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Fate of fall-out plutonium and americium in the environment: selected examples

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Abstract

At present, several isotopes of Pu and Am are in the biosphere as a result of above-ground nuclear weapons tests in the 1950s and 1960s. Even though their concentrations in most environmental samples are extremely low, they can be determined if methods are developed which allow convenient disintegration of very large samples before radiochemical analysis. In this way, information on the long-term behavior of actinides in the biosphere can be obtained under ideal natural conditions. The selected examples discussed here involve investigations on the following: (1) the filter effect of coniferous trees for Pu and Am during their dry deposition; (2) the rate of vertical migration of Pu and Am in the different soil horizons of forest and grassland soils; (3) the transfer of Pu and Am from soil to flour and bran from cereals; (4) the enrichment of Pu in some types of honey; (5) Pu and Am in the tissues of the general population in Germany.

1. Introduction

As a result of above-ground nuclear weapons tests, or of accidents at installations of the nuclear industry, actinides can be released into the environment. To estimate realistically the long-term radiation doses to humans, a detailed knowledge of the fate of actinides in the biosphere is necessary. Because actinides cannot be released deliberately into the biosphere to study their environmental behavior, information on the radioecology of actinides is obtained frequently from greenhouse experiments. Even though these investigations offer the advantage of controlled environmental conditions, it is very difficult to ensure in this way realistic environmental conditions, especially for long-term investigations, which have to be carried out for several years.

For this reason, it is useful also to study extensively the behavior of those actinides which have been in the biosphere for many years, resulting from the worldwide fall-out from above-ground nuclear weapon tests. Up to 1978, 0.79 PBq of ^{238}Pu (from which 0.51 PBq was released from the burn-up of the nuclear-powered satellite SNAP-9A in 1964) and 13 PBq of $^{239+240}$Pu were produced in this way [1–3]. Similar to ^{238}Pu and $^{239+240}$Pu, the radionuclide ^{241}Am found in our environment is also a long-lived alpha emitter, which results mainly from the decay of fall-out ^{241}Pu (beta emitter; $t_{1/2}=14.4$ years). By this route, 3.1 PBq of ^{241}Am had been produced by 1978 and, eventually, 5.5 pBq will be formed [1].

Because these fall-out actinides were deposited fairly evenly throughout the Northern hemisphere, the resulting activity concentration in an environmental sample (such as in soil, plant or food) is extremely low. Nevertheless, if sufficiently large samples are processed and analysed, the above actinides can be determined in many environmental samples. As a result, valuable information, especially on the long-term behavior of Pu and Am under ideal natural conditions, is obtained.

2. Experimental details

The determination of fall-out actinides in environmental samples usually involves the following steps: the addition of the yield tracers ^{242}Pu and ^{243}Am, and disintegration of the sample; radiochemical separation of Pu and Am from other elements in the sample; electroplating of Pu and Am on separate stainless steel planchets; the determination of Pu and Am by alpha spectrometry.

As mentioned, especially for those samples where the activity concentrations of the actinides are extremely low, sample sizes up to 100 kg have to be disintegrated for one analysis to obtain reasonably small analytical errors. For the wet disintegration of cereals, for example, we developed a special procedure which employs initial dissolution of the starch by alpha amylase at 50 °C, saccharification of starch by amyloglucosidase at 55–60 °C, fermentation by yeast below 35 °C and, finally, wet ashing of the residue by hydrogenperoxide + nitric acid

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(for details, see refs. 4 and 5). Blank determinations showed that the introduction of actinides by the reagents used was always negligible.

3. Results and discussion

3.1. Deposition of fall-out Pu and Am in forests and grasslands

Because the actinides in the global fall-out are attached to aerosol particles, it is conceivable that, as a result of the filter effect of trees, a higher deposition of these radionuclides may occur in forests as compared with on grasslands. Subsequent to the deposition of radioactive aerosols on tree leaves, needles or grass, most of the actinides are transferred over the years by rain and leaf-fall from the plant to the ground. Because these radionuclides are accumulated in the upper layers of the soil (see below), the cumulative deposition of fall-out actinides in an area since the beginning of the fall-out in the 1950s can be quite accurately estimated by determining their total activity per square meter of soil. To detect how large the enhanced deposition of fall-out actinides in forests will be, we determined at 25 sites in southern Bavaria, Germany, $^{239+240}$Pu and ^{241}Am in the soil of spruce forests and adjacent grassland [6]. Because the extent of the long-term deposition of a fall-out radionuclide may depend also on the mean annual precipitation, the precipitation was also evaluated at each site.

The results of these measurements are shown in Fig. 1, where we have plotted the cumulative deposition of $^{239+240}$Pu as a function of the mean annual precipitation, separately for the forest soils and the grassland soils. The graph reveals that the cumulative deposition of fall-out Pu increases for forest and grassland sites with increasing precipitation. This correlation is also statistically highly significant (Spearman correlation coefficient; $p<0.001$, two tailed). Calculation of the linear regression line yields

$$^{239+240}\text{Pu (Bq m}^{-2}) = 32.4 + 0.034 \times \text{mean annual precipitation (mm)}$$

for the forest soil and

$$^{239+240}\text{Pu (Bq m}^{-2}) = 12.3 + 0.039 \times \text{mean annual precipitation (mm)}$$

for the grassland soil.

Even though the values for the slopes of the two regression lines differ slightly from the above equations, they are actually not different in a statistically significant way. (This can be shown by a test of the comparison of two regression coefficients at the 0.05 level.) However, there is a significant displacement between the two parallel regression lines. This difference for the cumulative deposition of $^{239+240}$Pu in spruce stands and grassland sites is 14 Bq m^{-2} on average (median), with a 95% confidence limit of 10–17 Bq m^{-2}. If we calculate the average cumulative $^{239+240}$Pu deposition for all grassland sites, we obtain 49 Bq m^{-2} for the median. Thus, in spruce stands, the long-term cumulative deposition is increased by about 30%. A very similar result is also observed for fall-out ^{241}Am. The corresponding values for this element can be obtained by dividing all the values given above for $^{239+240}$Pu by 3.4.

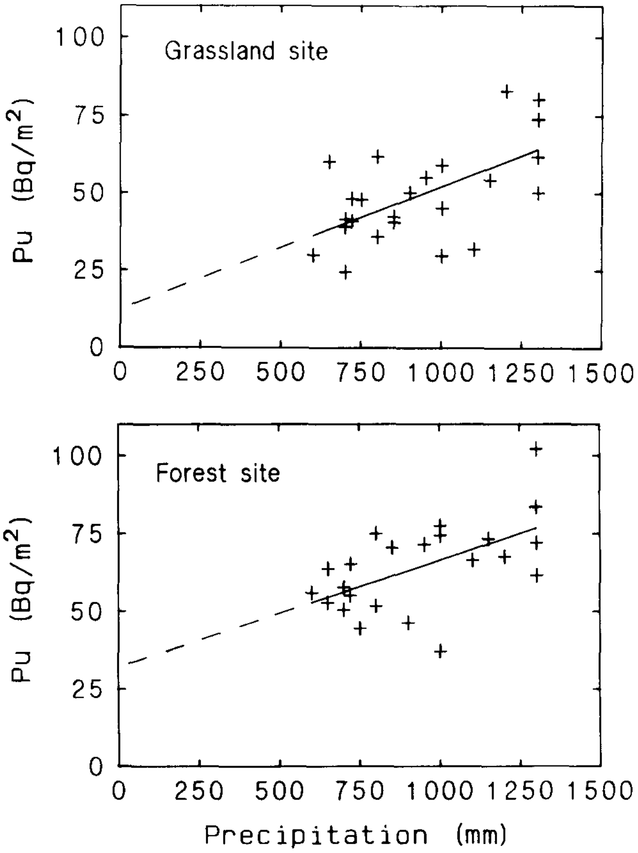

Fig. 1. Cumulative deposition of $^{239+240}$Pu from global fall-out in forest and grassland soils in Southern Bavaria, Germany, as a function of the mean annual precipitation.

3.2. Migration of fall-out Pu and Am in soils

Quantitative information on the long-term rates of the vertical migration of actinides in a soil can be obtained by evaluating the depth profiles of these radionuclides observed at various times after their deposition on the soil surface by the fall-out event. For this purpose, soil samples were taken in layers about 2 cm thick to a depth of about 40 cm at various sites in a spruce forest [7] (a podzolic parabrown earth soil; Hapludult) and in a grassland [8] (slightly wet Alfisol). At depths greater than 40 cm, fall-out actinides are not yet observable in these soils. These depth profiles reveal that the major part of the activity deposited in the 1950s and the 1960s on the soil surface is found for the forest soil in the upper layers of the mineral horizon (5–8 cm in depth below the surface). In the grassland soil, fall-out Am and Pu were essentially observed in the Ah horizon about 7–10 cm below the surface. In both soils, however, these actinides are also found to some extent in the other soil layers sampled.

To obtain the vertical rates of migration of Pu and Am from the measured depth profiles in each soil layer, we used a compartment model [9]. The results obtained in this way for the soils investigated are given in Table 1. The results demonstrate that the vertical transport for both actinide elements is very slow (in the order of about 1 cm per year) and quite similar, even though the mobility of Am in the forest soil is somewhat higher than that for Pu [10, 11]. Although there is a considerable spatial variability, the migration rates increase with increasing depth in the mineral horizon. The reason for this behavior is not known as yet. Because the mechanisms of the transport of actinides are not well understood, and because the fall-out actinides are still mainly in the upper soil layers only, it is currently not possible to predict with confidence whether or not the increase in the migration rates with depth will continue also in even deeper soil layers. In recent years, however, because of increasing interest in the geochemical behavior of disposed radionuclides, the hydrolysis and interactions of actinides with soil components has been studied in more detail (see, for example, refs. 10–12). These results should be very useful also for a better understanding of the migration behavior in the upper soil layers.

TABLE 1. Long-term rates of vertical migration (in cm per year) of fallout $^{239+240}$Pu and ^{241}Am in forest soil and grassland soil as a function of depth

Depth (cm)	$^{239+240}$Pu	^{241}Am
Forest soil		
0–2	>0.7	>0.7
2–3.5	>0.5	>0.5
3.5–5	0.2	0.3
5–7	0.08	0.1
7–10	0.25	0.3
10–15	0.7	2
Grassland soil		
0–2	0.3	0.3
2–5	0.4	0.4
5–10	0.5	0.5
10–15	0.7	0.5
15–20	1	1
20–30	1	1

3.3. Transfer of fall-out Pu and Am from soil to cereals

Even though details of a number of greenhouse and field experiments on the contamination of cereals are available, the transfer of actinides has been studied mostly for wheat and barley, and, in general, only for the whole grain. However, usually only the milled grain, *i.e.* either the flour or the bran of a given cereal, is processed for foodstuffs. Because very few data are available for fall-out Pu and Am, in particular, we determined the soil-to-plant transfer separately in the flour and bran fractions of wheat, rye, barley and oats [4]. To ensure the most realistic conditions possible, the crops were grown in fields where the soil was contaminated only with radionuclides from global fall-out. The crops were machine harvested using a combine, and milled in a commercially operated flour mill. To minimize surface contamination of the grains by re-suspended soil particles, the grains were washed before milling with water.

For comparison, in all the soils and cereal samples, we also determined the concentrations of fall-out ^{137}Cs and ^{90}Sr, as well as those of the stable elements K, Ca and Fe. To characterize the soil-to-plant transfer of each radionuclide, we calculated the soil-to-plant transfer factors (TF), defined as

$$\mathrm{TF} = \frac{\text{Bq per kg air dry plant material}}{\text{Bq per kg air dry soil}}$$

The values obtained in this way are shown in Fig. 2. As can be seen, the TFs for the brans are considerably higher than those of the flours, with the exception of oats, where the TFs are quite similar. It is also evident that the TFs for the flours and brans increase in the sequence

$$\mathrm{Am} < \mathrm{Pu} \ll \mathrm{Cs} \ll \mathrm{Sr}$$

Thus, the amount of transfer of the actinides from the soil to the plant is very small for the actinides, especially that to the flours.

The observed difference in the TFs for the flours and brans is illustrated also in Fig. 3, where we have plotted for each cereal and each radionuclide the ratio of the activity concentration in bran to that in flour. For comparison, we have also included in Fig. 3 the corresponding values observed for the stable elements K, Ca and Fe. The ratio reveals that, for these stable elements, a similar enrichment in the brans occurs to that in the flours.

The close analogy in the chemical and metabolic behaviors of ^{137}Cs and K, and those of ^{90}Sr and Ca has been observed in many other cases, and is also obvious in the present investigation. In contrast, Fe exhibits bran/flour ratios which are more similar to those of the actinides (see Fig. 3). At present, not

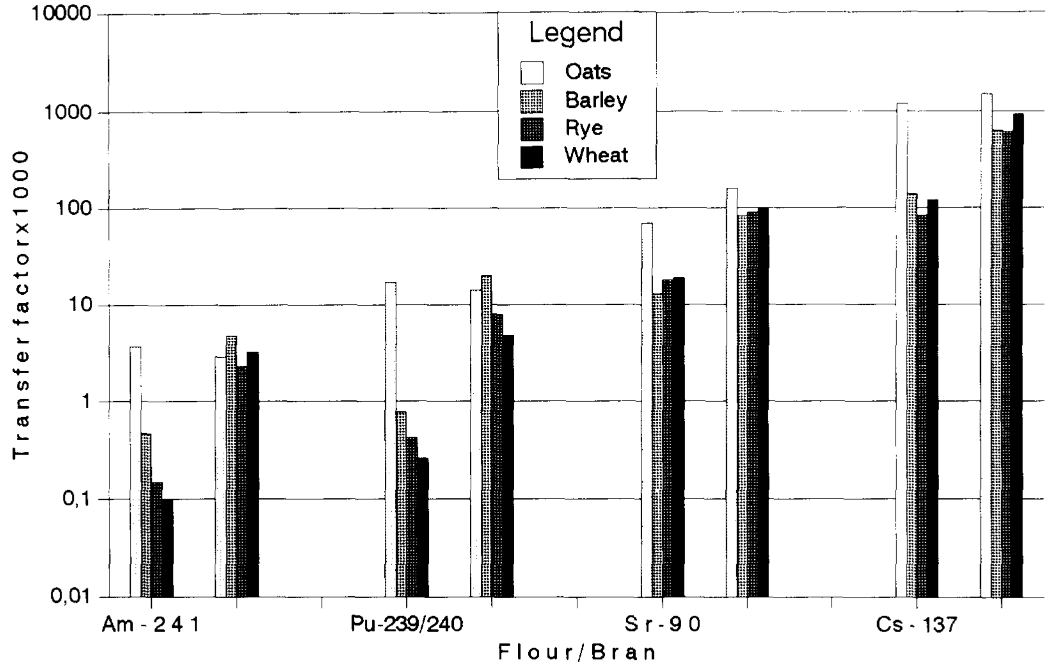

Fig. 2. Soil-to-plant transfer factors for flour and bran of oats, barley, rye and wheat. These cereals were grown on soils which were contaminated only by the global fall-out of above-ground weapons tests in the 1950s and 1960s.

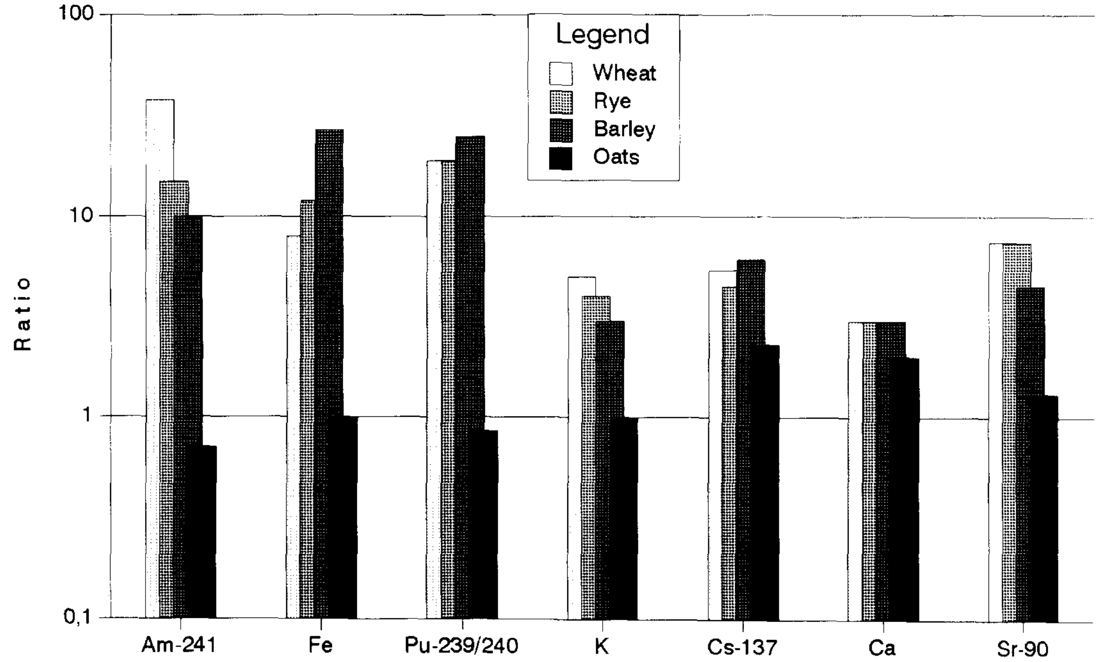

Fig. 3. Ratio of the activity concentrations of several radionuclides from the global fall-out in bran to those in flour for wheat, rye, barley and oats. The enrichment of some elements in the bran fraction is also observed for stable elements, such as K, Fe and Ca.

enough data are available to demonstrate that Fe is a suitable analog for the distribution of actinides in plants, but it might be worthwhile to include this element in future radioecological investigations on actinides.

3.4. Fall-out Pu in different types of honey

Information on the concentration of actinides in honey is of interest, not only because this foodstuff is consumed worldwide but also because it is used as a bioindicator for radionuclides. Thus, it is of interest to know when – and if so to what extent – the concentrations of radionuclides in the honey depend on the type of honey sampled by the bees. For this reason, we determined fall-out $^{239+240}$Pu, ^{137}Cs, ^{90}Sr and stable K in three important types of honey produced in Germany, namely honey from flowers, honey from honey dew and that from heather [13]. Because, as shown above, the extent of global fall-out in an area depends on the mean annual precipitation, the sampling areas were selected accordingly, to eliminate this effect. Again, for analytical reasons, large quantities (10 kg) had to be processed to determine the very low concentrations of fall-out $^{239+240}$Pu in this material. Up to 10 samples of each type of honey were analysed. The results are illustrated in Fig. 4, where we have plotted the average (median) activity concentrations of the above radionuclides as well as of stable K.

The data reveal that considerable differences in the activity concentrations of a radionuclide may arise for different types of honey. Especially low concentrations were always observed for honey from flowers. In comparison, honey from honey dew and honey from heather exhibited considerably higher concentrations (about one order of magnitude) for $^{239+240}$Pu, ^{137}Cs and ^{90}Sr. These differences are also statistically significant (*U*-test by Mann-Whitney). Seasonal variations of the radionuclide concentrations within three successive years were observed for the fall-out radionuclides, but the variations were small in comparison with the differences observed for different types of honey. The differences in the concentration of K in the different types of honey were comparatively small. The fairly high concentration of ^{137}Cs in honey from heather can be explained by the high root uptake of the perennial heather plants, which accumulate this radionuclide. The reason for the increased concentration of the radionuclides in honey from honeydew is not yet known. The above observations clearly demonstrate that, before a honey sample can be used as a bioindicator, its type also has to be determined, for example, by pollen analysis.

3.5. Fall-out Pu and Am in humans

Since the beginning of the period of global fall-out in the 1950s, various Pu isotopes have been taken up by the general population of the Northern hemisphere. The main route of uptake has been inhalation of the radioactive aerosols. Because, as mentioned above, ^{241}Am in the environment (and in humans) is produced mainly by the decay of fall-out ^{241}Pu, we determined in the human tissues of the general public not only $^{239+240}$Pu and ^{238}Pu but also the beta emitter ^{241}Pu.

The subjects selected (47 males from Munich) ranged from 20 to 79 years of age, and it was established that they were all victims of a sudden death. Because of the very low concentrations of actinides presently in humans, the entire lung, the entire liver as well as 300 g of vertebrae and 300 g of ribs were taken from each person and analysed [14].

The results (medians) of the $^{239+240}$Pu determinations in these tissues are shown in Fig. 5. They demonstrate that, at present, the lowest Pu concentrations are found

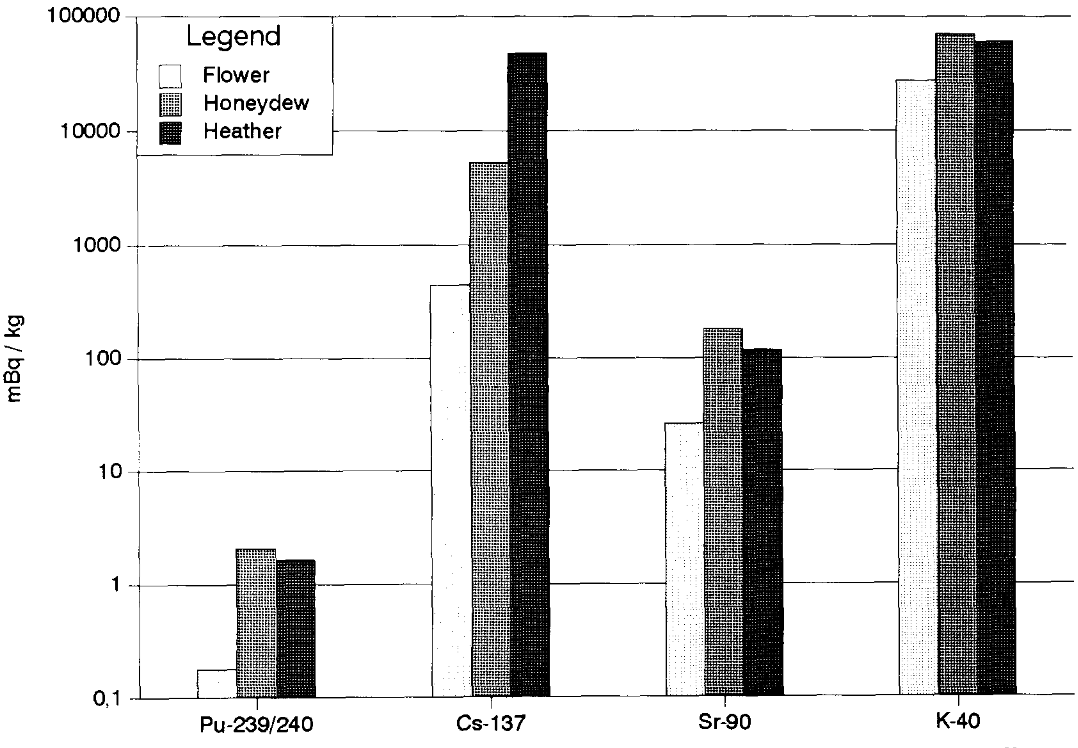

Fig. 4. Activity concentrations of several radionuclides from global fall-out ($^{239+240}$Pu, ^{137}Cs and ^{90}Sr), as well as those of ^{40}K in different types of honey (honey from flowers, honey from honey dew and that from heather). ^{40}K is a natural radionuclide, whose activity is proportional to the content of stable K in an environmental sample.

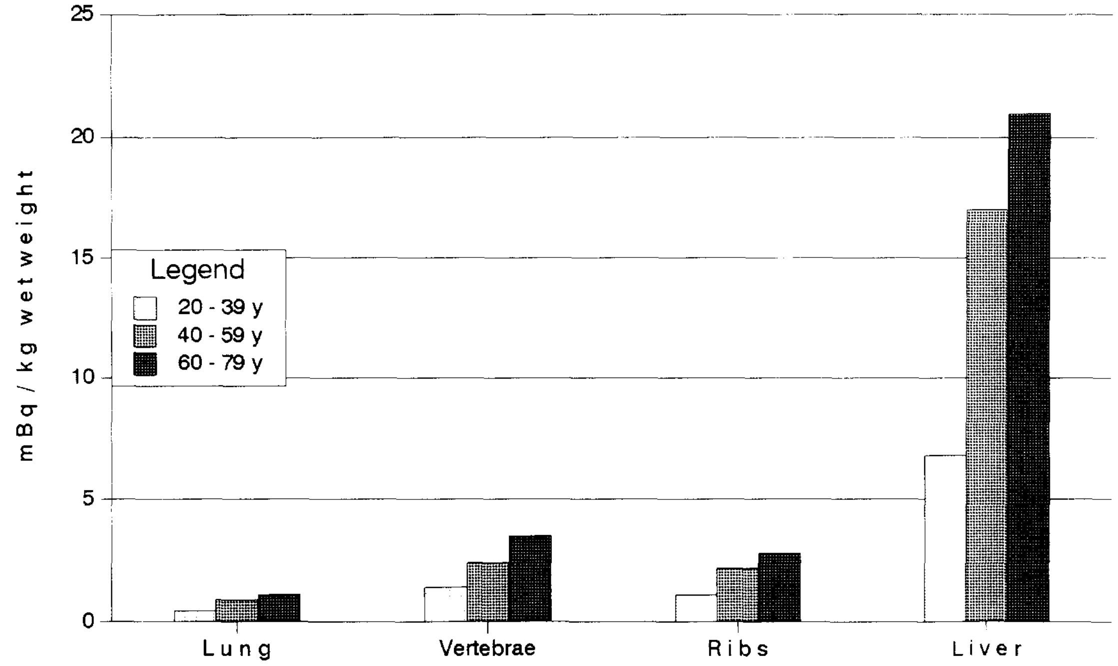

Fig. 5. Activity concentrations of $^{239+240}Pu$ from global fall-out in the lungs, bones and livers of the general population (males) in Munich, Germany, as a function of the age of the people.

in the lungs and the highest in the livers. In addition, for each tissue, the Pu concentration increased with age. This correlation was also highly statistically significant for all tissues (Spearman correlation coefficient; significance level 99.9%, two tailed). The frequency distributions of the data were mostly log-normal. The observed ratios for $^{238}Pu/^{239+240}Pu$ were between 0.03 and 0.05 for all tissues, while those for $^{241}Pu/^{239+240}Pu$ were between 4 and 7. These values are characteristic of the Pu isotopes of the global fall-out of weapons testing. Because the corresponding ratios in the emissions of nuclear power plants are much higher, the above values demonstrate that the origin of the Pu found presently in humans is indeed only the global fall-out.

While these observations are in good agreement with measurements in other countries [15–20] and support the metabolic model of the ICPR [21], much less information is available on the activities of fall-out ^{241}Am in the tissues of the general population. For this reason, we determined ^{241}Am in 29 human livers [22], where, similar to Pu, the highest concentrations can be expected. On average (median), we observed in this tissue 2.3 mBq per kg wet weight. A strong positive correlation was also observed for the concentrations of ^{241}Am and $^{239+240}Pu$ in the livers (Spearman correlation coefficient; significance level 99.9%, two tailed). The median ratio $^{241}Am/^{239+240}Pu$ in the livers was 0.133 on average. If one uses again the metabolic model of the ICRP [21] to compare the experimental results with predicted values, it seems that the model overestimates the activity concentrations in the liver.

4. Conclusions

The activity concentrations of fall-out Pu and Am in environmental samples are usually very low. Nevertheless, if sufficiently large samples are analysed, quantitative information on the long-term behavior of these elements in the biosphere can be obtained and compared with model calculations. In addition, the data can be used to establish the present background levels of Pu and Am in environmental samples, and may serve to detect possible contributions of emissions from the nuclear industry with high sensitivity, at a time before health hazards are observable.

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