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Long-Term Effects of Exposure to Low-Levels of Radioactivity: a Retrospective Study of ^{239}Pu and ^{90}Sr from Nuclear Bomb Tests on the Swiss Population

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1. Introduction

The production of nuclear energy may be a source of exposure to ionizing radiations, either for nuclear workers or the general population. Ionizing radiations are one of the most studied and ubiquitous carcinogens in our environment. The main basis for estimating the carcinogenic risk to humans are studies conducted on survivors of the Japanese atomic bomb, a population essentially exposed to acute high dose rates (Cardis et al., 2005b). Epidemiological studies of nuclear workers, who have been chronically exposed to much lower radiation doses show evidences that the classic approach of radiation protection based on the linear non threshold (LNT) model is valid: an excess relative risk (ERR) of cancers other than leukemia of 0.97 per Sv was found among a cohort of more than 400,000 nuclear workers in 15 different countries, all individually monitored for external exposure. Additionally, the study showed an ERR of 1.93 per Sv for leukemia, a disease that may be associated with radiation exposure (Cardis et al., 2005b). Nevertheless, these findings were not confirmed in a later study of workers from the French National Electricity Company (Rogel et al., 2005), and neither in the study of the United Kingdom Atomic Energy Authority (Atkinson et al., 2004). Conversely, both studies showed a strong healthy worker effect (HWE). Higher cancer rates are related to a significant cumulative radiation dose (Hunter and Muirhead, 2009). Besides, an adverse effect on the pulmonary function when exposed to ^{137}Cs has also been recently demonstrated for children living near the Chernobyl exclusion zone (Svendsen et al., 2010). Currently, there is a tremendous interest in the study of the Techa River cohort (TRC), a population which was exposed to low to moderate radiation doses at low-dose rates from radioactive releases from the Mayak plutonium production facilities into the Techa river between 1950 and 1956 (Degteva et al., 2000; Tolstykh et al., 2001; Krestinina et al., 2010). At total body doses as high as 10 Gy, 30% of cancer deaths have been attributed to plutonium exposure among the Mayak workers (Sokolnikov et al., 2008). In that particular case of worker exposure to high doses of radiation, the ERR due to plutonium exposure was 7.1 per Gy for males at age 60.
While numerous studies of the effect of ionizing radiation on nuclear workers exist, there are only a few studies which extend their research to the general population. Among any

given population, children are considered at higher risk (BEIR7, 2006) and several studies have focused on the risk of leukemia for children living in the proximity of a nuclear power plant (NPP)(Roman et al., 1999; Doyle et al., 2000; Cardis et al., 2005a; Kaatsch et al., 2008; Nussbaum, 2009). Laurier et al. (Laurier et al., 2008) have reviewed studies dealing with as much as 198 nuclear installations, three of them showing a significant excess of leukemia among children under 18 years of age (on the order of 5 cases detected for less than one expected). While the statistics tend to support the excess risk, the radioecological studies carried out on these sites failed to correlate the ERR with a significant increase of radioactivity burden in the environment or in food (Heasman et al., 1986; Black et al. 1994; Bithell et al., 1994; Hoffmann et al., 1997). The most recent study concerning the adverse effects of living in the proximity of a NPP revealed an odd ratio (OR) of 2.19 (with lower one-sided 95% confidence limit of 1.51) for leukemia in young children living in the vicinity of German NPP (Kaatsch et al., 2008). This study gave rise to new questions about the relationship existing between ERR and the distance to a NPP and the way to demonstrate the role, if any, of ionizing radiation in the observed ERR.

Most epidemiological studies on the effect of NPP on a general population, especially children, link leukemia statistics with the proximity to a NPP. No clear evidence came out of these studies and by their nature, these studies are subject to bias. There have been very few attempts to correlate leukemia cases with radiation dose because this latter is mostly unknown with respect to the general population, or considered much lower than the natural radiation background. In this respect, some authors have introduced a measure of doubt regarding the validity of the dose factors used for the fetus and newborn (Nussbaum, 2009; Shore, 2009); ERR of leukemia among children under 5 years of age living in the vicinity of a NPP may be the consequence of the father's exposure to radiation or direct fetus exposure, with a possibility that dose factors have been largely underestimated. While this assumption may be valid, Doyle et al. (Doyle et al., 2000) found no evidence of a link between exposure to low-level ionizing radiation before conception and an increased risk of adverse reproductive outcomes in men working in the nuclear industry. Nevertheless, the authors state that a possible link between a woman's exposure to radiation before conception and an increased risk of fetal death requires further investigation.

In addition to the lack of knowledge about the exposure dose to the population, dose rates due to in-vivo alpha-emitters, particularly transuranic elements, have been totally overlooked, mainly because of measuring difficulties (Schmitz-Feuerhake et al., 2005). Our opinion is that there is a lack of data on the radioactivity body burden of the general population, and in particular children. This information would facilitate the interpretation of leukemia statistics in terms of radiation dose rather than in terms of the distance to a radiation emission source (e.g. a NPP).

The determination of the external radiation dose in the vicinity of a NPP is obviously mandatory but may be of little use in the case of the risk related to very low doses of ingested or inhaled radionuclides. In this respect, the main challenge of national authorities is to survey diet and, possibly, the population body burden. While radioactivity surveys of food and the environment are a dedicated task of every laboratory dealing with environmental radioactivity surveys (Froidevaux et al., 2006a), the determination of very low-levels of radioactivity in the body is another challenge; whole body counting may provide data on ^{137}Cs (or other γ-emitters) contamination (Hodgson et al., 2004), but previous studies show that ^{90}Sr and plutonium could be the most radiotoxic isotopes that

should be searched for (Atkinson et al., 2004). Over the last ten years, mass-spectrometry has been applied as an alternative detection method for measuring actinides, especially plutonium. Using ICP-MS, the detection limit is lowered by a factor of ten compared to alpha-spectrometry. The detection limits at the femtogram-level needed for some particular environmental samples, are typically obtained by double focusing sector-field ICP-MS instruments or TIMS (Kim et al., 2000; Becker, 2003; Boulyga et al., 2003; Agarande et al., 2004; Baglan et al., 2004; Lindahl et al., 2010; Qiao et al., 2010). Using sf-ICP-MS, we were recently able to determine ^{239}Pu and ^{239}Pu/^{240}Pu ratio in the milk teeth of children born in Switzerland between 1951 and 1995 (Froidevaux and Haldimann, 2008) and in the vertebrae of individuals who died between 1962 and 2004 (Froidevaux et al., 2010).

Bones and teeth are both the main targets of plutonium and ^{90}Sr entering the blood stream. Approximately 100% of ^{90}Sr and 50% of the plutonium incorporated in humans is located in the skeleton. Another 30% of the plutonium burden is located in the liver (ICRP (International Commission on Radiological Protection), 1986). In this respect, the analysis of teeth and bones may be of great help to investigate low dose and low dose-rate exposure to radionuclides potentially emitted throughout the process of nuclear energy production (Culot et al., 1997). Among teeth, milk teeth (deciduous teeth) are very precious indicators of bone-seeking radionuclides that can be transferred from the mother's blood plasma to the fetus. Because deciduous enamel forms between week 13 in utero up to 9 months postnatally (thereafter essentially becoming inert), the analysis of milk teeth allows for the retrospective measurement of prenatal and early postnatal trace-element uptake during a critical period of child development (Dolphin et al., 2005). Another advantage of milk teeth as a tool for retrospective studies of bone-seeking radionuclide contamination is the fact that they are naturally shed at ages 6-12 years old. Conversely, there exists no other possibility of bone sampling except for collection at autopsy. Unfortunately, due to legal difficulties in obtaining samples for analysis, there are only limited data available worldwide, mostly concerning ^{90}Sr (Hodgson et al., 2004). The USTUR (United States Transuranium and Uranium Registries) is a source of very valuable information for human tissue studies, including bones, biokinetics, bioeffects and the dosimetry of uranium and transuranium in human. However, this database contains mainly tissues from workers exposed to radionuclides. As an example, USTUR studies show that, in addition to skeleton and liver, muscle is an appreciable reservoir for both plutonium and americium (Kathren, 1995).

We propose to use the ^{90}Sr and plutonium fallout of the nuclear bomb tests (NBT) of the sixties as a proxy to determine the radioactivity body burden that the population has received as a consequence of the tests. We used the Swiss databases on ^{90}Sr and ^{239}Pu in milk teeth and bones because this latter is the most complete that is currently available worldwide and covers a range of years from 1952-2002 (milk teeth) and 1960 to 2009 (vertebrae). We compared our results with other studies carried out worldwide. We hypothesized that the level of both radionuclides incorporated in the skeleton has been very low but significant as well as measurable using careful radiochemical and analytical procedures. We put the incentive on the analysis of ^{239}Pu and ^{90}Sr in milk teeth of children born from 1953 to 2005 to demonstrate the danger that these radionuclides may create if transferred to the fetus across the placental barrier. In addition, we collected vertebrae of deceased individuals (1960-2005) to measure the incorporation of very low-levels of bone-seeking ^{239}Pu and ^{90}Sr in the body.

2. Experimental

^{90}Sr analysis in milk teeth and bones is a very dedicated task and must be carried out with particular care to avoid bias due to other radionuclides that will be present in the sample, such as uranium isotopes, radium isotopes and their daughter products. As a consequence, a careful radiochemical separation of ^{90}Sr (or ^{90}Y) from the matrix and other radioisotopes is compulsory, because ^{90}Sr is a β-emitter whose counting, either by LSC or proportional counting, will be submitted to bias if other β-emitters are still present. In this respect, the analytical method used by the Radiation and Public Health project (RPHP, see www.radiation.org) for the determination of ^{90}Sr in milk teeth appears not satisfactory for the purpose of the authors and the conclusions that they draw from their studies. This is because the RPHP method does not use any chemical separation, except for a carbonate precipitation that will hardly remove ^{90}Y, radium isotopes, ^{210}Pb isotopes and its daughter ^{210}Bi, as well as thorium isotopes and many others (see for instance Mangano and Sherman, 2011). Thus, we propose below a method that is an alternative to the classical method using Eichrom Sr.spec resin in the analysis of ^{90}Sr, particularly for large sample sizes containing significant amounts of Ca, such as teeth, milk and bone. For ^{239}Pu determination, the main problem resides in the purification of plutonium from uranium, because the $^{238}UH^{+}$ hydride will interfere with the determination of ^{239}Pu; our methodological procedure is addressed in the corresponding sub-chapter.

2.1 Sampling

The vertebrae have been collected by pathologists over the last 45 years all over Switzerland. They were taken during autopsy from different adults (n=1200), of whom 80% were older than 60 and none younger than 8 at the date of death. 90% were older than 20 years in 1960. Thus our database represents the impact of nuclear bomb tests fallout on a population of adults, minimizing the problem of age-dependency in modelling. Milk teeth were collected by dentists (mainly school dentists). Both vertebrae and milk teeth were collected as part of the Sampling Program of the Swiss Federal Office of Public Health for environmental radioactivity survey, from the beginning of the nuclear era. The whole program is currently ongoing for ^{90}Sr determination in vertebrae and milk teeth. ^{90}Sr determination in vertebrae was carried out on individual cases as early as 1960 and required at least 5 g of bone ash; this study reports on the data set obtained during the last 50 years. ^{90}Sr analysis of milk teeth requires also at least 5 g of teeth ash, which necessitates about 10 milk teeth of children born during the same year in the same region. ^{239}Pu determination by mass spectrometry was carried out only after 2005, when adequate technology became available (see below). The samples for ^{239}Pu analyses are sub-samples of the vertebrae and milk teeth samples set used for ^{90}Sr determination that were preserved for further analyses. In this respect, the plutonium data set represents a retrospective study. To reach a satisfactory detection level and to minimize the effect of inter-individual variability, we pooled the samples of several individuals deceased in the same year to obtain at least 30 g of vertebrae ash per analysis. For milk teeth, we also used about 30 g of teeth ash, which required pooling about 50 teeth of children born the same year in Switzerland (Froidevaux et al., 2006b; Froidevaux and Haldimann, 2008; Froidevaux et al., 2010).

2.1 Radiochemistry

2.1.1 Plutonium analysis

Teeth ash was spiked with 10±0.5 mBq of ^{242}Pu and submitted to microwave digestion in a Milestone MLS Ethos Plus digester (MLS GmbH, Leutkirch, Germany) for 40 minutes at

170°C in 8 M nitric acid (100 ml). After filtration plutonium was extracted on a Bio-Rad (Bio-Rad, Reinach, Switzerland) ionic exchanger (100-200 mesh, 25 ml in 1 cm diameter chromatography column) and purified on a micro-column (100 mg) of Eichrom® TEVA resin (Eichrom Environment, Bruz, France) while taking care to use Ultrapur® reagents (Merck, VWR International, Dietikon, Switzerland) to minimize the uranium content (plutonium was extracted in 5 ml of HNO_3 8 M, the column was washed with 10 ml of HNO_3 3M, 3 ml of HCl 9 M and once again with 3 ml of HNO_3 3 M). Plutonium was measured in 5% HNO_3 by SF-ICP-MS (see below). The same analytical procedure was used for the vertebrae. Calcium was measured on all samples by AA with a Perkin Elmer 4100 apparatus (Perkin Elmer AG, Schwerzenbach, Switzerland) and results of plutonium isotopes activity are given as Bq g Ca^{-1} (Froidevaux and Haldimann, 2008).

2.1.2 ^{90}Sr analysis

From 1963 to 1971, milk teeth and bones were analysed using the nitric acid fuming method (Froidevaux et al., 2006b). After 1974 and until now, we used a method using two cation exchange chromatography columns (AG 50w-x8, 50 and 8 ml) and specific complexing agents such as EDTA and CyDTA (*trans*-1,2-cyclohexylen-dinitrilo tetraacetic acid) to purify ^{90}Sr. After ingrown into ^{90}Sr, ^{90}Y daughter product was purified using a cationic exchanger (AG 50w-x8, 8 ml) and a malonate eluting solution. After precipitation of ^{90}Y as oxalate, the micro-precipitated source was counted in a low-level (0.3 dpm) proportional counter (Tennelec LB 4100w). For details of the procedure, see (Froidevaux et al., 2006b)

2.2 Mass spectrometry

Determining ^{239}Pu levels in most samples is hampered by the ubiquity and abundant presence of uranium. Consequently, peak tailing from $^{238}U^+$ and the formation of $^{238}U^1H^+$ molecular interference isobaric to ^{239}Pu are the limiting factors of plutonium analysis. The hydrogen originates from the water solvent (Zoriy et al., 2004). Because the degrees of ionization of uranium and hydrogen atoms in argon plasma are 100 % and about 0.1 %, respectively, uranium hydride UH^+ is formed in the plasma, most probably through reaction:

$$U^+ + H \rightarrow UH^+$$

To minimize the formation of $^{238}U^1H$ ions, the chemical separation of uranium and plutonium using a TEVA resin prior to analysis did not yield sufficient removal of uranium. Therefore, it is essential to further reduce the formation of uranium hydride interference and correct for the residual signal contribution at m/z 239 in order to obtain unbiased plutonium measurements at ultra trace levels. The reduction of hydride formation of uranium requires special sample introduction systems, such as a micronebulizer with a desolvator. In this study, we equipped the instrument with an APEX Q desolvation device (Elemental Scientific, Scientific, Omaha, NE, USA) in combination with an ACM membrane unit (Elemental Scientific) that was effective in removing the solvent from liquid samples, thereby greatly reducing the formation of $^{238}U^1H^+$ while retaining plutonium for transport to the plasma ion source in the form of a dry aerosol. Additionally, this special configuration improved the signal-to-noise ratio by a factor of about 10, which was necessary to reach the very low plutonium concentrations. The $^{238}U^1H^+/^{238}U$ ratio was determined experimentally and a correction factor was applied to calculate the net ^{239}Pu response. The measured

$^{238}U^{1}H^{+}/^{238}U$ ratio of 1.4 x 10^{-5} ± 1 x 10^{-6} was constant under the selected experimental conditions and falls well into the range of 1.2 x 10^{-4} - 5.0 x 10^{-6} reported in the literature (Becker et al., 1999; Kim et al., 2000; Boulyga and Becker, 2001; Taylor et al., 2001; Wyse et al., 2001). We used a double focusing SF-ICP-MS (Element2; Thermo, Bremen, Germany) for measuring the ^{239}Pu, ^{240}Pu, and ^{242}Pu isotopes. Furthermore, we monitored the ^{238}U isotope to correct for residual background contributions to the ^{239}Pu signal as a result of variable uranium concentrations in the actual sample solutions. Moreover, ^{199}Hg was measured to check for potential $^{199}Hg^{40}Ar$ interference. We performed 36 consecutive scans on each sample. The total measurement time per sample was 94 s. We applied the low-resolution mode ($m/\Delta m = 300$) to obtain maximum ion transmission. Optimum argon flow conditions in the PFA-100-1036 nebulizer (Elemental Scientific) were in the range of 0.9–1.1 l/min at a sample uptake rate of 235 µL/min. All other parameters of the ICP-MS corresponded to normal operating conditions. We conducted quality controls of the chemical separation and ICP-MS measurements by spiking 100 mL of 8 M HNO_3 with a reference solution of ^{239}Pu and subjected it to the overall ^{239}Pu determination process. The measured value of 2.7 ± 0.1 mBq (n = 7) agreed well with the reference value of 2.8 ± 0.3 mBq.

3. ^{90}Sr in milk teeth

The level of ^{90}Sr in milk teeth has been recorded in several countries since the beginning of the nuclear weapon tests. Data show that there are two peaks in the concentration of ^{90}Sr in milk teeth versus the year of birth, in 1958 and in 1965, followed by an exponential decrease after the entry into force of the Nuclear Test Ban Treaty (Rosenthal et al., 1964; Aarkrog, 1971; Rytomaa, 1972). There were several further studies on teeth, including permanent teeth, initiated punctually to answer specific questions, such as how much radiation the population received following the Chernobyl NPP accident (Kulev et al., 1994; Stamoulis et al., 1999), the contamination from the Sellafield reprocessing plant (ODonnell et al., 1997) or the contamination of the Techa river (Tolstykh et al., 2003; Tolstykh et al., 2008). In Switzerland, a monitoring program of ^{90}Sr in milk teeth was initiated by the Federal Office of Public Health in the late fifties and is still ongoing (Froidevaux et al., 2006b). Figure 1 displays the features of the ^{90}Sr content of milk teeth monitored in Switzerland (1951-2002, (Froidevaux et al., 2006b)), Denmark (1953-1963, (Aarkrog, 1971), United-Kingdom (1960-1971, (Hodgson et al., 2004)), Germany and Northern Ukraine (1982-1986, (Schmitz et al., 2004)) and shows the maximum value determined in Norway (1965) and Finland (1963) and, between 1976 and 1979, in French Polynesia (Badie et al., 1987). Dates must be understood as the birth year of the children. For the United-Kingdom, the data represent activities found in premolar roots of milk teeth from children aged 10 (Hodgson et al., 2004). In this respect, and because the roots of teeth have a bone-like structure (Gulson and Gillings, 1997), these data correspond better to the activities that can be found in trabecular bones, rather than in the enamel of milk teeth. Figure 1 shows that ^{90}Sr (Bq/gCa) activities found in Switzerland are situated between the maximum values found in Norway and Finland and the activities determined in Denmark. Activities of premolar roots from the UK are significantly lower, showing that milk teeth may actually accumulate ^{90}Sr to a larger extent than bones. ^{90}Sr originating from the Chernobyl disaster (1986) has not significantly impacted Switzerland and Germany, but possibly the slightly higher values found in milk teeth of children from northern Ukraine may reflect the contamination of foodstuff with ^{90}Sr of Chernobyl or other origins in this region. Current values found in Switzerland are close to 25 mBq/gCa.

^{90}Sr is mostly incorporated through ingestion of contaminated food, a situation which may last for decades after fallout deposition on soil. Worldwide studies on milk teeth contamination demonstrate that NBTs fallout from the sixties has significantly increased the ^{90}Sr activity of food, especially milk, so that a very significant increase in ^{90}Sr (up to a value of 0.3 Bq/gCa in 1963 in Switzerland) is observed in milk teeth. Figure 2 shows the results of ^{90}Sr analysis of milk for Switzerland (1954-2009) and Norway (1953-1972, (Christensen et al., 1975)). Additionally, data for milk from New-Zealand are added for comparison of both Northern and Southern hemispheres (Badie et al., 1987). While several large NBTs were conducted in the Southern hemisphere, this latter presents an overall contamination several times less than the Northern hemisphere. This fact is reflected by the very low activity (average 14 mBq/gCa) determined in the milk teeth collected in French Polynesia, 4 times less than in Switzerland for the same date of birth. For Switzerland, the data presented in Figure 2 are issued from the analysis of milk collected in the Swiss lowlands. As a matter of fact, milk activities considerably differ if the milk is sampled from cows who grazed in Alpine pastures, with a ten times increase in activity for grazing land located at 2000 m above sea level (Pourcelot et al., 2007).

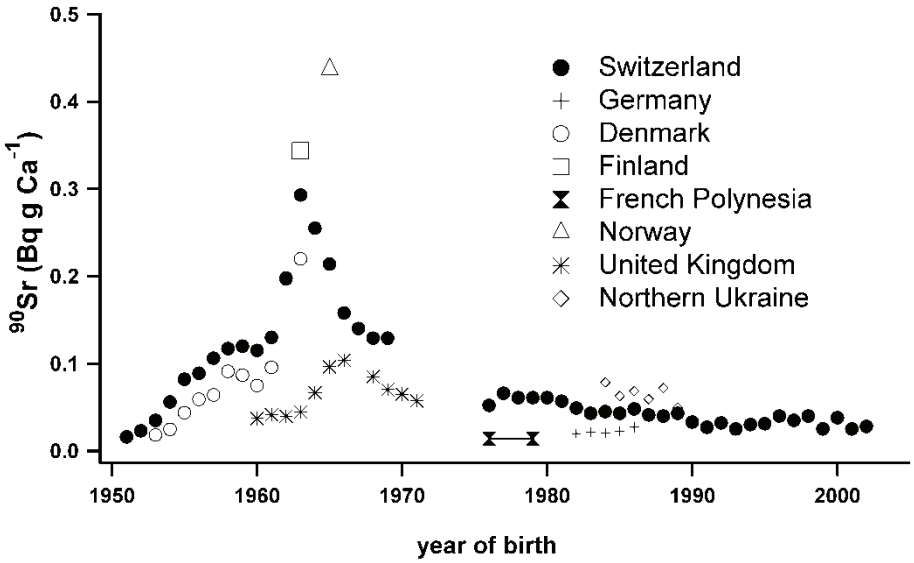

Fig. 1. ^{90}Sr activities (Bq/gCa) in milk teeth as a function of the year of birth of the children.

Activities in milk samples display essentially the same features as milk teeth, with a maximum activity observed in 1963, just before the entry into force of the Partial Nuclear Test Ban Treaty that ended American, British and former Soviet Union atmospheric nuclear weapon tests. Accordingly, we can conclude that the food (milk)-mother-fetus-milk teeth pathway is the most important mechanism of milk teeth contamination. A very significant correlation is observed between milk contamination and milk teeth contamination in Switzerland (Pearson correlation coefficient of 0.92) with a milk-to-milk teeth transfer factor of 0.2. In this respect, milk contamination by ^{90}Sr is a very good proxy of milk teeth contamination.

3.1 Influence of the Swiss nuclear power plants on ^{90}Sr activities of milk teeth

The Swiss database on ^{90}Sr contamination of milk teeth contains information concerning three distinct regions of Switzerland; the first region includes the canton of Zürich and a large part of the Swiss German lowlands. The Gösgen, Beznau and Leibstadt nuclear power plants (NPPs) are included in this region. The second region is the French-speaking part of Switzerland, mainly the canton of Vaud. There is a potential influence of the Mühleberg NPP in the Eastern part of this region. The third region is the Italian-speaking part of Switzerland, which is shielded from the Swiss NPPs by the Alps and borders Italy which has no active nuclear program (Froidevaux et al., 2006b).

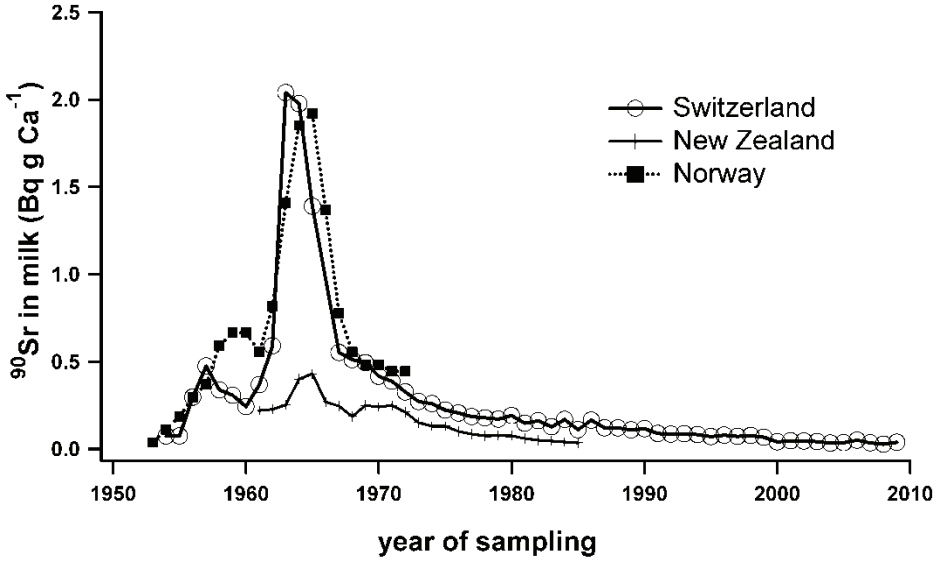

Fig. 2. ^{90}Sr activities (Bq/gCa) in milk sampled in Switzerland, Norway and New-Zealand.

Figure 3 shows the results of an analysis of the ^{90}Sr activities of the cantons of Tessin and Zürich as a function of the ^{90}Sr activities measured in the canton of Vaud, for the same years of birth. Both correlations are very strong (Pearson coefficient =0.99), which means that the contamination of milk teeth originated from the same event, e.g. the NBT fallout of ^{90}Sr. Nevertheless, both slopes differ from unity (0.84 for Zürich and 0.82 for Tessin). This fact may reflect a difference in consumption habits in the canton of Vaud compared to the cantons of Tessin and Zürich . Additionally, the canton of Vaud includes large areas of the Jura mountains and pre-alpine mountains that possibly contribute to a higher contamination of milk (Pourcelot et al., 2007). The canton of Zürich , which is potentially under the influence of 4 among the 5 nuclear reactors located in Switzerland, presents no statistical differences compared with the canton of Tessin county, which is shielded from these same reactors by the Alps. This indicates that the Swiss NNPs have had no significant impact on the ^{90}Sr activities of milk teeth and that the ^{90}Sr in these human samples arises only from the NBT fallout of the sixties that contaminated foodstuff and the mother-to-fetus contamination across the placental barrier.

4. Plutonium in milk teeth

There exist only a few studies on teeth contamination by plutonium. The lack of data on plutonium in teeth is a result of the difficulty of the analysis, which requires a large sample size to yield results above the detection limit. This is because the plutonium activity is so low that a classical alpha-spectrometry technique is barely satisfactory to produce activities and uncertainties that could be used to demonstrate the potential influence of NPP on the plutonium body burden.

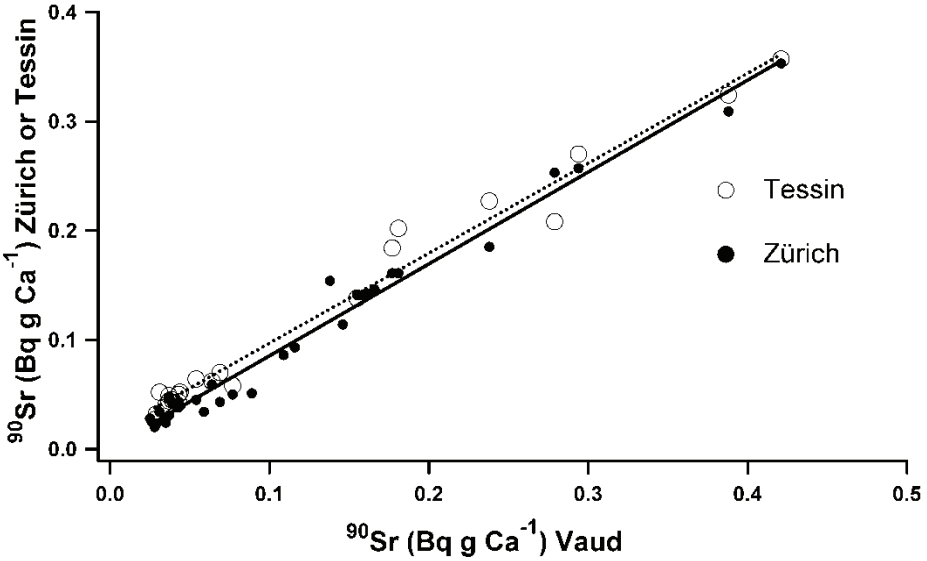

Fig. 3. ^{90}Sr activities (Bq/gCa) of milk teeth of the cantons of Tessin and Zürich as a function of the ^{90}Sr activities of milk teeth of the canton of Vaud, for the same years of birth (1953-1996).

Long et al. (1991) have determined the activity of $^{239+240}$Pu in milk teeth of 35 different geographically located child populations in the United Kingdom. The average concentration was 5.2 mBq/kg of teeth ash. They concluded that there was no correlation with mean annual rainfall, that can be used as a proxy for the intensity of the radionuclide deposition (Renaud et al., 2003; Pourcelot et al., 2007), or with the distance from the Sellafield reprocessing plant. In contrast, O'Donnel et al. (1997) found a significant correlation between the plutonium content of milk teeth and the distance to the Sellafield plant, with activities near the plant close to 6 mBq/kg ash. At a distance above 150 miles from Sellafield, the activity drops to 3.0 mBq/kg ash.

The use of sf-ICP-MS allows us to analyse our set of milk teeth sampled from 1951 to 1995 for ^{239}Pu content and ^{239}Pu/^{240}Pu ratio (Froidevaux and Haldimann, 2008). In fact, we used the ^{239}Pu activity in milk teeth to probe for the potential of plutonium, a highly radiotoxic nuclide, to cross the placental barrier. We hypothesized that the plutonium inhaled by the mother will possibly cross the placental barrier, leading to a similar activity profile as observed for ^{90}Sr in milk teeth. Surprisingly, results show a very different situation, with

^{239}Pu activity peaking for the years of birth 1954-1956, some 9 to 10 years earlier than the ^{90}Sr peak (Figure 4). Nevertheless, the ^{240}Pu/^{239}Pu isotopic ratio is 0.21±0.06 (n=23) from which we infer that the plutonium source is indeed the NBT fallout (Kelley et al., 1999; Warneke et al., 2002).

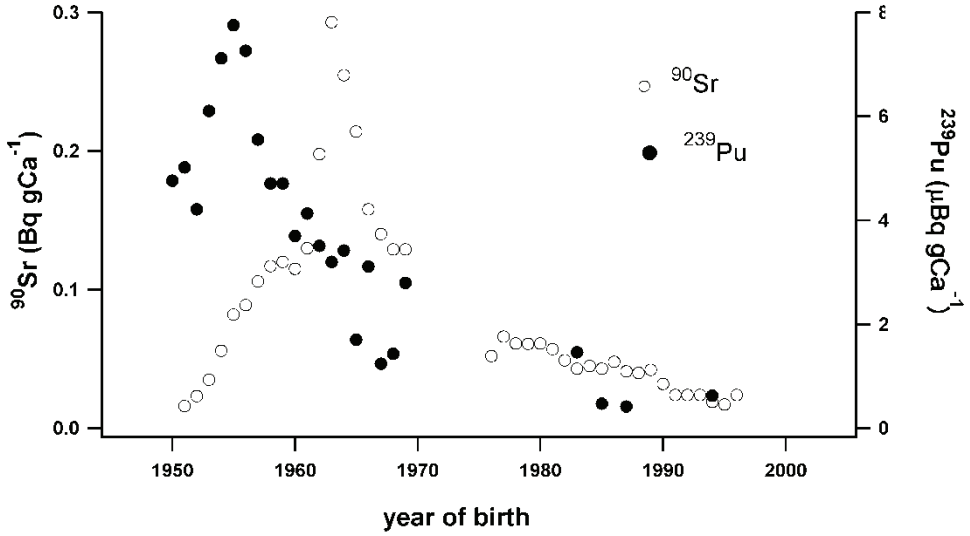

Fig. 4. ^{239}Pu (mBq/gCa) and ^{90}Sr (Bq/gCa) activities in milk teeth of children born in Switzerland 1950-1996.

A careful analysis of the year at which children shed their teeth shows that most of the milk teeth collected in the Swiss study are coming from children about 9 to 11 years old. This is because the teeth are collected by dentists, usually by school dentists. Most of the teeth sampled in this way are thus larger teeth (e.g. premolar or molar) that are shed at an older age compared to other teeth e.g. incisors. The offset between the highest activities of plutonium found in milk teeth and the maximum highest activities that were measured in air during the sixties represents exactly the time necessary to shed the teeth. Thus we interpret this offset as a proof that plutonium has not crossed the placental barrier, otherwise the plutonium activities in the milk teeth would have peaked for the same year of birth than ^{90}Sr. Instead, the plutonium activities that we measured in the teeth reflect the plutonium concentration in the air during the year or so before tooth shedding, which takes place on average 10 years after birth according to our study (Froidevaux and Haldimann, 2008). Because our study used whole teeth, we measured plutonium that was present in the root and cementum structures of the teeth, both remodelling at the same rate as the trabecular bone socket (Gulson and Gillings, 1997). To confirm this hypothesis, we roughly separated the roots of milk teeth from the rest of the teeth for two samples (born 1954 and 1965, 30 teeth) before measurement and found that ^{239}Pu in the roots has activities up to 9 times higher than the rest of teeth (mainly enamel). It is likely that the low residual ^{239}Pu activity found in the enamel represents a contamination by residual root and dentine structures still present in the enamel sample. Therefore, it is highly probable that the enamel laid down in utero is virtually free of ^{239}Pu. Consequently, neonates were probably born free

of plutonium, even when the environmental plutonium was at its highest, in the early 1960s. Our interpretation of the plutonium activity in deciduous teeth as a function of the year of birth is that neonates start to incorporate plutonium through inhalation of contaminated air after birth, until they shed their milk teeth. Because of the exchange of the bone-like structure of the teeth with the blood plasma, a higher concentration in milk teeth will show up for children shedding their whole teeth when the plutonium concentration in the air was at its highest, i.e. 1963. Thus ^{239}Pu activity will peak in milk teeth of children born circa 1953, as observed in Figure 4.

To compare our results with the results from the literature, we used a conversion factor of 0.35 g Ca/g ash (Swiss database, n=182, mean= 0.35±0.04) because results were given as mBq/kg ash in the former literature. We also used a multiplication factor of 1.68 of our ^{239}Pu value because the literature data concern the activity of both ^{239}Pu and ^{240}Pu isotopes as given by alpha-spectrometry. O'Donnell (ODonnell et al., 1997), found a mean activity of 5 mBq/kg ash in permanent teeth sampled in 1990 in the UK to compare with our value of 0.6 mBq/kg ash. Long et al. found similar values in a study of 35 different regions of the UK, with a mean value of 5.2 mBq/kg ash. The factor of 9 found between milk teeth in our study and permanent teeth in the UK studies confirms that plutonium does not cross the placental barrier and does not contaminate the enamel of the deciduous teeth that was laid down during pregnancy. In this respect, the activity of the permanent teeth possibly represents a proxy for the activity that might be found in bone. We did not discover any potential influence of the 5 Swiss NPPs on the ^{239}Pu contamination of milk teeth, as demonstrated by the ^{240}Pu/^{239}Pu isotopic ratio of 021±0.06, close to the reference value of 0.187 for NBT fallout (Kelley et al., 1999; Warneke et al., 2002; Froidevaux and Haldimann, 2008). A significant decrease of this ratio would have supported the hypothesis that some of the plutonium arises from other sources, e.g. burnt nuclear fuel (Ketterer et al., 2004a), while a significant impact of the Chernobyl accident would have increased it up to 0.35 (Ketterer et al., 2004b).

5. ^{90}Sr in bones

Similarly to milk teeth, several countries have initiated a program of measuring the ^{90}Sr body burden through analysis of bones sampled at autopsy (Aarkrog, 1971; Christensen et al., 1975; Bauman et al., 1977; Dehos and Kistner, 1980; Kalmykov et al., 1997; Stamoulis et al., 1999; Hodgson et al., 2004; Froidevaux et al., 2010). In the particular case of bones analysis, the choice of the type of bones sampled is very critical. The strontium levels in bone vary according to the bone structure, and higher amounts of strontium are found in cancellous bone than in cortical bone after stable strontium ingestion. However, strontium levels at different skeletal sites are strongly correlated, and the strontium content of the lumbar vertebra may be estimated from iliac crest bone biopsies in the monkey (Dahl et al., 2001). Usually cancellous bone such as vertebrae, ribs or the iliac crest are used for a ^{90}Sr survey because the remodeling rate is higher compared to compact bones (Martin and Seeman, 2008; Zebaze et al., 2010). In this way, a better evaluation of the radioecological situation in a given location is obtained using cancellous bones because the activity found in the bone will reflect more accurately the current contamination of foodstuff. Nevertheless, the use of cortical bone such as the tibia may be of help when an archive of contamination is needed.

In Switzerland, the Federal Office of Public Health initiated a ^{90}Sr survey program using an analysis of ^{90}Sr in vertebrae of individuals deceased between 1960 and now. To our

knowledge, the Swiss database is the only one reflecting the ^{90}Sr contamination of the human skeleton for the last 50 years and which is still ongoing. Results are presented in Figure 6, in which data from several other studies from the literature are also given for comparison; the ^{90}Sr activities for Switzerland are situated between the average activities found worldwide in adults and activities found in Denmark for children deceased before their first year. Higher values were also found in children less than 5 years in Norway (highest activity=0.52 Bq/gCa for the year of death 1965) with a children/adults (>20y) activity ratio of 2.5 for children less than one year, of 3.3 for children less than 5 years and 1.7 for children between 5 and 20 years at the time of death (Christensen et al., 1975). Current activities are in the order of 0.028 Bq/gCa), a value that is typically found elsewhere (Stamoulis et al., 1999).

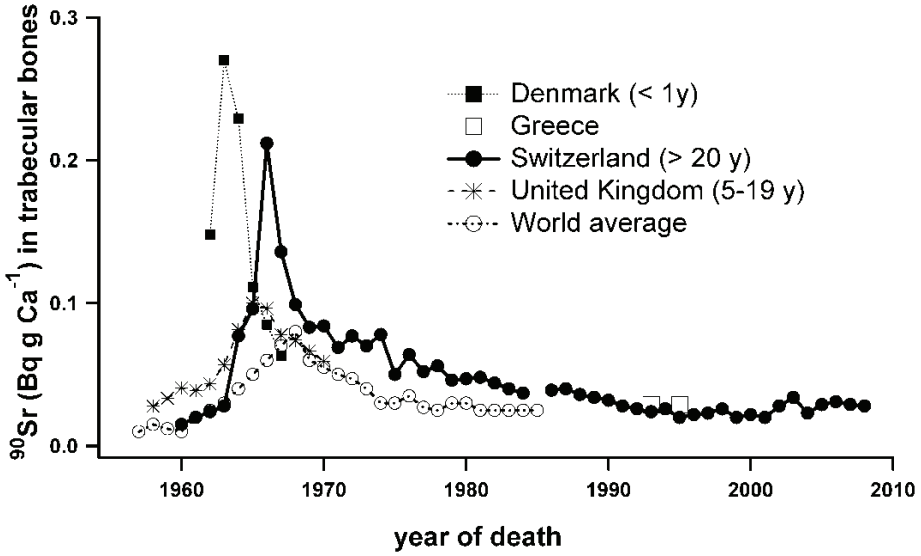

Fig. 6. ^{90}Sr (Bq/gCa) in cancellous (trabecular) bones sampled at autopsy as a function of the year of death.

Swiss data are significantly above the world average data or United Kingdom data, which is most likely a reflection of the Swiss population's high consumption of dairy products. The highest activity is found for the year 1965-1967 in adults in Switzerland and worldwide and in children 5-19 years in UK, but significantly earlier in children less than one year old in Denmark and Norway (1963-1965; (Aarkrog, 1971; Christensen et al., 1975)). This observation is supported by the higher remodeling rate of bones and bone growth measured in younger children. As a consequence, the incorporation of ^{90}Sr in bones is faster (and higher) for young children than for adults (see also (Papworth and Vennart, 1984) for a discussion on the uptake and turnover of ^{90}Sr in the human skeleton). In this way, ^{90}Sr contamination of the environment and foodstuff will have a greater negative impact on children than on adults, leading to an absorbed dose in the marrow to age 70 about 30 times higher if ^{90}Sr is incorporated when less than one year old, compared to a similar ingestion at the age of 20 years (Papworth and Vennart, 1984). No effect of the Chernobyl accident was

observed on the vertebrae activities in Switzerland, the pre-Chernobyl activities being somewhat higher (0.044 Bq/gCa, 1980-1985) before the accident than after (0.037 Bq/gCa, 1986-1989). Vertebrae were collected in different regions of Switzerland, and two regions are still currently being sampled; for the same year of death, the activities in vertebrae sampled in the canton of Vaud are strongly correlated with the activities in vertebrae collected in the canton of Tessin (Pearson=0.98), in the Basel area (Pearson=0.96), and in the St-Gallen area (Pearson=0.99). Nevertheless, the slopes differ from unity for all correlations, showing that vertebrae collected in the canton of Vaud contain 18% more ^{90}Sr than vertebrae collected in the Basel area, 2% more than in vertebrae from the St-Gallen region and 25% less than in the canton of Tessin. These data confirm that the contamination of the skeleton is coming from the same event (the NBT fallout) and that the canton of Tessin , which has received more deposition from the NBT fallout because of a higher average rainfall rate, shows a slightly higher ^{90}Sr body burden. Our results do not support the assumption that the Swiss NPPs have a potential influence on the ^{90}Sr body burden of the population living within range of the NPPs emission (mostly the Basel region here). Conversely, a higher ^{90}Sr skeleton burden is found in the canton of Tessin, an area which is shielded from the Swiss NPPs by the Alps.

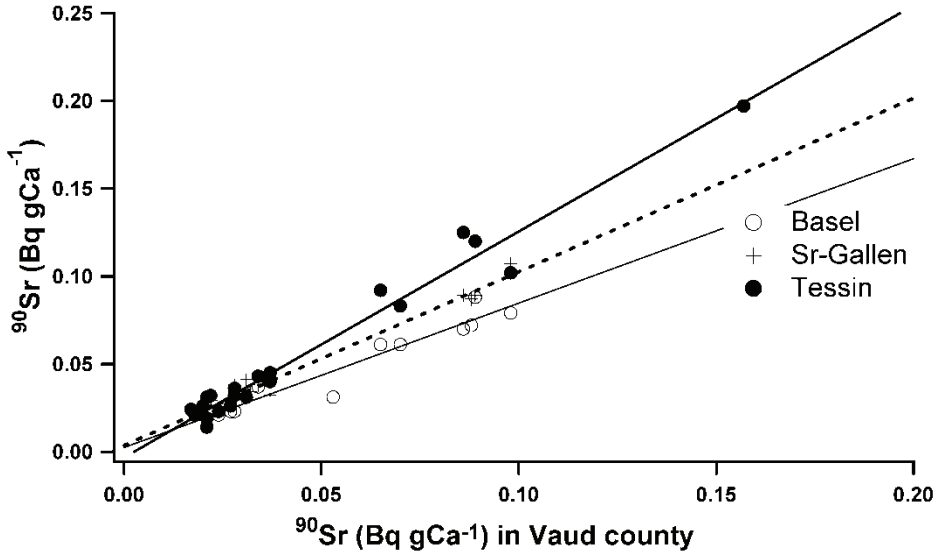

Fig. 7. ^{90}Sr activities (Bq/gCa) in vertebrae of the Basel area, of the St-Gall region and of the canton of Tessin as a function of the ^{90}Sr activities in vertebrae of the canton of Vaud, for the same year of death.

6. Plutonium in bones

If human data for ^{90}Sr are widely available, they are rather scarce for plutonium. USTUR represents probably the most important database for biokinetic modelling of actinides (Kathren, 1995). A review of contributions of human autopsies to the dosimetry of plutonium in man has been further proposed by Kathren (Kathren, 2004). Nevertheless, most of the data concern individuals that have been previously exposed to plutonium

because of their activities in the nuclear industry. Only few studies concern the effect of the NBT fallout on the general population (Taylor, 1995; ODonnell et al., 1997; Takizawa et al., 2000; Froidevaux and Haldimann, 2008); the most extensive is probably McInroy's work on plutonium in autopsy tissues which contains data on more than 260 people, both from the general public in the US as well as workers from the nuclear industry (McInroy et al., 1979). Nevertheless, all these studies were carried out using alpha-spectrometry, which is a less sensitive technique for plutonium determination than mass spectrometry. Moreover, alpha-spectrometry cannot separate ^{239}Pu and ^{240}Pu signals while the ^{240}Pu/^{239}Pu is a very good signature of the origin of the plutonium contamination.

In our work, we determined the ^{239}Pu and ^{240}Pu activities in vertebrae of individual who were mostly older than 20 years at the time of the highest fallout rate of plutonium, using high sensitivity sf-ICP-MS. Data contain results for people who died between 1962 and 2004 in Switzerland. In this respect, our database is probably the most complete database covering the last 40 years for a given population. This allows us to determine, with a significant confidence level, how long plutonium is retained in the skeleton, a very important data in dose assessment that was not previously ascertained (Froidevaux et al., 2010). Results are presented in Figure 8, in which a comparison with published data is included. Because published data usually are given as mBq per kg of wet weight in the former literature, we apply a factor of 4.8% Ca per kg of wet weight and a factor of 1.68 to take into account the ^{240}Pu/^{239}Pu isotopic ratio of the NBT fallout to recalculate the activity in mBq/gCa.

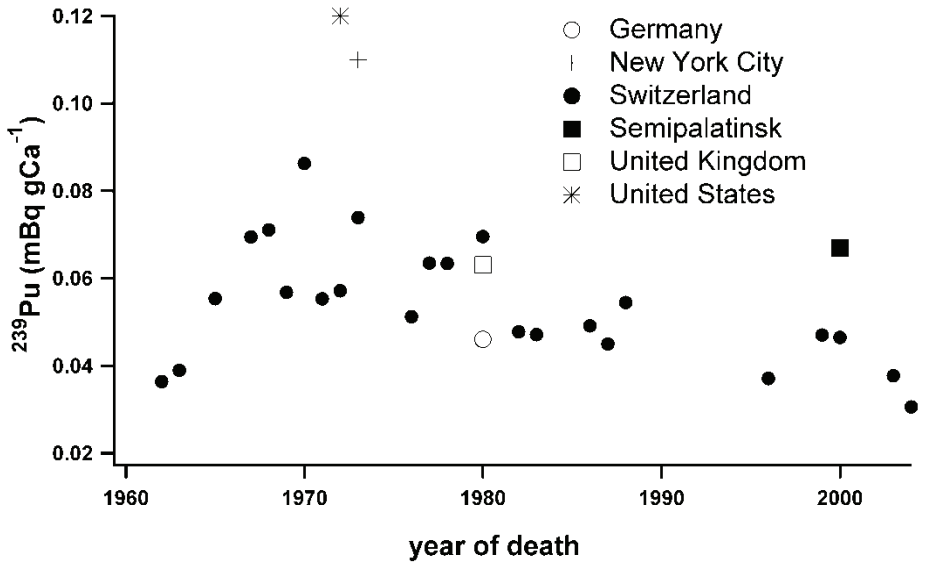

Fig. 8. ^{239}Pu activity (mBq/gCa) as a function of the year of death.

Figure 8 shows that the activities of ^{239}Pu in vertebrae in Switzerland increased similarly to ^{90}Sr, peaking in 1970 at a value of 0.086 mBq/gCa. The ^{240}Pu/^{239}Pu isotopic ratio is 0.18±0.01 (n=48), from which we infer that plutonium measured in this study originated only from the fallout of the atomic bomb tests (Froidevaux et al., 2010). Significantly higher values were found in the US (McInroy et al., 1979) and in New York City (Fisenne et al., 1980), possibly

because these databases contain autopsy tissues from both the general public and workers of the nuclear industry. Similar values to Switzerland were determined in Germany (Bunzl and Kracke, 1983) and in the UK (Popplewell et al., 1985) for the years around 1980. Higher values were obtained at the Semipalatinsk test site (STS) during the 2000's, indicating an effect of the test site fallout in the plutonium body burden of the population (Yamamoto et al., 2006). Using ICP-MS, (Yamamoto et al., 2008) found a significantly lower ^{240}Pu/^{239}Pu isotopic ratio of 0.125 in autopsy tissues (bone) of individuals from the STS, confirming the influence of the STS fallout on plutonium incorporation.
There were too few bone ash samples in our study to separate individuals from different regions, especially the ones potentially affected by the Swiss NPPs. Accordingly, our data represent a pool of bone samples from all over Switzerland. Nevertheless, the ^{240}Pu/^{239}Pu isotopic ratio of 0.18 indicates, beyond any reasonable doubts, that the plutonium inhaled by the Swiss population comes from the fallout of the NBTs of the sixties.

7. Retention half-times in the skeleton of ^{90}Sr and plutonium

The retention half-time in the skeleton of bone-seeking radionuclides such as ^{90}Sr and plutonium is a key parameter used for their dosimetry in humans. Currently, only a partial answer is given to the question of how long plutonium will stay in the body. Values found in the literature are situated between 15 to 100 years, with a proposed value by ICRP 56 or Kathren (1995) of 50 y. Our long-term study of ^{90}Sr and plutonium in the vertebrae allowed us to determine, with a high statistical significance, the retention half-time of both radionuclides in cancellous bones. It is of 40±15 y (95% confidence) for plutonium and 13.5±1.5 for ^{90}Sr (Figure 9). Meanwhile, the retention time of ^{90}Sr is very close to the retention time found in milk teeth, milk, grass and soil (0-5 cm, Table 1).

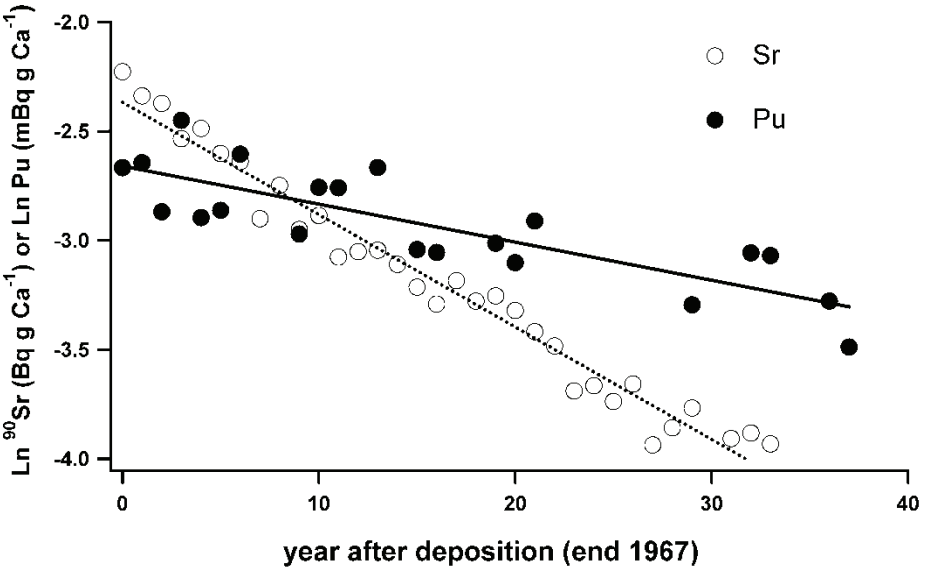

Fig. 9. The use of the data from our long-term study for the determination of the retention time of ^{90}Sr and plutonium in cancellous bones.

Site	Soil (0-5 cm)	Grass	Milk	milk teeth	Vertebrae
Grangeneuve	12.3±3.6	11.6 ±3.9	14.8 ±2.3		
Mühleberg	9.0 ±1.3	7.6 ±1.3	14.5 ±2.6		
Gösgen	7.8 ±0.9	6.7 ±1.1	10.1 ±2.7		
Leibstadt	8.9 ±1.5	12.3 ±3.9	12.5 ±2.5		
Switzerland	**9.5±2**	**9.5±3**	**13±2**	**10.0 ±3**	**13.0 ±1**

Table 1. Retention half-time of ^{90}Sr in different compartments of the environment, food and human for different locations in Switzerland.

These results demonstrate that the calculated retention half-time for ^{90}Sr is in fact an apparent retention half-time because ^{90}Sr is still incorporated in bones after the Nuclear Test Ban Treaty, due to ingestion of contaminated food, especially milk. In this respect, the ^{90}Sr activity in vertebrae is a better reflection of the contamination of the food chain and the environment rather than any mechanism of ^{90}Sr excretion. Consequently, bones remain contaminated by ^{90}Sr as long as environmental contamination lasts (Froidevaux et al., 2010).

8. Conclusion

In this work we show that plutonium and ^{90}Sr from NBTs fallout have contaminated the Swiss population. The level of the contamination is very low and the potential effect of this contamination can be classified within the very low dose effects. In this respect, the NBTs contamination can be viewed as a surrogate for the potential effect that a NPP could have on a nearby population in case of accidental release of low intensity. Compared to other studies conducted worldwide on the same problem, we see that the Swiss population received NBTs fallout similar to other Northern Hemisphere regions but that the incorporation of ^{90}Sr might have been slightly higher because the diet of the Swiss population includes a significant portion of dairy products. The determination of plutonium in milk teeth at a very low-level using sensitive sf-ICP-MS technique allowed us to demonstrate that plutonium does not cross the placental barrier and that the babies were probably born free of plutonium. Nevertheless, the determination of significant amounts of plutonium in bones of adults shows that the incorporation of NBT plutonium in the skeleton of the babies starts as soon as they begin to breathe and continues as long as the plutonium is present in air. ^{90}Sr has been incorporated as a consequence of food contamination, as demonstrated by the strong correlation between the milk activity and the milk teeth activity, and ^{90}Sr in the body will stay in equilibrium with the ^{90}Sr present in the environment. We also show that the analytical part of such a study has to be handled with great care because the levels measured are so low that contamination of the samples by other radionuclides easily happens. In this respect, careful radiochemical work must be carried out on the samples, either for ^{90}Sr or plutonium analyses, otherwise results are submitted to significant bias. In addition, our long-time study allowed us to determine the retention half-time of plutonium and ^{90}Sr in the skeleton. We think that this kind of study forms a very good basis for epidemiological studies involving the effects of a low dose of radiation (Wakeford et al., 2010). We thus conclude that a survey of the population by yearly sampling of milk teeth and vertebrae is very useful to demonstrate an increase in the population body burden that may be attributed to air and/or environmental contamination. In view of the presence of 5 NPPs in Switzerland, this program helps to determine any potential negative effect of the NPPs on the population in case of accidental release. This survey program is well accepted

by the population and offers reassurance that people are not submitted to unacceptable doses of radiation.

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