Natural and man-made radionuclides in the global atmosphere

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The impact of nuclear power on the global environment is due mainly to the release of radionuclides into the atmosphere. The importance of effluents from nuclear plants may be assessed by comparing them with natural releases and with emissions from other anthropogenic sources such as the production and testing of nuclear weapons or the coal fuel cycle.

Making a comparison of this sort is a simplified approach to environmental impact assessment, different from calculating tissue-absorbed doses in man and from the concept of dose-commitment to the human generations which are supposed to suffer the burden of present practices "over all future years" i.e. to the end of time [1]. These latter calculations need many arbitrary assumptions and are subject to uncertainty as the ecological, geological and demographic condition of the Earth in future millenia is very difficult to estimate. Estimating the radiation energy emitted to the environment from man-made sources here and now might be more helpful in understanding the relative impacts than such long-term assessments.

Indeed, as a result of the comparisons to be outlined in this article, it would appear that the discharge of ^{222}Rn from nuclear power operations might have a greater impact on the global environment than conventional dose-commitment calculations would have suggested. This nuclide seems therefore to deserve more research than it has hitherto been accorded, since conventional calculations have stressed ^{14}C releases as the major contributor to the impact of nuclear power on the environment. However, the comparisons to be made in this article underline once again that the current environmental impact of civilian nuclear energy is a very small fraction of the effect due to natural sources of radiation.

One may also regard it as improper to limit the assessments of the absorbed radiation-dose exclusively to human populations. Nowadays the effects of man's activity have reached global dimensions: each drop of rain, each plant and animal on the land and in the sea, contains traces of stable or radioactive substances released into the environment by humans. Man is usually exposed to the adverse effects of these releases much less than are species in lower positions in the ecological chain of food and energy transfer in the biosphere. The anthropocentric assumption that, when we keep the radiation-dose to human individuals at safe levels other species are automatically not endangered, simply ignores the burden imposed on those species. This was practically and perhaps ethically acceptable at earlier stages of industrial development. But as environmental impacts of this development are now of planetary scale, our responsibility or at least assessments, should embrace all living components of the biosphere.

To compare the environmental impact of radioactive emissions from three anthropogenic sources with natural ones, I have chosen arbitrarily ^{3}H, ^{14}C, ^{137}Cs, ^{238}U, ^{235}U, ^{226}Ra, ^{222}Rn, ^{210}Pb and ^{239}Pu because they are the radionuclides contributing most to the total radiation-dose received by the world's population. The estimates of annual flows of these nuclides are given in Table 1.

The energy which can be imparted to the environment from one disintegration or from one unit of activity differs by orders of magnitude for various nuclides. To estimate the relative impact, it is interesting to calculate the product of the radiation energy of each particular nuclide and the flow of its activity into the global atmosphere. The values of this product, i.e. the annual flows of *radiation energy*, calculated for various sources and nuclides, are given in Table 2.

Natural sources

The annual emissions of ^{3}H and ^{14}C into the global atmosphere are taken from UNSCEAR [2]. The average flows of ^{235}U, ^{238}U, ^{226}Ra, and ^{210}Pb were estimated from their concentrations in ice, deposited during the past three decades on the surface of nine glaciers located in widely dispersed geographical regions

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Table 1. Annual flows of activity into the global atmosphere (Ci)

Natural sources		Nuclear weapons between 1945 and 1981		Nuclear power in 1981		Coal burning in 1980	
^{222}Rn	9×10^{8} [4, 5]	^{3}H	1.9×10^{8} a [6]	^{3}H	1.5×10^{6} [10]	^{222}Rn	2.3×10^{4}
^{3}H	2×10^{6} [2]	^{3}H	7.6×10^{7} b	^{222}Rn	4×10^{5}	^{210}Pb	6.6×10^{2}
^{210}Pb	4.9×10^{5} [3]	^{137}Cs	7×10^{5} a [6]	^{14}C	3.4×10^{3} [10]	^{238}U	3.3×10^{2}
^{14}C	3.8×10^{4} [2]	^{14}C	1.7×10^{5} a [6]	^{210}Pb	2×10^{2}	^{226}Ra	65
^{226}Ra	6.6×10^{3} [3]	^{222}Rn	1.1×10^{4} b	^{238}U	3	^{235}U	16
^{238}U	4×10^{3} [3]	^{239}Pu	6×10^{3} a [6]	^{137}Cs	1.7 [1, 2]		
^{235}U	1.9×10^{2} [3]	^{239}Pu	1×10^{2} c [2]	^{226}Ra	1.4		
^{137}Cs	d	^{239}Pu	13 e [2]	^{235}U	1.5×10^{-1}		
^{239}Pu	d [2]	^{210}Pb	5.4 b	^{239}Pu	9.4×10^{-3} [1, 2]		
		^{235}U	1.7×10^{-1} a				
		^{238}U	1.6×10^{-1} a				
		^{238}U	8.2×10^{-2} b				
		^{226}Ra	3.6×10^{-2} b				
		^{235}U	2.1×10^{-7} b				

a = weapon tests 1945–1980
b = weapon production 1972–1981
c = weapon accidents
d = below detection limit
e = dispersed in stratosphere by spacecrafts

The source of information for every entry is given in parenthesis. Where no reference is given, the values are those estimated in this article.

in the northern and southern hemispheres [3]. The flow of ^{222}Rn was estimated as a mean flow from several countries [4, 5].

Nuclear weapons

Testing:

The total injections of ^{3}H, ^{14}C, ^{137}Cs and ^{239}Pu into the atmosphere by atmospheric weapons tests between 1945 and 1980 have been recently estimated by UNSCEAR [6]. These injections, averaged as annual flows, are given in Table 1. However, UNSCEAR did not estimate the flows of ^{235}U and ^{238}U from weapon testing.

I assume that among 423 past atmospheric explosions, of total energy yield of 545.4 Mt [6], 74 per cent had a yield less than one Megaton (Mt) and 26 per cent a yield greater than 1 Mt [7]. I assume also that in half the explosions, 20 kg of ^{235}U was used per weapon; and that the average burn-out efficiency in explosions of less than 1 Mt was 15 per cent, and 80 per cent in those greater than 1 Mt. With these assumptions, the tentative estimate of dispersion of ^{235}U in the atmosphere between 1945 and 1980 is about 2800 kg i.e. 1.7×10^{-1} Ci per year.

To estimate the dispersion of ^{238}U, I assume that in the 110 explosions greater than 1 Mt [7], which make up 92 per cent of the total energy yield (502 Mt), half the yield was from the fission of ^{238}U with 40 per cent burn-out efficiency, and that for 1 Mt energy release 56 kg of ^{238}U was fissioned. From this, the tentative estimate of dispersion of ^{238}U in the atmosphere between 1945 and 1980 is about 16 900 kg, i.e. 1.6×10^{-1} Ci per year.

Production:

Eisenbud et al. [8] reported that in 1980 the radiation-dose to humans in the northern hemisphere from the past production of nuclear weapons was a factor of 2.5 lower than the dose from nuclear tests. From this one may infer that the average annual flow of ^{3}H from this source, between 1945 and 1980, was 7.6×10^{7} Ci.

Except for this I was unable to find in the literature any information on the global releases of radionuclides from weapons production. A rather tentative assessment of these releases is presented here, based on SIPRI [9] data on nuclear-weapons' production. Between 1972 and 1981, 90 699 nuclear warheads have been produced by the two super-powers; 72 470 warheads having a yield less than 1 Mt and 18 229 greater than 1 Mt. I do not take into account in this estimation the contribution

Table 2. Average annual flow of radiation energy into the global atmosphere (J s^{-1})

Natural sources		Nuclear weapons between 1945 and 1981			Nuclear power in 1981		Coal burning in 1980	
^{222}Rn	1.2×10^{8}	^{3}H	2.7×10^{4}	a	^{222}Rn	5.3×10^{4}	^{222}Rn	3×10^{3}
^{210}Pb	1.9×10^{4}	^{3}H	8×10^{3}	b	^{3}H	1.6×10^{2}	^{210}Pb	2.6×10^{1}
^{226}Ra	1.1×10^{3}	^{137}Cs	4.5×10^{3}	a	^{210}Pb	8	^{238}U	1.3×10^{1}
^{3}H	2×10^{2}	^{222}Rn	1.5×10^{3}	b	^{235}U	5	^{226}Ra	1×10^{1}
^{238}U	1.6×10^{2}	^{239}Pu	1.8×10^{2}	a	^{14}C	3	^{235}U	4.5×10^{-1}
^{14}C	3.5×10^{1}	^{14}C	1.6×10^{2}	a	^{226}Ra	2.2×10^{-1}		
^{235}U	5.3	^{239}Pu	3	c	^{238}U	1.2×10^{-1}		
^{137}Cs	d	^{239}Pu	4×10^{-1}	e	^{137}Cs	1.1×10^{-2}		
^{239}Pu	d	^{210}Pb	2.1×10^{-1}	b	^{239}Pu	2.9×10^{-4}		
		^{238}U	6.2×10^{-3}	a				
		^{226}Ra	5.8×10^{-3}	b				
		^{238}U	3.2×10^{-3}	b				
		^{235}U	5.6×10^{-9}	a				

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of other countries. I assume that each of the warheads contained 20 kg of ^{239}Pu. With a conversion factor of 0.5 this means that about 3600 tons of natural uranium was used to produce the plutonium. I assume also that in the greater than 1 Mt warheads of total yield of 71 575 Mt half the yield is from the fission of ^{238}U, the burn-out efficiency of which is 40 per cent. This corresponds to 5040 tons of natural uranium. Therefore, the average annual supply of natural uranium for production of warheads between the years 1972 and 1981 was at least 864 tons. From the UNSCEAR data on release of radionuclides during the mining, milling, conversion and enrichment processes [10] it may be inferred that production of this amount of natural uranium introduced into the atmosphere 6.8×10^{-2} Ci of ^{238}U, 3.5×10^{-2} Ci of ^{226}Ra, 9.1×10^{3} Ci of ^{222}Rn, and 2.3×10^{-3} Ci of ^{235}U each year. With particulate emission only 3.0×10^{-2} Ci of ^{210}Pb was introduced, but its activity resulting from ^{222}Rn emission was 5 Ci per year.

Nuclear weapon and space-craft accidents:

A review of few accidents involving nuclear weapons and re-entry of spacecrafts into the atmosphere indicates that small amounts of ^{239}Pu were dispersed in the atmosphere from these sources [2].

Nuclear power

In 1981 the world installed nuclear generating capacity was 144.4 GWe [11], and with an assumed load-factor of 0.6 the energy produced was 86.6 GWe yr. Following UNSCEAR [10], I assume that about 200 tons of natural uranium are required to generate one GWe yr. i.e. 1.7×10^{4} t for electricity generated in 1981.

The values given in Table 1 are calculated from the UNSCEAR [10] data on the atmospheric emissions associated with dust particles from mining, milling, and enrichment operations, corresponding to 1.7×10^{4} t of uranium used in 1981. In addition, 25 Ci of ^{210}Pb associated with decay of gaseous ^{222}Rn are included in the corresponding value in Table 1. The atmospheric emissions of ^{3}H and ^{14}C from power reactors and reprocessing plants are also inferred from UNSCEAR data, as well as emissions of ^{137}Cs and of ^{239}Pu [1,2,10].

Coal burning

In 1980, 3900 million tons of coal were produced [12], 47 per cent of it outside Europe and North America. In various regions of the world different qualities of coal are burned with emission control devices of different efficiency. In the United States, the average atmospheric

emission of particulates was reported to be up to an order of magnitude higher than the recommended emission standards for power generation, which – depending on ash-content in coal – range between 0.5 to 1.0 g of particulates per kg of coal [13]. The efficiency of emission-control devices depends on the size of particles and is in general lower for small particles. The efficiency of electrostatic precipitators for the whole range of particle size is different for various types and in the United States was reported to be as low as 70 per cent in older units while new units may reach 99.8 per cent [14]. In other countries using coal with an apparently higher ash-content, 15 to 40 per cent [15, 16, 17], and less efficient control devices, the emissions from power plants may be as high as 20 g/kg to 35 g/kg [15, 18].

Fewer particulates are emitted in the generation of electricity than in other industrial and household uses of coal. Emissions due to household coal burning were reported to be three times higher than for electricity generation [15]. In 1976 only about 2 per cent of coal in the United States was used for household heating [19], whereas in Poland the figure was 15 per cent [20]. It would be difficult to find out the situation for all countries contributing to the world production and consumption of coal.

For an assessment of the global emission of radionuclides from coal burning, I assume that 50 per cent of coal produced in 1980, i.e. 1.95×10^9 ton, had an ash content of 10 per cent and was burned with a low particle-emission coefficient of 3.3 g/kg [1], i.e. 0.33 per cent of the original mass of coal, or 6.44×10^6 tons, was released into the atmosphere as fly-ash. I assume that the other half of the produced coal had an ash content of 20 per cent and was burned with a particle emission coefficient of 30 g/kg, i.e. 3 per cent of the original mass of coal, or 5.82×10^7 tons entered the atmosphere as fly-ash. From both types of coal-burning then, about 6.5×10^7 tons of fly-ash were released into the global atmosphere in 1980.

Following UNSCEAR 1977 [1], I assume that in the escaping fly-ash the average concentration of ^{238}U is 5 pCi/g; of ^{226}Ra, 1 pCi/g; and of ^{210}Pb, 10 pCi/g. This corresponds to a global flow of these nuclides of 325 Ci U^{238}; 65 Ci ^{226}Ra; and 650 Ci of ^{210}Pb. The activity of ^{235}U associated with ^{238}U flow, calculated from the activity concentrations of these isotopes in natural uranium is about 16 Ci. In addition to the flow of ^{210}Pb associated with particulate matter, this nuclide is produced in the atmosphere from its gaseous parent ^{222}Rn released during the burning and excavation of coal. In coal ^{222}Rn is in radioactive equilibrium with ^{226}Ra, the average concentration of which is 0.54 pCi/g [10]. Being a noble gas, ^{222}Rn is not collected by particulate control devices. In 1980 with the total mass of coal burned, about 2100 Ci of ^{222}Rn escaped into the atmosphere. There is no data available on the emission of ^{222}Rn from coal mines: however, it may be assessed by comparison with the known average emission from uranium mines, which is 5.4×10^{-3} Ci per ton of 0.2 per cent grade ore [10]. This emission is associated with an activity concentration of ^{222}Rn in ore of 540 pCi/g [10], i.e. a factor of about 1000 higher than in coal. Using this factor, one may estimate that with the total mass of coal excavated in 1980, about 21 000 Ci of ^{222}Rn was released into the atmosphere, and the total emission of ^{222}Rn from coal was 23 100 Ci. The activity of ^{210}Pb which originated from this flow of ^{222}Rn is about 11 Ci.

As may be seen in Table 1 the natural annual activity flow of radionuclides into the global atmosphere is several orders of magnitude higher than the corresponding flows from the three anthropogenic sources studied, except for the nuclides ^3H, ^{14}C, ^{137}Cs and ^{239}Pu. The average flows of these four nuclides from nuclear weapons testing and production between 1945 and 1981 dominated the other sources and were by 2 to 6 orders of magnitude higher than the flows from nuclear power in 1981.

The natural flow of ^{222}Rn activity is the highest of the average atmospheric flows of all radionuclides from all sources. The radiation from this nuclide and its daughters contributes the greatest part of the natural radiation-dose received by the population of the world [21]. Therefore ^{222}Rn may serve as a convenient basis for comparison of relative importance of particular flows. The flow of ^{222}Rn from nuclear power in 1981 was a factor of more than 2000 lower than the natural flows and the flow from nuclear weapons a factor of more than 80 000. It is interesting to see in Table 1 that in 1981 the ^{222}Rn flow from coal burning and production was twice as high as from nuclear weapons production.

The flows of other members of ^{238}U family and of ^{235}U from coal burning exceeded the flows from nuclear power and nuclear weapons.

The radiation energies emitted by the radionuclides released into the atmosphere, and listed in decreasing order in Table 2, may help the relative assessment of environmental risk more than comparison of activity flows. The energy emitted by disintegration of radionuclides released is calculated from data given in [22].

As may be seen in Table 2 the energy released from natural sources is dominated by ^{222}Rn and is 4 to 5 orders of magnitude higher than from all anthropogenic sources studied.

Only in the case of nuclear weapons is ^{222}Rn not the main contributor to the emission of radiation energy from atmospheric flows of radionuclides, being third after ^3H and ^{137}Cs. Both in the case of nuclear power and of coal burning, the energy released into environment from ^{222}Rn is 2 orders of magnitude higher than from ^3H and ^{210}Pb. It seems therefore that ^{222}Rn deserves greater interest and study than might have been suggested by assessments based on dose-commitment calculations, which indicate that the impact of nuclear

power production due to releases of ^{14}C would be much higher than due to other radionuclides [1]. As may be seen in Table 2 the current radiation environmental impact of nuclear energy consists a very small fraction of the natural impact and it does not seem possible that in the foreseeable future the flows of radionuclides into the global atmosphere from nuclear power will approach the natural levels.

The comparison of activities and radiation energies released into the global environment from four sources, gives a different perspective from a comparison based on the more anthropocentric and less parsimonious concept of tissue-absorbed dose or of dose-committment. I hope that it may be helpful for the public and decision-makers perception of radiation risk involved in large-scale activities of man.

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