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DOSSIER

DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS : RECHERCHES ACTUELLES

FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Natural radiation sources, including some lessons for nuclear waste management

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Abstract The average effective dose at the global level is, according to UNSCEAR, estimated to be 2.4 mSv from naturally occurring sources. This average value can be divided as follows: 1.3 mSv associated with radon, 0.39 mSv from cosmic radiation, 0.46 mSv from terrestrial radiation, and 0.23 mSv from internal radiation, radon excluded. These values can vary quite significantly depending on the place of habitation. Despite this large variation, no sound epidemiological study has yet shown the health effects on the most exposed populations, apart from a few studies concerning radon, but in which the predominant role of tobacco is difficult to determine. ***To cite this article: H. Métivier, C. R. Physique 3 (2002) 1035–1048.***

ionizing radiation / natural irradiation / radiotoxicity / biological effects

Les sources naturelles d'irradiation, leçons pour la gestion des déchets nucléaires

Résumé La dose efficace moyenne au niveau mondial est estimée selon l'UNSCEAR à 2,4 mSv pour les sources d'origine naturelles. Cette valeur moyenne se décompose en 1,3 mSv liés au radon, 0,39 mSv au rayonnement cosmique, 0,46 mSv au rayonnement tellurique, et 0,23 mSv liée à l'irradiation interne à l'exception du radon. Ces valeurs peuvent varier très significativement selon le lieu d'habitation. Malgré cette variation importante aucune étude épidémiologique solide n'a à ce jour montré d'effets sur la santé dans les populations les plus exposées, si ce n'est quelques études concernant le radon, mais où le rôle écrasant du tabac est difficile à préciser. ***Pour citer cet article : H. Métivier, C. R. Physique 3 (2002) 1035–1048.***

radiation ionisantes / irradiation naturelle / radiotoxicité / effets biologiques

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1. Introduction

Exposure to ionising radiation arises from two main sources: natural radiation, to which Man is constantly exposed, and 'human' radiation sources, that is, associated with modern technology. For most countries, irradiation from natural sources remains the more significant, followed by medical irradiation, which depends closely on the technological level of the country. An inventory of these types of irradiations is updated regularly by the United Nations Scientific Committee on the Effects of Ionising Radiation (UNSCEAR), the latest being published in 2000 [1]. For France, the average individual radiation due to natural radioactivity is approximately 2 mSv per year in terms of effective dose, but it can vary from one region to another by a factor of three.

2. Radioactivity and exposure: basic notions

Radioactivity is associated with the disintegration of the nuclei of certain special atoms, for this reason referred to as 'radioactive'. During this disintegration, transmission of alpha or beta particles occurs, and may be accompanied by electromagnetic radiation – gamma radiation – when the atoms produced return to their stable state. Such disintegration can be observed in nature – where it is known as *natural radioactivity*, or is produced by man, whence the expression *artificial radioactivity*.

Whether the source be natural or artificial, radioactivity has the same impact, is measured using the same methods, and produces the same effects on man and matter in general. The phenomenon (radioactivity) is today defined in *becquerel* (Bq), corresponding to one disintegration per second.

The time required for one-half of the atoms initially present to disintegrate is known as *half-life.* By analogy, the time necessary for one-half of a radioactive nuclide to disappear from the human body is known as *biological half-life*. If for an equal weight a product is more radioactive than another, it is said that its *specific radioactivity* is greater – it is inversely proportional to the half-life.

The irradiation of individuals is a question of *external exposure* when the radiation source is on the outside of the body; exposure ceases as soon as the radiation source is sufficiently remote or a sufficiently thick screen can be interposed. Irradiation increases with the activity of the radiation source, its proximity to the subject, and the duration of the exposure. Conversely, if the radioactive source is on the inside of the body, irradiation is a question of *internal exposure*, and it continues until the radioactive nuclide is removed, even if the human exposure to the source of the contamination has ceased. Exposure resulting from the incorporation of radioactive nuclides therefore depends on the ingested or inhaled activity, the nature of the pollutant, and the time required by the human organism to eliminate it. In the case of inhalation, the incorporated activity (in Bq) can be deduced from the activity concentration of the air breathed (in $\mathrm{Bq{\cdot}m^{-3}}$), and from the average respiratory rate of the exposed individual ($\mathrm{m^3{\cdot}h^{-1}}$). In the case of ingestion, the question is to estimate the specific activities ($\mathrm{Bq{\cdot}kg^{-1}}$) and the average quantities of the foodstuffs consumed ($\mathrm{kg{\cdot}j^{-1}}$ or $\mathrm{kg{\cdot}yr^{-1}}$).

3. Notions of dose

The irradiation of an organism causes a greater or lesser degree of injury. The magnitude used for determining the quantity of radiation received is the *dose*. It is therefore the estimation of *dose*, known as dosimetry, which allows the degree of human attack resulting from the various types of exposure to be quantified. In order to compare the biological effects of all ionising radiation, irrespective of origin, the International Commission on Radiological Protection (ICRP) has created the concept of equivalent dose, and, to take account of varying sensitivities in tissues at stochastic risk, that of effective dose. These will be discussed in this section.

3.1. Absorbed dose

The radiation emitted by radioactive substances interacts with matter, releasing energy into the latter. The quantity of energy transferred is called the *absorbed dose*. It is expressed in Gray (Gy), corresponding to an energy level of 1 J transferred into one kilogram of material ($1\ \mathrm{Gy} = 1\ \mathrm{J{\cdot}kg^{-1}}$). This transfer of energy is a source of disturbance in the exposed matter, namely, causing the ionising of the constituent atoms.

3.2. Equivalent dose

Not all radiation produces the same effects. In place of the former so-called Quality Factor (Q), ICRP Publication 60 [2] now selects radiation weighting factors, w_R, based on a review of the biological information on the harmful effects of radiation. They are closely related to the previously used quality factor (Q) and are also related to the *linear energy transfer* (LET) and *relative biological effectiveness* (RBE) for tumour induction. The concept of *equivalent dose* allows the expression of these two effects in equivalent terms, and thus the comparison of them, by admitting the notion of 'radiation harmfulness factor'.

The equivalent dose to an organ $H_{T,R}$ in a tissue T associated with exposure to radiation R, is given by the equation:

$$H_{T,R} = w_R D_{T,R},$$

where $D_{T,R}$ is the absorbed dose by organ T due to irradiation R and w_R, the radiation weighting factor associated with radiation type R. The total equivalent dose, H_T, is the sum of $H_{T,R}$ for all types of radiation. Since the quality factor has no dimension, the unit is the same as for the absorbed dose; $\mathrm{J{\cdot}kg^{-1}}$ and employs a special unit known as the sievert (Sv).

ICRP Publication No. 60 [2] allocates the following radiation weighting factors w_R given in Table 1 to each radiation type.

For alpha radiation, the equivalent dose is therefore equal to twenty times the absorbed dose, whereas for gamma radiation, it is equal to the absorbed dose.

3.3. Effective dose

Not all tissues have the same sensitivity to radiation. The notion employed for estimating the risks of inducing late effects (cancers or genetic anomalies) into the entire organism, is therefore the *effective dose*, which takes account of the radiosensitivity of the various tissues in question.

Table 1. Radiation weighting factors for different types of radiation [2]

Photons from all energy sources	1
Electrons and muons from all energy sources	1
Energy neutrons < 10 kev	5
10–100 kev	10
100 keV–2 MeV	20
2 MeV–20 MeV	10
> 20 MeV	5
Protons other than recoil protons, energy > 2 MeV	5
Alpha, fission fragments, heavy nuclei	20

Table 2. Tissue weighting factors [2]

Gonads	0.20	Breast	0.05
Bone marrow	0.12	Oesophagus	0.05
Colon	0.12	Thyroid	0.05
Lung	0.12	Skin	0.01
Stomach	0.12	Bone surface	0.01
Bladder	0.05	Other tissues or organs[a]	0.05

[a] For computational reasons, 'other tissues or organs' in includes adrenal glands, brain, upper large intestine, small intestine, kidney, muscle, pancreas, spleen, thymus and uterus.

The effective dose is the sum of the equivalent doses for each organ or tissue in the organism, weighted by a factor relating to the sensitivity of the irradiated tissue to stochastic effects, w_T. Since the weighting factors are standardised, their sum is equal to 1.

The effective dose can therefore be expressed by the equation:

$$E = \sum_T w_T \, H_T .$$

ICRP publication No. 60 [2] allocates the following tissue weighting factors given in Table 2 for the organs specified

Since the tissue weighting factor has no dimension, the unit is the same as for the absorbed dose and equivalent dose for organs: $\mathrm{J{\cdot}kg^{-1}}$; it employs the same unit as the equivalent dose for organs – the sievert (Sv).

Thus, using a measurable given quantity – the absorbed dose – it is possible to estimate a risk to an individual through the concept of 'effective dose'.

3.4. Special consideration applying to internal exposure: the notion of dose-commitment

In the case of internal exposure, irradiation of the organism continues until the latter has eliminated the radioactive nuclide. To take account of the overall dose received during the time the radioactive nuclides remain in the organism once committed, the ICRP has created the concept of *committed dose*. This defines the equivalent dose to the organ, or the effective dose to the entire organism, committed as soon as the radioactive nuclides are ingested or inhaled. If the exposed individual is an adult, the estimation of the committed dose takes account of the irradiation of the subject's organism for 50 years following receipt, irrespective of his/her age. For a child, from infancy to adulthood, the effective committed dose estimation is performed by convention until age 70. If the calculation is made over a shorter period, the dose is known as the *truncated dose* and the period over which it was calculated shall be specified: 1, 3 or 5 years for example. The dose coefficients published by ICRP are calculated for 50 years, or up to age 70; therefore their use in estimating medium-term effects must demand precautions.

The method of calculating the committed equivalent dose takes into account for each target tissue, T, the exposure resulting from the committed radiation placed in the subject, and any other source organ 'S' when the radiation is of a sufficiently penetrating nature, and continues to incorporate the phenomena in the years following commitment.

The ICRP proposes two types of models – input models (pulmonary, gastro-intestinal, skin, injuries) which depend on the physicochemical form of the contaminant, and systemic models which depend only on the biological form carrying the contaminant around after it has dissolved and been transformed at the entry site.

3.5. Latest ICRP recommendations

The ICRP published new recommendations in 1991 [2] taking account of changes in scientific knowledge at this time, and especially data concerning survivors of the Hiroshima and Nagasaki bombs, and the UNSCEAR conclusions of 1988 [3]. Given the time of occurrence of the various cancers observed, the ICRP modified the tissue weighing factors (w_T) of the various organs. Since the previous recommendations [4], new scientific work has shown that the toxicity of neutrons had been underestimated; the ICRP has therefore doubled the harmfulness value factor of neutrons (w_R). Allowing for re-evaluation of the doses received by the bomb survivors, confirmed by the UNSCEAR, the ICRP has modified its factors of risk per unit dose.

Given the foregoing factors, the ICRP has recommended new limits: a limit of 100 mSv over five years for workers, provided that a value of 50 mSv is not exceeded in any one year, and a yearly limit of 1 mSv for the public, limit values which must not be added to the natural radiation. Natural irradiation is not taken into account with regard to dose limit. Finally, in the category of workers exposed to ionising radiation, it has added workers exposed to natural radiation sources, when their working conditions do not correspond to normal conditions (miners, airline flight crews, etc.).

Since this publication, the hypothesis that the dose-response relationships are linear down to near zero dose, and that the risks of cancer induction can be estimated approximately from observations of the effects seen with moderate doses, have both come under attack. The most notable criticism has come from a committee of the French Academy of Sciences [5]. However, although the conclusions of the report of the French Academy of Sciences were convincing, the problem is what to put in its place as a basis for public policy and the avoidance of harm. This problem is particularly acute for regulators and radiotoxicity is a good illustration of this difficulty, as discussed below.

4. Radiotoxicity

The radiotoxicity of a radioactive nuclide is defined currently in terms of the effective committed dose that an individual will receive, following commitment of a radioactive nuclide. The dose per unit of intake (DPUI) is expressed in Sv per Bq. The greater the DPUI, the greater the assumed radiotoxicity of the radioactive nuclide. Its calculation, as we have seen, takes account of the half-life of the radioactive nuclide, its emitted energy level, and its biological half-life. Some values are given in Table 3.

Radiation-protection standards are currently based, as a precautionary measure, on the hypothesis that irrespective of the radiation dose received by an organism, a stochastic effect will be possible; only the

Table 3. Specific and radiotoxicity activities

Radionuclide	Physical half-life (years)	Specific activity ($Bq \cdot g^{-1}$)	Committed effective dose after ingestion [6] ($Sv \cdot Bq^{-1}$)
^{129}I	$15.7 \cdot 10^6$	$6.5 \cdot 10^6$	$1.1 \cdot 10^{-7}$
^{135}Cs	$2.3 \cdot 10^6$	$3.3 \cdot 10^7$	$2.0 \cdot 10^{-9}$
^{137}Cs	30.2	$3.2 \cdot 10^{12}$	$1.3 \cdot 10^{-8}$
^{238}U	$4.47 \cdot 10^9$	$1.2 \cdot 10^4$	$4.5 \cdot 10^{-8}$
^{235}U	$7.04 \cdot 10^8$	$7.1 \cdot 10^4$	$4.7 \cdot 10^{-8}$
^{237}Np	$2.14 \cdot 10^6$	$2.6 \cdot 10^7$	$1.1 \cdot 10^{-7}$
^{238}Pu	87.7	$6.3 \cdot 10^{11}$	$2.3 \cdot 10^{-7}$
^{239}Pu	$2.41 \cdot 10^4$	$2.3 \cdot 10^9$	$2.5 \cdot 10^{-7}$
^{241}Am	432	$1.3 \cdot 10^{11}$	$2.0 \cdot 10^{-7}$

frequency of appearance will be a function of the received dose. The relationship between the dose and the effect is assumed to be linear without threshold. The radiotoxicity, based on the dosimetric approach alone, subtends this linear non threshold dose–effect relationship, irrespective of the biological effects actually observed.

The system of 'equivalent dose' and 'effective dose' allows the comparison of the effects of all radiations, and in a relatively simple manner, despite its uncertainty level, and enables management of radiation protection in all laboratories and factories. It would appear difficult to question this method, especially in installations where the potential hazard are mixed. The results of the last fifty years show that it is operationally reliable.

Conversely, for situations where the problem is not to legislate for human activities, but to foresee the potential hazard of such-and-such an activity, or estimate the consequences of an accident, it becomes perfectly feasible – even necessary – to avoid adopting this type of attitude, in favour of the actual hazard, if scientific knowledge so allows. This is the case with long-term waste management, and also exposure to certain natural sources.

When the irradiation is the result of internal exposure following contamination by long-lived radionuclides, this relationship can be challenged in favour of a supralinear relationship or a relationship with threshold or threshold-like. The acquisition of sound scientific knowledge could allow a specific approach to risk-assessment, which could appropriately answer the problem of managing long-lived nuclear wastes, which by definition have a lower specific activity [7]. Their return to man via the food chain would only result in low exposures at low dose rates. The notion of specific activity in this case is preponderant. It will be remembered that a human thyroid contains between 10 and 15 mg of iodine [8], which would represent between 65 000 and 100 000 Bq were it all ^{129}I, that represent an equivalent dose to the thyroid higher than 40 Sv.

A nonlinear dose-effect relationship has been observed after inhalation of oxide of ^{239}Pu by a rat [9] or inhalation of ^{239}Pu or ^{238}Pu oxide by dogs [10,11]. This phenomenon is also observed in humans for bone cancers induced at low dose rates by ^{226}Ra [12–14], and according to some, after inhalation of plutonium oxide by workers at the Mayak complex [15]. To all these examples, compromising alpha emitters, must be added the work of the Davis University of California, among dogs that had ingested ^{90}Sr. Although the latter is a beta emitter, there is also a threshold for all cancers observed in this study: 13.3 Gy for leukaemia, and around 20 Gy for other cancers including bone cancers [13,14].

5. Natural radiation sources [1,16]

5.1. Cosmic radiation

5.1.1. *Cosmic rays*

Cosmic rays are fluxes of very high-energy charged particles streaking through space in all directions. They are deflected by magnetic fields, especially that of the Earth, which returns the lower-energy particles back into space. Their presence is greatest around the poles.

Cosmic rays are composed of hydrogen nuclei (85%) and helium nuclei (12.5%). The remainder consists of electrons (1.5%), and approximately 1% of particles of atomic nuclei ranging from 4 to 26, sometimes higher. The average energy of these particles is 10^{10} eV, and the maximum energy, 10^{20} eV. The flux decreases with increasing energy – the major part comprises protons of 10^{9} eV. Conversely, one square metre of the Earth's surface receives one proton with an energy level greater than 10^{17} eV, once a century. Their origin is essentially galactic.

Radiation of solar origin has relatively lower energy, which explains that solar eruptions have little effect on the intensity and composition of cosmic radiation at sea level. Their relative importance is large outside the atmosphere.

Cosmic rays interact with the atmosphere, to produce electrons, gamma ray photons, neutrons and mesons. At sea level, mesons represent 80% of cosmic radiation, and electrons, 20%.

The atmosphere is an effective screen protecting us from cosmic radiation. It is considered that only 0.05% of such radiation arrives at sea level, but its intensity is twice that at 1500 metres. Buildings also absorb a part of this radiation – approximately 20%.

At sea level, it is considered that the effective dose associated with the cosmic component is approximately 240 µSv per year, to which must be added 30 µSv associated with neutron irradiation. This is therefore the annual dose level – approximately 300 µSv – received by each individual for this type of radiation. Populations living at high altitudes receive larger doses: the inhabitants of Denver, Colorado (alt. 1600 m) receive almost twice the dose received at sea level (570 µSv for the cosmic component). For inhabitants of Mexico City (alt. 2240 m), the figure is 820 µSv. Finally, in La Paz, Bolivia (alt. 3900 m), inhabitants receive 2000 µSv per year, divided almost equally between the directly ionising component and the neutron component. Given the geographical distribution of the inhabitants, and their way of life (outside or inside), the UNSCEAR estimates the average dose per habitant to be 380 µSv, 300 associated with directly ionising radiation, and 80 with the neutron component.

5.1.2. *Radioactive nuclides of cosmic origin*

As discussed above, the interaction of cosmic radiation with nuclei present in the atmosphere produces elementary particles and also a series of radioactive nuclides such as ^{3}H, ^{7}Be, ^{14}C, ^{22}Na, to mention only the most important in terms of their role in the irradiation of populations.

Carbon 14 ($T_{1/2} = 5730$ years) comes from the interaction of slow cosmic neutrons with nitrogen 14. Transformed into $^{14}CO_2$, it participates in the photosynthesis cycle. Its specific activity has very markedly diminished through dilution since the creation of CO_2 by the burning of fossil fuels. Today, the specific activity of carbon is approximately 230 Bq·kg^{-1}, and man ingests 20 000 Bq per year, the resulting annual effective dose being 12 µSv.

The main source of naturally occurring tritium ($T_{1/2} = 12.3$ years) results from the interaction of cosmic radiation with nitrogen and oxygen nuclei. The tritiated water produced in this mechanism participates in the water cycle. Its concentration level in continental water is 400 Bq·m^{-3}, and 100 Bq·m^{-3} in the oceans. Man on average receives 500 Bq, and the resulting dose is approximately 0.01 µSv [17].

Beryllium 7, with a half-life of 53.6 days, has a concentration of 3 mBq·m^{-3} in air. It returns to earth in rainwater, a process that contributes to an annual commitment for each individual (through fresh vegetables) of approximately 1000 Bq·yr^{-1}, delivering an effective dose of 0.03 µSv.

Finally, the annual commitment of ^{22}Na by man is approximately 50 Bq, but this contributes an effective dose of approximately 0.15 µSv, that is, considerably more that tritium.

The irradiation of populations through cosmogenic radioactive nuclides is therefore essentially linked with the production of ^{14}C; it is very slightly greater than 12 µSv per year.

5.2. Terrestrial sources

Naturally occurring radioactivity stems mainly from primordial radioactive nuclides and their descendants. Approximately 340 nuclides have been found in nature, and of these, 70 are radioactive – the radioactive nuclides. All elements with atomic number greater than 80 have radioactive isotopes, and all elements with $Z > 83$ have all their isotopes radioactive.

5.2.1. *Primordial radioactive nuclides*

Since the Earth's creation, all radioactive nuclides with half-lives of less than 10^8 years have become undetectable. Those primordial radioactive nuclides that remain have half-lives going from $7 \cdot 10^8$ years for ^{235}U, to $5 \cdot 10^{15}$ years for ^{142}Ce. The list is limited to around twenty radioactive nuclides.

Three primordial radioactive nuclides are particularly important since they produce secondary radioactive nuclides through radioactive decay. They are known as the 'decay series'. They include the decay uranium 238 series ($T_{1/2} = 4.47 \cdot 10^9$ years), thorium 232 ($T_{1/2} = 1.4 \cdot 10^{10}$ years) and uranium 235 ($T_{1/2} = 7.4 \cdot 10^8$ years), also known as the 'actinium series'. The quantity of descendants in equilibrium with the primordial

radioactive nuclide depends on the half-life of the latter: one tonne of uranium contains only 1 mg of ^{226}Ra and 1 µg of ^{210}Po.

In the Uranium 238 series, radium 226 is a more important element. With a half-life of 1600 years, it disintegrates into radon 222. It is encountered in significant concentrations in mineral water 40 mBq·l^{-1} of ^{226}Ra and 4 mBq·l^{-1} of ^{228}Ra in Evian water; 250 mBq·l^{-1} and 270 mBq·l^{-1} respectively in Badoit water, before treatment; and 240 mBq·l^{-1} and 170 mBq·l^{-1} in Vichy Saint Yorre water. With a chemistry similar to calcium, radium concentrates in the skeleton, which on average contains 850 Bq, leading to an annual effective dose of 7 µSv. According to the concentration values for radioactive nuclides published [18,19] the committed effective dose resulting from daily consumption of mineral water could vary from 30 µSv (Evian) to 280 µSv (Badoit and Saint Yorre). For an infant, whose intestinal absorption of these radioactive nuclides is approximately 10 times greater than for an adult, the committed dose would be 350 µSv during the first year of life in the case of an infant consuming only Evian water on a daily basis [20].

Brazilian hazel trees concentrate barium, and also radium, which has similar chemical properties. The content may go from 10 to 300 Bq·kg^{-1}, that is, 1000 times more that the average concentration in standard foods.

Uranium, present in phosphates in fertilisers, is also found in trace concentrations in certain foodstuffs.

Out of the descendants of these primordial radioactive nuclides, radon is the main source of irradiation of populations; therefore it will be discussed individually.

The concentration of primordial radioactive nuclides in the ground is greater in igneous rocks than in sedimentary rocks. The average concentration in ^{226}Ra varies from 0.4 to 4.1 Bq·kg^{-1} in basalts (average value: 11 Bq·kg^{-1}), to between 1 and 370 Bq·kg^{-1} in granites (average value: 78 Bq·kg^{-1}), but the variations may be still greater – from 1 to 1800 Bq·kg^{-1} in gneiss (average value 50 Bq·kg^{-1}). The result is a highly variable external irradiation between one region and another. Regions of the world exist where the irradiation is far greater than the world average: Kerala in India, where monazite is to be found; certain regions of Brazil; and to a lesser degree, certain regions of Italy, France, Sweden, Iran, Madagascar and Nigeria.

Besides these series, certain primordial radioactive nuclides disintegrate to give a stable element. The most important, in terms of dose, are potassium 40 ($T_{1/2} = 1.28 \cdot 10^9$ years) and rubidium 87 ($T_{1/2} = 4.7 \cdot 10^{10}$ years). Potassium 40 is ubiquitous: it has been estimated that 100 TBq are dispersed annually in fertilisers in the United States. Seawater contains 10 Bq·l^{-1}. Potassium is an essential element for life – it is the essential source of radioactivity in human food. Under homeostatic control, man contains 2 g of potassium per kg of body weight. The average concentration of potassium 40 is approximately 60 Bq·kg^{-1} – it is not influenced by environmental variations. The corresponding annual effective dose is approximately of 165 µSv for adults and 185 µSv for children. The average concentration of rubidium 87 is approximately 8.5 Bq·kg^{-1}, the resulting annual effective dose being estimated at 6 µSv approximately.

In short, the standard man (70 kg) is radioactive, containing an average – as regards the main radioactive nuclides – of 4400 Bq of ^{40}K; 3700 Bq of ^{14}C; 455 Bq of ^{87}Rb; 40 Bq of ^{3}H; 4 Bq of ^{226}Ra, and somewhat less than 4 Bq of ^{232}Th.

According to UNSCEAR, the average value for the annual effective dose resulting from external irradiation by radioactive nuclides in the terrestrial layer, is 0.46 µSv, the committed dose resulting from annual commitment of these radioactive nuclides being 0.25 µSv.

5.2.2. *Radon*

Radon (see [21]) has three natural isotopes: ^{219}Rn, ^{220}Rn and ^{222}Rn, resulting respectively from ^{235}U, ^{232}Th and ^{238}U. All of these are α emitters, but they have associated γ emissions linked to their descendants. Radon has its main source in soils, where it is formed by the disintegration of the radium atoms present in rocks. The amount produced therefore depends on the radium content, and also on the half-lives, which are, respectively, 4.05 s, 55.6 s and 3.82 d. Only a fraction, depending on the porosity of the soil, the size of the grains and the humidity, manages to escape. Once in the air, radon dilutes depending on the

Table 4. External exposure sources from terrestrial radiation ($nGy \cdot h^{-1}$) in the least- and most-exposed countries [1]

Country	Dose rate in $nGy \cdot h^{-1}$	Range	Population (in millions)
Cyprus	18	9–52	0.76
Iceland	28	11–83	0.27
Egypt	32	8–93	63
Netherlands	32	10–60	15.6
Brunei	33	3–70	0.3
United Kingdom	34	8–89	58
France	68	19–250	58.5
Portugal	84	4–230	9.8
Malaysia	92	55–130	20.6
Australia	93		18

Table 5. The most exposed regions of the world, from UNSCEAR [1]

Country	Region	Source of exposure	Dose rate ($nGy \cdot h^{-1}$)	Population
Brazil	Guarapari	Monazite	90–170 (streets)	73 000
			90–90 000 (beaches)	
	Mineas Gerals	Volcanic intrusions	2800	350
India	Kerala and	Monazite	200–4000	100 000
	Madras		(Average: 1800)	
China	Yangjiang	Monazite	370	80 000
Iran	Ramsar	^{226}Ra sources	70–17 000	2000
France	South-West	Uranium ores	10–10 000	
	Central Region	Granites	20–400	7 000 000

atmospheric conditions. Generally, a vertical concentration gradient is observed, with time-based variations depending on a nycthemeral cycle: during the day, diffusion is high and the rate is lower; at night, where frequent temperature inversions are observed, diffusion is poor, the radon stagnating at ground level; its concentration can thus increase by a factor of 10 to 100. At the surface of the earth, the average emitted flow is 0.022 $Bq \cdot m^{-2} \cdot s^{-1}$ for ^{222}Rn and 1 $Bq \cdot m^{-2} \cdot s^{-1}$ for ^{220}Rn. Various measurements of ^{222}Rn concentration around the world, in different climates and atmospheric conditions, have shown values from 0.1 $Bq \cdot m^{-3}$ to more than 100 $Bq \cdot m^{-3}$; therefore an average value of 10 $Bq \cdot m^{-3}$ is retained. The same average value is retained for radon 220 (thoron) [20].

Above the oceans, the average concentration of ^{222}Rn is 2 $Bq \cdot m^{-3}$.

In dwellings, radon has its main source in the subjacent ground structure, and sometimes in construction materials. For a given type of ground, the radon content depends on: the characteristics of the building (presence of a crawl space, cellar etc.); the presence of transfer routes from the ground to the various levels of habitation (such as pipes, stairways etc.); the level of ventilation; and the life-styles of the occupants. Degassing of tapwater originating from a well in granitic ground is a possible but much rarer source.

Average concentrations in dwellings (10–10 000 $Bq \cdot m^{-3}$) are very variable, depending on the location and country. UNSCEAR considers an average value of 40 $Bq \cdot m^{-3}$ on a world scale. This value also corresponds to the average value for France although the French distribution is largely dissymmetric (from 10 $Bq \cdot m^{-3}$ to about 5000 $Bq \cdot m^{-3}$. The proportion of dwellings with radon concentrations higher than 1000 $Bq \cdot m^{-3}$ (action level recommended for public buildings by the Conseil supérieur d'hygiène publique de France (CSHPF)) is estimated in France to be around 0.4%.

Whereas all the other natural radioactive nuclides affect man through foodstuffs, irradiation due to radon is associated with inhalation. In this case, irradiation will be heterogeneous, essentially affecting the lung and more particularly, the tracheobronchial tree.

As a noble gas, radon does not itself interact with the organism: irradiation of the organism will depend on its daughters irradiating the tracheobronchial tree, in which they lodge, and also the remainder of the organism, where they circulate if their half-life so allows. Calculating the delivered dose due to inhalation of radon and its descendants depends on several factors, particularly the type of aerosols on which the descendants can fix themselves. The result will differ depending on whether the subject lives outdoors or indoors; it will also depend on tobacco habits, for example. On the basis of these various parameters, UNSCEAR in 1982 recommended an effective dose factor of 0.009 µSv per $Bq \cdot m^{-3} \cdot h^{-1}$, and the ICRP recommendation of 1993 was for a factor of 0.003 µSv per $Bq \cdot m^{-3} \cdot h^{-1}$.

The annual irradiation for populations is on average 1200 µSv for ^{220}Rn and 73 µSv for ^{220}Rn: this therefore constitutes the main source of irradiation for man. In terms of internal radiation, ^{222}Rn and its descendants represent 70% of the dose; ^{40}K, 13%; ^{220}Rn and its descendants 13%; and ^{210}Pb and ^{210}Po, 13%. The remaining radioactive nuclides impart doses of little significance.

5.2.3. *Geographical variations*

External exposure due to terrestrial radiation varies from one region of the globe to another, depending on rock types. Table 4 gives the average dose rates of the least-exposed and the most-exposed countries. The values are national averages and do not representative any great local diversity.

These average values may be complemented by an inventory of the most exposed regions of the world (Table 5), according to UNSCEAR [1].

For radon, the local geology and type of dwelling ventilation can lead, for certain houses, to very varied levels of up to 100 times the world average.

6. Naturally occurring sources modified by technology

Numerous products are naturally radioactive, but only result in irradiation of populations if they are transformed (such as prior to processing). Thus fertilisers originating from natural minerals irradiate the population that uses them. Furthermore, if irradiation due to cosmic radiation is inescapable, recourse to airline travel exposes people to additional irradiation, which would not happen in the case of surface travel. All of the said activities represent sources of exposure to natural radiation, brought about by the development of technology.

6.1. Extractive industry

6.1.1. *Fossil energy*

Coal contains more radioactive products that any other source of fossil energy. The principal component is associated with ^{40}K. Burning coal produces ash that is more radioactive than the coal itself.

A 1000 MW electricity generating station, burning coal, releases approximately $4 \cdot 10^6$ tons per year of ash, that is, a total of approximately 150 to 600 GBq of radioactive products, depending on the type of coal and its sources (Table 6).

Table 6. Concentration (in $Bq \cdot kg^{-1}$) of coal and combustion ash

Radionuclide	Coal	Ash
^{40}K	50–100	250–700
^{238}U series	16–27	200–900
^{232}Th series	8–27	50–150

The resultant average effective dose for the population is 2 $\mu Sv \cdot yr^{-1}$. Modern plants, representing one-third of production, limit their waste by filtering the ash, thereby reducing their contribution to the annual effective dose by at least a factor of 10.

Due to its domestic usage, coal delivers an annual effective dose estimated at 0.4–8 µSv to populations using it for heating and cooking.

6.1.2. *Phosphate fertilisers*

Phosphatic minerals are the main source of materials for producing fertilisers. The main producers are China, Morocco, the ex-USSR, and the United States. Concentrations in ^{40}K and radioactive nuclides belonging to the ^{232}Th decay series are similar to those found in rocks of other composition; on the other hand, the concentration of radioactive nuclides belonging to the ^{238}U series is particularly high for phosphatic minerals of sedimentary origin (1500 $Bq \cdot kg^{-1}$ of ^{238}U).

The contribution of the uranium 238 decay series is large in phosphate fertilisers. Activity levels of 4000 $Bq \cdot kg^{-1}$ of ^{238}U, and 1000 $Bq \cdot kg^{-1}$ of ^{226}Ra can be measured in P_2O_5. Use of fertilisers represents the major source of contamination of the Planet due to ^{226}Ra. The annual effective dose per individual is estimated at approximately 2 µSv.

The two most resourced-based commodities produced by the phosphates industry are phosphogypsum and calcium silicate. These products are used in building materials, plasters and cements. It is considered that the phosphogypsum contained in plasters and cements delivers – due to inhalation of the radon it releases – an annual effective dose in the order of 0.6 mSv per year, associated with the presence of approximately 900 $Bq \cdot kg^{-1}$ of ^{226}Ra contained in the phosphogypsum. Calcium silicate has a higher concentration of ^{226}Ra (1300–2200 $Bq \cdot kg^{-1}$), and can lead to irradiation levels of 0.3 $\mu Gy \cdot h^{-1}$ in concrete dwellings.

6.2. Air transport and space activity

Two types of human activities can lead to higher radiation exposure from cosmic radiation: air transport and space activity [22].

Measurements were performed onboard the Russian MIR orbiting station between 1988 and 1995. An average equivalent dose rate of 30 to 40 $\mu Sv \cdot h^{-1}$ was thus measured. The station was particularly exposed when crossing what is known as the South Atlantic anomaly, where the equivalent dose rate can attain a value fifty times greater (1.5 $\mu Sv \cdot h^{-1}$). Given the factors of quality, and variations associated with solar eruptions, it is considered that the personnel onboard orbiting stations receive on average a daily effective dose of 1 mSv.

At the cruising altitude of long-range aircraft (between 9000 and 11 000 metres), the average equivalent dose rate is 2 or 3 $\mu Sv \cdot h^{-1}$ on a flight from Paris to Buenos Aires, and is minimal when crossing the Equator. On a polar flight from Paris to Tokyo, the equivalent dose rate is six to seven times greater in proximity to the magnetic pole. At the cruising altitude of the supersonic Concorde (18 000 metres), the dose rate during a Paris–New York flight is three to four times larger than for a subsonic flight.

Summing up, the annual effective dose associated with air transport attains an average of 5 mSv per year for flight personnel and certain frequent-flier passengers. It can attain 10 mSv for certain crews undertaking frequent polar flights. The exposure is 3 to 5 times lower for domestic flights.

These results have led the ICRP to include flight personnel in the category of workers subjected to occupational exposure. This proposal was incorporated in the European directive dated 13 May 1996.

6.3. Consumer products

Numerous consumer products contain radioactive nuclides that are in some cases added deliberately, while constituting as many radiation sources.

Watches can be made luminous through use of paint loaded with ^{226}Ra, ^{147}Pm and ^{3}H. Today, only tritium is used in watchmaking. The wearer of such a watch receives an annual irradiation of 0.3 µGy. The contamination of France's Rhone river was mainly associated with the watchmaking trade in Switzerland and the east of France. These activities are becoming increasingly limited.

Antistatic devices contain sources of between 20 and 30 mBq of ^{210}Po. Thorium is added into gas-lamp mantles, and uranium and thorium to certain porcelain. Finally, the consumer product delivering the highest dose is without a doubt tobacco, which contains ^{210}Pb and ^{210}Po [23].

To summarise, the most significant sources are the result of conservation of energy and exposure to radon from construction materials, as found in dwellings. Of all other activities, tobacco remains a significant source of exposure.

7. Health effects

Several epidemiological studies have been performed in some of the most exposed, inhabited regions of the planet. Of these, two showed positive results for heavy exposures to radon – one by S. Darby, which shows an increase in leukaemia on the basis of risk factors extrapolated from the survivors of Hiroshima and Nagasaki [24]. Nonetheless, UNSCEAR in its latest report [1] refutes this affirmation and declares that there is no significant association between natural radiation and leukaemia. Another study from Sweden [25] shows a significant risk tendency associated with radon exposure in dwelling houses. Other studies from Canada and Finland show no increase in risk [26–28], and the same is true of two American studies [29,30]. The role of tobacco is not elucidated in the Swedish study.

Population comparisons are of the best quality in a Chinese study comparing mortality through leukaemia in two regions, one (Yangjiang) representing a high content of monazite, and Taishan, the control community. The populations are sedentary and lend themselves to very efficient comparison. For women, the mortality rate is respectively 2.21 and 3.56 per year; and 3.32 and 3.82, for men in respectively the high- and low-rate regions. The results are not significant but suggest a lower rate in the most exposed regions [31].

UNSCEAR concludes that comparative studies into various groups of populations exposed to natural radiation levels from gamma radiation have never demonstrated any significant increase in cancer in the most exposed regions [1].

8. Conclusions

To summarise, the average effective dose at world level, as estimated by UNSCEAR, is 2.4 mSv from naturally occurring sources. This average value can be broken down as follows: 1.3 mSv associated with radon (0.2–10); 0.39 mSv from cosmic radiation (0.3–0); 0.46 mSv from terrestrial radiation (0.3–0.6); and 0.23 mSv attributable to internal radiation excluding radon (0.2–0.8), and 1.2 mSv attributable to inhalation, mainly radon (0.2–10).

Despite such a wide variation, no sound epidemiological study has to date shown any health effects associated with high natural irradiation. Some studies involving radon are ongoing but they are compromised by the predominant role of tobacco, which is always difficult to take into account.

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Question de J. Lafuma

Aujourd'hui aux USA, on a analysé les caractères fonctionnels de la cellule cancéreuse qui la différencient des cellules normales :
(1) capacité à se donner à elle-même l'ordre de se multiplier, ordre qui ne vient que des autres cellules dans un tissu normal ;
(2) capacité à 'ignorer' les signaux d'arrêt de prolifération ;
(3) capacité à ignorer les signaux d'autodestruction ;
(4) capacité de développer un système vasculaire nouveau ;
(5) capacité à l'invasion locale à la métastase.
Existe-t-il des publications expliquant comment on passe de la mutation initiale à la *vraie* cellule cancéreuse ?

Réponse de H. Métivier

MM. Dutrillaux et Masse sont mieux placés que moi pour répondre à cette question.

A propos des cancers de la thyroïde il faut noter que des épidémiologistes suédois et finlandais n'ont pas montré d'augmentation significative de ces cancers, bien que ces pays aient été particulièrement touchés, lors des premières phases de rejets riches en iode.