38	0.37	0.28				0.38		
Milk and dairy products	0.14	0.12	0.13				0.14	0.04	0.19
Green vegetables, fruits and pulses	0.81	0.56	0.48				0.32	0.17	0.81
Root vegetables	0.40	0.22	0.26				0.10	0.02 ^a	0.07 ^a
Water	~ 0.02	~ 0.03	~ 0.01				0.07	0.06	0.29
Total pCi/day	~ 2.5	~ 2.1	~ 1.7	~ 0.7	~ 3 (2.2-4.3)	~ 5 (2.5-6.5)	~ 1.2	~ 0.7	~ 2.8
pCi Ra ²²⁶ /g Ca	2.2	2.0	1.6	1.3	1.9	2.5	1.1		

¹ Miscellaneous.

TABLE XI. Ra²²⁶ IN HUMAN BONE AS REPORTED AFTER 1962

Location of area	pCi/g ash	pCi/g Ca	Total ^a in the skeleton (pCi)	References
NORMAL AREAS				
CENTRAL AMERICA				
United States				
Puerto Rico	0.006	0.017	17	190
EUROPE				
Federal Republic of Germany				
United Kingdom	0.013	0.040	36	203
0.008-0.02				
NORTH AMERICA				
United States				
Illinois	0.012 ^b		32	183
New England	0.014		39	209
New York, N.Y.	0.012	0.032	32	186
Rochester, N.Y.	0.010; 0.017		28, 48	204
San Francisco, Cal.	0.0096	0.026	27	186
HIGH LEVEL AREAS				
ASIA				
India				
State of Kerala	0.096		~ 270	180, 216
(monazite area)	(0.03-0.14)			
NORTH AMERICA				
United States				
Illinois	0.037 ^c		~ 100	165
Illinois	0.028 ^c		78	183

^a Skeleton of 7,000 g fresh weight yielding 2,800 g ash was assumed.

^b In people consuming water with "normal" levels of Ra²²⁶.

^c In people consuming water with elevated Ra²²⁸ concentration.

TABLE XII. Pb²¹⁰ AND Po²¹⁰ IN HUMAN BONE

(Number of samples in brackets)

Area or location	Pb ²¹⁰		Po ²¹⁰		Po ²¹⁰ / Pb ²¹⁰	Total Po ²¹⁰ in the skeleton ^a (pCi)	References
	pCi/g fresh bone	pCi/g ash	pCi/g fresh bone	pCi/g ash			
NORMAL AREAS							
FEDERAL REPUBLIC OF GERMANY	0.032 (20)	0.11 (20)	0.031 (20)	0.13 (20)	1	290	213
	0.037		0.030		0.8	210	203
POLAND	0.040 (20)					250	237
	0.026 (5)					160	238
UNITED KINGDOM			0.017 (6)			120	239
			0.017 (9)			120	214
UNITED STATES							
Illinois (mostly) ...		0.150 (128)			1	410	165
Illinois		0.080 (32)				200	183
New England		0.140 (25)				360	209
Rochester, N.Y. ...	0.015 (18)					95	212
San Juan, P.R.		0.120 (28)				300	240
HIGH LEVEL AREAS							
CANADA (Eskimos) ..	0.140 [0.08-0.71] (10)					960	193

^a Calculated directly from Po²¹⁰ or from data on Pb²¹⁰. In the latter case, if no data for Po²¹⁰/Pb²¹⁰ ratio were reported the value of 0.9 was assumed. Skeleton of 7,000 g fresh weight yielding 2,800 g ash was further assumed. If the data were reported both for fresh tissue and tissue ash, the values in pCi/g ash were used to estimate the total skeletal burden.

TABLE XIII. NATURAL ALPHA EMITTING RADIO-NUCLIDES IN HUMAN SOFT TISSUES IN AREAS OF NORMAL BACKGROUND RADIO-ACTIVITY
VALUES ARE GIVEN IN pCi/kg FRESH WEIGHT

For Po^{210} the ranges include the average values reported by various authors

Tissue	Ra^{226}	Th^{232}	Po^{210}
Soft tissues in general	~ 0.1203, 206	~ 0.04203	~ 3203
Liver	~ 0.2204	—	11-17116, 193, 198, 203, 214, 239
Kidney	0.1204	—	5-17193, 198, 203, 214, 239
Gonads	—	—	3-4193, 198, 214, 239
Spleen	0.1204	—	3203, 239, 240
Lung	—	—	2-5193, 198, 214, 228, 239
Skeletal muscle	0.06204	—	1-6165, 193, 203, 239

TABLE XIV. Ra^{226} IN HUMAN TEETH FROM DIFFERENT AREAS

Values in pCi/g ash; number of samples in parentheses

Locality	Mean	Range	References	Remarks
NORMAL AREAS				
BRAZIL				
Vitoria	0.030	0.008-0.079 (14)	215	
Rio de Janeiro	0.037	0.006-0.123 (13)	215	
Poços de Caldas	0.015	0.006-0.031 (13)	215	
UNITED STATES				
New England	0.016	0.01-0.062 (25)	209	
AREAS OF HIGH TERRESTRIAL RADIO-ACTIVITY				
BRAZIL				
Guarapari	0.036	0.006-0.104 (23)	215	Monazite sand areas
Mcaipe	0.023	0.006-0.077 (15)	215	Monazite sand areas
Araxa and Tapira	0.077	0.008-0.204 (52)	215	Areas of high radio-activity due to volcanic intrusives

TABLE XV. Po^{210} CONTENT OF VEGETABLES AND ANIMAL TISSUES¹⁹³

Materials	Number of samples	Po^{210} specific activity (pCi/kg)	Pb^{210}/Po^{210} activity ratio	Materials	Number of samples	Po^{210} specific activity (pCi/kg)	Pb^{210}/Po^{210} activity ratio
Grass (dried) United Kingdom	24	400-16,000	1-5	Beef and lamb kidney (United Kingdom)	3	48-270	0.05-1
Dry lichen (<i>Caloplaca elegans</i>) United Kingdom	2	7,800; 10,000	1	Lamb kidney (north Wales)	6	90-1,800	0.2
Dry lichen (<i>Cladonia alpestris</i>) Lapland	3	6,600-8,100	1	Reindeer (Lapland) summer killed			
Dry lichen (<i>Cladonia alpestris</i>) Canada	1	3,500	1	Muscle	6	15-50	—
Edible green vegetables (United Kingdom)	5	6-90	1-3	Liver	5	350-750	—
Carrots and potatoes (United Kingdom)	2	1	—	Kidney	4	110-490	—
Breads and cereals (United Kingdom)	4	1-7	—	Reindeer (Canada, Northwest Territory) winter killed			
Dried milk powder (United Kingdom)	3	2-6	—	Muscle	2	200; 210	—
Beef and lamb muscle (United Kingdom)	2	3; 3	—	Liver	2	2,400; 5,600	—
Beef and lamb liver (United Kingdom)	3	4-100	0.7	Kidney	2	4,200; 2,300	—
				Spleen	1	980	—
				Cockles (United Kingdom, east and west coasts)	3	400-900	0.1-0.2
				Crab (United Kingdom, south coast)	2	1,300; 1,400	—
				Plankton (south Pacific) ..	1	2,000	—

TABLE XVI. INTERNAL DOSES FROM NATURAL RADIO-ACTIVITY;^a VALUES IN MILLIRADS PER YEAR

(In parentheses, fraction in per cent of total yearly dose derived from alpha radiation)

Nuclide	Gonads	Bone		Bone marrow (trabecular bone)
		Haversian canals (50 μ diameter)	Osteocytes (50 μ diameter)	
K ⁴⁰	20	15	15	15
Rb ⁸⁷	0.3	< 0.3	< 0.3	< 0.3
C ¹⁴	0.7	1.6	1.6	1.6
Ra ²²⁶	— ^e	0.6 ^{f, d}	1.4 ^{f, d}	0.03 ^f
Ra ²²⁸	— ^e	0.7 ^{f, b}	1.1 ^{f, b}	0.03 ^f
Po ²¹⁰	0.3 ^f	2.1 ^{f, c}	4.2 ^{f, c}	0.3 ^f
Rn ²²²	0.3 ^f	0.3 ^f	0.3 ^f	0.3 ^f
TOTAL	21.6	20.3	23.6	17.3
	(3)	(18)	(30)	(4)

^a Doses of alpha radiation to tissues of respiratory tract are discussed in paragraphs 141-148. The highest doses are most probably those delivered to basal layers of bronchial epithelium in segmental and lobar bronchi. They may reach the level of some hundreds of millirads per year at assumed average concentrations of Po²¹⁸, Pb²¹⁴ and Po²¹⁴.

^b Accepted concentration of Ra²²⁸ (in equilibrium with daughter products) of 1.4×10^{-8} pCi/g fresh bone.

^c Accepted concentration of Po²¹⁰ of 3.3×10^{-2} pCi/g fresh bone.

^d Accepted concentration of Ra²²⁶ (plus one-third of short-lived daughter products through Po²¹⁴) of 4.3×10^{-8} pCi/g fresh bone.

^e Doses to gonads from Ra²²⁶ and daughter products and Ra²²⁸ and daughter products could not be estimated with reasonable accuracy. The upper limits seem to be 0.02 and 0.03 mrad/year, respectively.

^f Doses of alpha radiation.

TABLE XVII. DOSE RATES DUE TO EXTERNAL AND INTERNAL IRRADIATION FROM NATURAL SOURCES IN "NORMAL" AREAS

(In italics, estimates given in the 1962 report)

Source of irradiation	Dose rates (mrad/y)			Paragraphs
	Gonads	Haversian canal	Bone marrow	
<i>External irradiation</i>				
Cosmic rays				
Ionizing component	28	28	28	48
Neutrons	0.7	0.7	0.7	49
Terrestrial radiation (including air)	50	50	50	58
<i>Internal irradiation</i>				
K ⁴⁰	20	15	15	136
Rb ⁸⁷	0.3	< 0.3	< 0.3	136
C ¹⁴	0.7	1.6	1.6	136
Ra ²²⁶	—	0.6	0.03	135-139
Ra ²²⁸	—	0.7	0.03	135-139
Po ²¹⁰	0.3	2.1	0.3	135-139
Rn ²²² (dissolved in tissues) ..	0.3	0.3	0.3	135-139
TOTAL ^a	100	99	96	
Percentage from alpha particles and neutrons	1.3	4.4	1.4	
	3	2.8	3	

^a Totals were rounded off to two significant figures.

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