

# Radionuclides in the Environment in Switzerland: A Retrospective Study of Transfer from Soil to the Human Body

Pascal Froidevaux\*, Pierre-André Pittet, Ruslan Cusnir, François Bochud, and Marietta Straub

**Abstract:** Natural radionuclides are ubiquitous in the environment. In addition, artificial radionuclides are present in the Swiss environment after the fallout of the nuclear bomb tests of the 1950s and 1960s, after the accident of the Chernobyl nuclear power plant, or after authorized discharges from the Swiss nuclear power plants and research centres. These radionuclides can create a radiological hazard to the environment and humans because of the increased risk of cancer due to the ionizing radiation they produce. Here we show that some of these radionuclides have made their way from the air or the soil to the human body, where they target mostly the skeleton. However, the activity levels of  $^{90}\text{Sr}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Pb}/^{210}\text{Po}$  found in the human body remain very low and do not represent a public health issue at the current body burden.

**Keywords:** Environment · Human body · Radiation dose · Radionuclides · Transfer factors

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## 1. Introduction

Natural and artificial radionuclides are unstable chemical elements which disintegrate with emission of ionising radiation.  $\gamma$ -Radiation is the most studied because, as a photonic radiation, it can be measured directly with dedicated instruments.  $^{137}\text{Cs}$  produced during nuclear fission is a typical  $\gamma$ -emitting radionuclide that has been widely studied in the context of nuclear accidents such as Chernobyl and Fukushima.<sup>[1]</sup> On the other hand,  $\beta$ -decay only produces electrons (and antineutrinos). These electrons are difficult to measure and  $\beta$ -emitting radionuclides require a complex chemical separation to reach satisfactory detection limits during analysis. This is why  $\beta$ -emitting radionuclides are monitored to a lesser extent in the framework of a radiation survey plan. However,  $\beta$ -emitting radionuclides such as  $^{90}\text{Sr}$ , also a result of nuclear fission, can be very damaging to human because of their high mobility in the environment.<sup>[2]</sup> As an alkaline-earth cation

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similar to calcium,  $^{90}\text{Sr}$  targets bones and deposits its energetic  $\beta$ -radiation in the proximity to the bone marrow.<sup>[3]</sup> In this context,  $^{90}\text{Sr}$  should be systematically determined where nuclear fission is of concern.  $\alpha$ -Radiation is the most damaging radiation for living species. The dose factor for  $\alpha$ -radiation is typically 20 times higher than for a  $\gamma$ -radiation. The  $\alpha$ -particle deposits its energy within a few  $\mu\text{m}$  from the source point, creating numerous damaging ionisations along the track.  $\alpha$ -Emitting radionuclides represent a significant threat for human tissues through inhalation or ingestion.  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  belong to radionuclides from the  $\alpha$ -emitters family. They are also difficult to determine for reasons similar to those of  $\beta$ -radiation. Natural radionuclides such as  $^{238}\text{U}$ ,  $^{235}\text{U}$  or  $^{232}\text{Th}$  are also  $\alpha$ -emitters. During disintegration, they give off a series of unstable daughter nuclides, either  $\beta$ -,  $\gamma$ - or  $\alpha$ -emitters. Each series contains at least 20 radioactive members, which decay consecutively to a final stable nuclide – an isotope of lead.

The danger represented by a radionuclide is not only related to its decay mode,  $\alpha$ -emitters being more deleterious than  $\beta$ - and  $\gamma$ -emitters, but mostly to its capability to be transferred from the soil, where it is deposited, to the human food chain.<sup>[2e,4]</sup> In this respect, the chemical properties of a given radionuclide are more important than its decay mode. Radionuclides that have similar chemical properties to those of stable elements involved in biological metabolism can cross biological membranes.<sup>[2c]</sup> For instance,  $\text{K}^+$  is an alkaline cation of importance in the living species metabolism. Thus,  $^{137}\text{Cs}$ , as an alkaline cation, is able to cross biological membranes as a side effect of K-metabolism, even if Cs has no significant role in the metabolism. In humans, K is mostly found in muscles, as  $^{137}\text{Cs}$  will be. One of the most radiotoxic elements from the nuclear industry is  $^{90}\text{Sr}$ . As an alkaline-earth cation, it follows Ca all along the Ca metabolism, targeting bones. Likewise, natural  $^{226}\text{Ra}$  and its progeny  $^{210}\text{Pb}$  behave in a similar way.<sup>[5]</sup> Even if  $\text{Pb}^{2+}$  is not directly related to the alkaline-earth cation family, its biokinetics in human is very similar to  $\text{Ca}^{2+}$ .<sup>[6]</sup> Once incorporated into the bone structure as  $\text{Ca}(\text{Pb})$ -hydroxyapatite, it decays to  $^{210}\text{Bi}$ , a very energetic  $\beta$ -emitter, and to the very radiotoxic  $\alpha$ -emitter  $^{210}\text{Po}$ .<sup>[3c,d,7]</sup> In this study, we show that  $^{90}\text{Sr}$ , deposited in the Swiss environment because of the nuclear bomb tests fallout, is easily transferred from soil to plants, to milk and human bones, following the Ca metabolism pathway. Similarly,  $^{90}\text{Sr}$  found in milk teeth, which form already *in utero* during pregnancy, shows that  $^{90}\text{Sr}$  can cross the placental barrier.<sup>[3a,b,8]</sup> We also were able to quantify  $^{226}\text{Ra}$  and  $^{210}\text{Po}$  in human vertebrae, because Ra is a member of the alkaline-earth cations family, and  $^{210}\text{Po}$  is the progeny of  $^{210}\text{Pb}$ , an element with similar metabolism as Ca. On the other hand, we demonstrate that Pu cannot cross the placental barrier, because we did not find significant quantity of the Pu present in air during the fallout of the nuclear bomb tests in milk teeth formed during pregnancy. Following Pu deposition and integration in soils, no transfer to the food chain and human was observed and no Pu was found in human vertebrae of persons born after 1967.

## 2. Experimental

All the samples measured in this study come from the National Radioactivity Survey Plan of the Swiss Federal Office of Public Health and have been analysed for this purpose. From 1971 to 2004,  $^{90}\text{Sr}$  has been determined as described by Froidevaux *et al.*<sup>[3b]</sup> From 2004 to 2010,  $^{90}\text{Sr}$  has been determined as described by Guillaume *et al.*<sup>[2e]</sup> and, starting from 2010, with a specific ion-imprinted polymer as described by Chauvin *et al.*<sup>[9]</sup> and Froidevaux *et al.*<sup>[10]</sup>  $^{239+240}\text{Pu}$  was determined by alpha spectrometry as described in Luisier *et al.*<sup>[11]</sup>  $^{239}\text{Pu}$  in bone and teeth was determined by mass spectrometry as described by Froidevaux and Haldimann<sup>[8]</sup> and Froidevaux *et al.*<sup>[3a]</sup>  $^{226}\text{Ra}$  in bones was determined as described by Straub *et al.*<sup>[5]</sup>  $^{210}\text{Po}$  was determined in vertebrae as described by Froidevaux *et al.*<sup>[7]</sup>

Transfer factors are expressed as the ratio of compartment 1 (in a given unit) to the compartment 2. Eqns (1) to (3) show the different expression for  $^{90}\text{Sr}$  transfer factor from soil to grass, grass to milk and milk-to-milk teeth and bones.

$$TF_{\text{soil-grass}} = \frac{A_{\text{grass}} \left[ \frac{\text{Bq}}{\text{kg}} \right]}{A_{\text{soil}} \left[ \frac{\text{Bq}}{\text{kg}} \right]} \quad (1)$$

$$TF_{\text{grass-milk}} = \frac{A_{\text{milk}} \left[ \frac{\text{Bq}}{\text{g Ca}} \right]}{A_{\text{grass}} \left[ \frac{\text{Bq}}{\text{kg}} \right]} \quad (2)$$

$$TF_{\text{milk-milk teeth}} = \frac{A_{\text{milk-teeth}} \left[ \frac{\text{Bq}}{\text{g Ca}} \right]}{A_{\text{milk}} \left[ \frac{\text{Bq}}{\text{g Ca}} \right]} \quad (3)$$

## 3. Results and Discussion

### 3.1 $^{90}\text{Sr}$ Transfer from Soil to Human

$^{90}\text{Sr}$  is present in the Swiss environment mostly because of the nuclear bomb tests fallout of the 1960s and marginally as the consequence of the Chernobyl accident. Results of numerous  $^{90}\text{Sr}$  analyses from the 1960s up to now show that  $^{90}\text{Sr}$  was transferred from the soil to the human food chain and to the skeleton.<sup>[3a-c]</sup> Fig. 1 displays the results obtained in Switzerland for soil, grass, wheat, milk, milk teeth and vertebrae. Each single value in Fig. 1 is the average of the yearly determination of about 20 soil samples, 20 grass samples collected above the soil samples, 15 wheat samples, 20 milk samples, 12 vertebrae from 12 different individuals collected mostly by the Lausanne and the Lugano pathology institutes and 12 milk teeth samples. A milk tooth sample is formed by merging at least 10 teeth from children born the same year.<sup>[3b]</sup> The main feature of Fig. 1 shows that  $^{90}\text{Sr}$  has decreased from 1965, after the nuclear test ban treaty, in all the measured compartments with the same biological half-life, close to  $13.1 \pm 2$  y. The soil is the compartment that directs the mechanism of transfer to the food chain. The biological half-life of  $^{90}\text{Sr}$  in the root zone (rhizosphere) of soil is 12.6 y. It follows that the biological half-life of the other compartments is linked to that of soil.

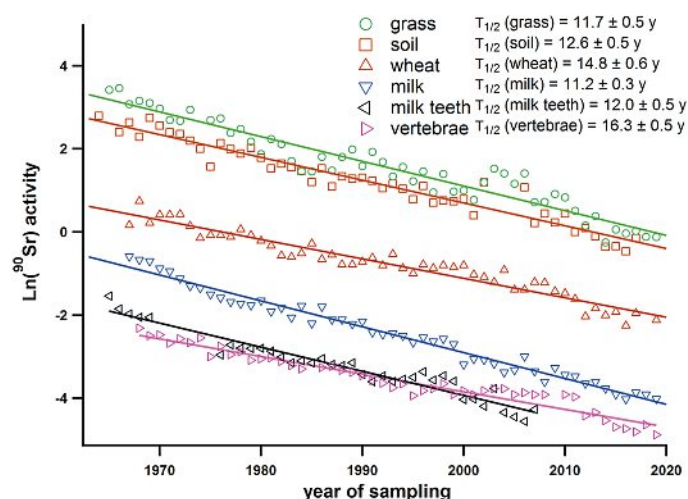


Fig. 1. Logarithm of the  $^{90}\text{Sr}$  activities, in  $\text{Bq kg}^{-1}$  for soil, grass and wheat and in  $\text{Bq g Ca}^{-1}$  for milk, milk teeth and vertebrae, and related biological half-life for the environment of the Swiss Tableland.

Because the half-lives are the same for all measured compartments, it is possible to calculate the transfer from one compartment to another with the concept of the transfer factor. The transfer factor (TF) links two compartments with a simple relationship. It does not reveal the geochemical mechanism or biological mechanism of transfer. Nevertheless, it allows a rapid evaluation of the possible consequence of a radioactive contamination for a given radionuclide if sufficient data are available to produce a histogram of the TF values. For instance, the histogram of TF values for  $^{90}\text{Sr}$  from soil to grass in Switzerland based on the data of Fig. 1 ( $n = 53$ ) shows that the values are centred on  $\text{TF} = 1.3$ . Table 1 gives the TF values for  $^{90}\text{Sr}$  from soil to human, based on our data set of values collected from 1965 to now. TF values less than 1 for the transfer from milk to milk teeth and vertebrae show that the human metabolism makes a preference for Ca over Sr; the latter having no known role in human metabolism.

Table 1. TF values for  $^{90}\text{Sr}$  in the Swiss environment. Compartments taken into account are soil, grass, wheat, milk, milk teeth and vertebrae. TF are evaluated as histogram values based on data of Fig. 1 ( $n = 53$ ).

| Compartments                  | TF value $\pm u(k = 1)$              |
|-------------------------------|--------------------------------------|
| soil $\rightarrow$ grass      | $1.3 \pm 10\%$                       |
| soil $\rightarrow$ wheat      | $0.15 \pm 25\%$                      |
| grass $\rightarrow$ milk      | $0.02 \pm 20\% \text{ kg g Ca}^{-1}$ |
| milk $\rightarrow$ milk teeth | $0.33 \pm 25\%$                      |
| milk $\rightarrow$ vertebrae  | $0.35 \pm 25\%$                      |

Fig. 2 shows the pulse of  $^{90}\text{Sr}$  activity measured in Switzerland during and after the fallout of the nuclear bomb tests of the 1950s and 1960s. The activity profile of  $^{90}\text{Sr}$  in milk teeth matches perfectly the one for milk. Considering that the enamel of milk teeth forms during pregnancy and during breast-feeding, these results demonstrate that  $^{90}\text{Sr}$  has crossed the placental barrier.<sup>[3b,8]</sup> The  $^{90}\text{Sr}$  profile in human vertebrae also matches the one of milk, but with a delay of about 2 years. This delay is the result of the bone-remodelling rate, which recirculates the Ca in the organism, incorporating new  $\text{Ca}^{(90}\text{Sr)}$  in the bone structure. The bone-remodelling rate is dependent on age, bone functionalities and varies between individuals. It is faster at younger ages and for trabecular bones such as vertebrae. While our database contains samples from individuals deceased mostly after 60 years (80%) and none who died before 8 years, the results show that  $^{90}\text{Sr}$  was readily incorporated into the bone structure.<sup>[3a]</sup> Similar to milk teeth,  $^{90}\text{Sr}$  is rapidly incorporated into the skeleton of the foetus and newborns. Thus, our data show that  $^{90}\text{Sr}$  is very mobile in the environment, easily transferred to the food chain, and reaches through placental

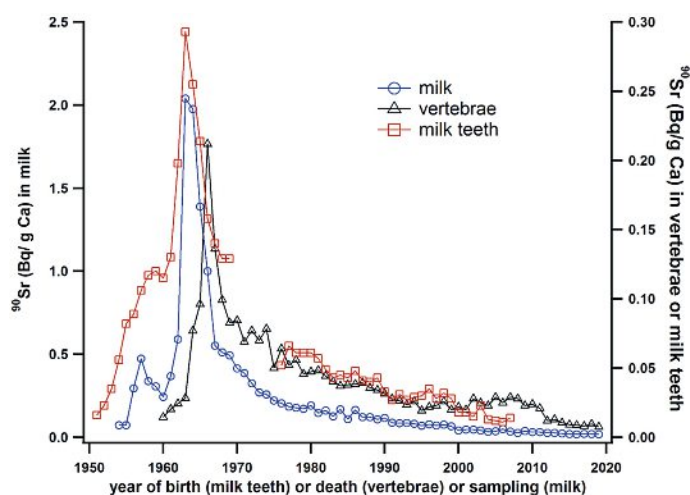


Fig. 2.  $^{90}\text{Sr}$  activity profiles in milk, milk teeth and vertebrae in Switzerland from the beginning of the nuclear era to 2019. The activities of milk teeth are reported to the year of birth and the activities of vertebrae to the year of death.

transfer the skeleton and milk teeth of the foetus. In this respect, pregnant women should be particularly protected against this particular radionuclide.

### 3.2 $^{226}\text{Ra}$ and $^{210}\text{Po}$ Transfer to Humans

$^{226}\text{Ra}$  and  $^{210}\text{Po}$  belong to the decay series of  $^{238}\text{U}$ , which is present in soil at an average activity of  $30 \text{ Bq kg}^{-1}$ . However, it can be accumulated in wetlands to exceptional values above  $100'000 \text{ Bq kg}^{-1}$ .<sup>[12]</sup>  $^{226}\text{Ra}$  is the seventh member of the series and has a long half-life of 1602 y. As an alkaline-earth cation, it is rather mobile and, in a way similar to Sr, it can be transferred from soil to human.  $^{226}\text{Ra}$  has been used in the Swiss watch industry and can be present in excess in the Swiss environment as a radioactive heritage of the past. In 2015, the Swiss Federal Office of Public Health launched a program of rehabilitation of contaminated sites in the Jura, Soleure, Neuchâtel and Bienne areas.<sup>[13]</sup> In this context, it appeared important to establish a database of  $^{226}\text{Ra}$  values in bones, using samples collected in the framework of the National Radioactivity Survey Plan for  $^{90}\text{Sr}$  measurement. Results of the  $^{226}\text{Ra}$  analyses on 33 vertebrae sampled from 2014 to 2019 showed that the accumulated activities are low, with an average of  $1.82 \text{ mBq g Ca}^{-1}$ , a minimum value at  $0.73 \text{ mBq g Ca}^{-1}$  and a maximum value at  $4.58 \text{ mBq g Ca}^{-1}$  (Table 2). These values are within or close to the worldwide average value of  $0.37\text{--}4.0 \text{ mBq g Ca}^{-1}$ .<sup>[14]</sup>

$^{210}\text{Po}$  is also a member of the  $^{238}\text{U}$  decay series. Its chemistry is more related to the metalloids rather than to metals.  $^{210}\text{Po}$  bears a high charge density (+4) leading to fast hydrolysis, even at low pH. In this respect, it is relatively immobile in soil and not easily transferred to the food chain. It has a half-life of 138.4 days and thus, is mostly supported by  $^{210}\text{Pb}$  ( $T_{1/2} = 22.3 \text{ y}$ ). Secular equilibrium between  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  means that the presence of

Table 2.  $^{226}\text{Ra}$ ,  $^{210}\text{Po}$  and  $^{90}\text{Sr}$  activities ( $\text{mBq g Ca}^{-1}$ ) in vertebrae of the Swiss population (Tessin and Vaud regions) between 2010 and 2019.

| Radionuclide        | Number of cases | Average activity [ $\text{mBq g Ca}^{-1}$ ] | Minimum activity [ $\text{mBq g Ca}^{-1}$ ] | Maximum activity [ $\text{mBq g Ca}^{-1}$ ] |
|---------------------|-----------------|---------------------------------------------|---------------------------------------------|---------------------------------------------|
| $^{226}\text{Ra}$   | 33              | 1.82                                        | 0.73                                        | 4.58                                        |
| $^{210}\text{Po}$   | 80              | 29.6                                        | 5.95                                        | 101.6                                       |
| $^{90}\text{Sr}$    | 134             | 12.2                                        | 3.8                                         | 38.0                                        |
| $^{239}\text{Pu}^a$ | 42              | 0.054                                       | 0.030                                       | 0.086                                       |

<sup>a</sup>Average of measurements on vertebrae collected from 1963 to 2004

$^{210}\text{Po}$  in a compartment is due to the presence of  $^{210}\text{Pb}$  in the same compartment.  $^{210}\text{Pb}$  oxidation state in natural environment is +2, which makes the  $\text{Pb}^{2+}$  chemistry rather similar to that of  $\text{Ca}^{2+}$ . Biokinetics of Pb is very similar to Ca, thus  $^{210}\text{Pb}$  is found mostly in the skeleton.<sup>[6]</sup> In this respect,  $^{210}\text{Po}$  found in bones reflects the presence of  $^{210}\text{Pb}$  in the skeleton. The results of the analysis of 80 cases since 2006 shows that the distribution of the activity is log-normally distributed, with an average value at 29.6 mBq g  $\text{Ca}^{-1}$  (Fig. 3). The minimum value is 5.9 mBq g  $\text{Ca}^{-1}$  and the maximum value is 101.6 mBq g  $\text{Ca}^{-1}$  (Table 2). The large interval of values covering almost two orders of magnitude reflects the different mechanisms of introduction of  $^{210}\text{Pb}/^{210}\text{Po}$  in the body. As a decay product of the  $^{222}\text{Rn}$  gas,  $^{210}\text{Pb}$  can be inhaled in significant quantities in regions with high level of  $^{222}\text{Rn}$ , such as in the Tessin canton.<sup>[15]</sup> Seafood consumption is also of concern, as  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are found at higher level in these samples.<sup>[16]</sup> In addition, tobacco habits are also of concern because  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are found in significant activity in the main stream of tobacco smoke.<sup>[17]</sup>

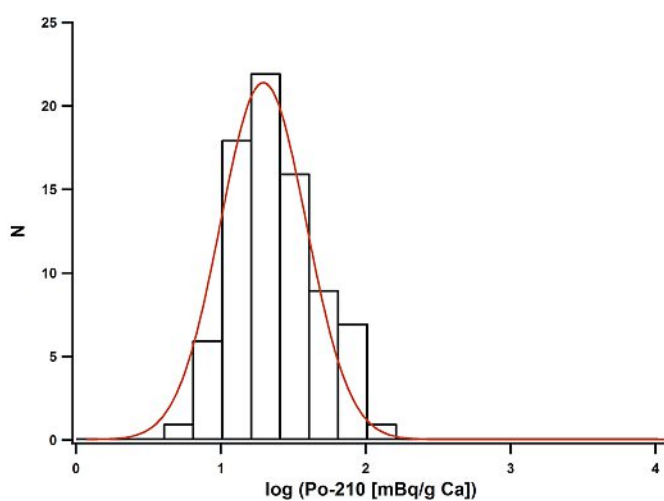


Fig. 3. Distribution of the log ( $^{210}\text{Po}$ ) activities from the Swiss  $^{210}\text{Po}$  database on  $^{210}\text{Po}$  content in vertebrae ( $n = 80$ ), showing a log-normal distribution with an average value of 29 mBq g  $\text{Ca}^{-1}$ .

### 3.3 Plutonium Transfer to Humans

Plutonium is present in the Swiss environment because of the fallout of the atmospheric nuclear bomb tests of the 1950s and 1960s and to the burn-out of a SNAP-9A satellite (1964), powered by a radioisotope thermoelectric generator (RTG) containing  $^{238}\text{Pu}$  (1 kg). In Switzerland, all the soils measured in the framework of the National Radioactivity Survey Plan contain traces of  $^{239+240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  from these events. This results in the determination of a  $^{238}\text{Pu}/^{239+240}\text{Pu}$  isotopic ratio of  $0.029 \pm 0.001$  and a  $^{241}\text{Am}/^{239+240}\text{Pu}$  ratio of  $0.424 \pm 0.012$  for soil samples collected from 2015 to 2019 (Fig. 4). Most of the activities of  $^{239+240}\text{Pu}$  are below 0.3 Bq  $\text{kg}^{-1}$ , except for some soils of higher altitude where higher fallout deposition are observed.<sup>[18]</sup> Two studies were conducted in Switzerland to investigate the fate of Pu deposited in the environment. In one of these studies, we took advantage of the milk teeth samples collected in the sixties to re-investigate the Pu content of milk teeth, using modern mass spectrometry techniques.<sup>[8]</sup> The aim was to use milk teeth to demonstrate that Pu does not cross the placental barrier. Results showed that  $^{239}\text{Pu}$  was found mostly in the root of the milk teeth and peaked in the whole milk teeth at 8  $\mu\text{Bq g Ca}^{-1}$  for children who were about 10 years old when the  $^{239}\text{Pu}$  activity peaked in air (1963). This means that the  $^{239}\text{Pu}$  activity in milk teeth was due to inhalation of contaminated air during the sixties, with a transfer to the roots due to the presence of vascularization. Enamel, formed during pregnancy was not impacted and the ratio of the activity of enamel to the

activity of the root was 0.10–0.20.<sup>[8]</sup> Thus, in contrast to  $^{90}\text{Sr}$ , Pu is not transferred across the placental barrier to the foetus skeleton.

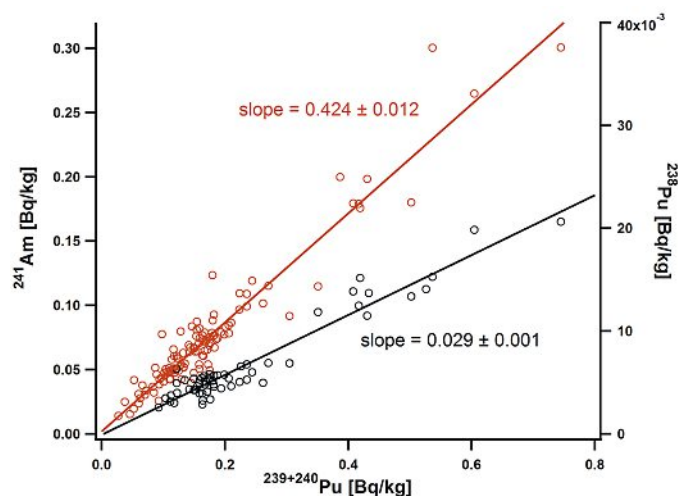


Fig. 4.  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  activities in function of the  $^{239+240}\text{Pu}$  activities of soils sampled from 2015 to 2019 in Switzerland.

$^{239}\text{Pu}$  was also found in vertebrae of the Swiss population.<sup>[3a]</sup> Similarly to the case of milk teeth,  $^{239}\text{Pu}$  was inhaled during the contamination of the air, which stopped shortly after the nuclear test ban treaty was enforced (1963). From the analyses of our database samples of vertebrae, we found that individuals who died before 1960 had undetectable  $^{239}\text{Pu}$  in vertebrae. On the other hand, we also found that individuals who were born after 1970 had undetectable or very low  $^{239}\text{Pu}$  activity. In this respect,  $^{239}\text{Pu}$  concerns individuals who inhaled contaminated air during the sixties. By measuring vertebrae of individuals who died between 1962 and 2004, we were able to estimate the retention time of  $^{239}\text{Pu}$  in the skeleton of people who inhaled  $^{239}\text{Pu}$  before 1967. After 1967, the  $^{239}\text{Pu}$  concentration in air was considered negligible. The retention half-life of  $^{239}\text{Pu}$  in the skeleton was  $40 \pm 14$  y ( $k = 2$ ).<sup>[3a]</sup> The average activity was 0.054 mBq/g Ca, a value very close to the 0.048 mBq g  $\text{Ca}^{-1}$  found in the root zone of milk teeth, showing similarity between these two calciferous tissues.<sup>[8]</sup>

### 4. Conclusions

This study demonstrates that some radionuclides, either natural or artificial, are transferred from soil to the food chain and human. The ability of a radionuclide to be transferred depends mostly on its chemistry or, in case of natural radioactivity series, on the chemistry of its parent radionuclides. The very radiotoxic fission product  $^{90}\text{Sr}$  is easily transferred to the human skeleton because of its chemical similarity with Ca. However, results show that the human metabolism has a preference of Ca over Sr, the TF of milk-to-milk teeth and bone being close to 0.3. In addition, results show a much higher preference for Ca over Ra. This is probably due to the larger cationic radius of Ra compared to Ca and Sr, preventing its passage across biological membranes and its introduction into the structure of the apatite of bones and teeth. Thus, Ra is found in much lower concentration in vertebrae than  $^{90}\text{Sr}$ , given that the  $^{226}\text{Ra}$  activity in soil is currently about 60 times higher than the activity of  $^{90}\text{Sr}$ .  $^{210}\text{Po}$  is probably not directly transferred to bones but its presence in vertebrae is the consequence of decaying  $^{210}\text{Pb}$  having similar biokinetics as Ca in human. Even if Pu does not cross the placental barrier, it is present in detectable activities in vertebrae. In addition, its retention half-life is 40 years, which means that once inhaled, Pu remains in significant amounts in the human body for a lifetime, causing a potential irradiation danger. While the measured activities of the considered

radionuclides in this study have become low (Table 2) and do not represent a significant radiation dose to the public, it is necessary to monitor the fate of these radionuclides. The (radio) analytical work of the past 60 years yielded data that form the basis of all numerical modelling of radioactivity transport. Switzerland still has four working nuclear power reactors with another one being currently decommissioned, thus the necessity to detect any potential environmental contamination well deserves the analytical efforts.

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