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^{90}Sr in deciduous teeth from 1950 to 2002: The Swiss experience

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Abstract

Switzerland has recorded the level of activity of ^{90}Sr in the milk teeth of children from different regions of the country since the first atomic explosions in the atmosphere. Activity peaked at 0.421 Bq g^{-1} Ca at the beginning of the sixties, coinciding with the detonation of many large nuclear devices. Following the Nuclear Test Ban Treaty that ended atmospheric nuclear weapon tests, a steady and significant decrease in ^{90}Sr activity in milk teeth has been observed—down to a value of 0.03 Bq g^{-1} Ca for children born in 1994. The apparent half-life of ^{90}Sr in milk teeth is 9.8±3 years. With the exception of the period from 1962 to 1964, there is no correlation between the activity in the teeth of children born in a given year and the year of extraction. Between 1953 and 1992, the milk teeth of children born in Zürich county showed 16% less activity than teeth from children born in Vaud county. Dairy consumption habits might be responsible for this trend. The effect of the ^{90}Sr deposition from Chernobyl is barely measurable in milk teeth, and no effect is seen from the five Swiss nuclear reactors. This paper emphasizes the necessity of a very high purity chemical separation of ^{90}Sr or ^{90}Y to determine ^{90}Sr activity in milk teeth or other samples.

Keywords: Strontium-90; Deciduous teeth; Nuclear bomb tests; Chernobyl; Nuclear power plant

1. Introduction

^{90}Sr is an artificial radionuclide produced by nuclear fission during the explosion of a nuclear device and in nuclear energy plants (Stamoulis et al., 1999). In Switzerland, environmental ^{90}Sr comes from atmospheric nuclear tests performed in the sixties, and more recently from the Chernobyl accident. It has been estimated that about 10^{18} Bq of ^{90}Sr was released in the atmosphere from 1945 to 1965 and subsequently deposited on the earth and in the oceans (Christensen et al., 1975). Hardy et al. (1973) studied the deposition of plutonium from nuclear weapon tests. They reported that the region from 40–50°N was the most heavily contaminated, and that the Arctic region (80–90°N) was the least contaminated, with activities of around 80 Bq m^{-2} and 8.0 Bq m^{-2} respectively. Apostoaei (2002) studied the absorption of strontium from the gastrointestinal tract to the plasma in human adults and found that the fraction of strontium transferred to the plasma (f_1) depends on dietary calcium intake. There is a high variability in f_1 between individuals, but no significant dependence on age or sex. As an alkaline earth cation, ^{90}Sr follows calcium in the food chain–in particular in dairy products–before being deposited in the skeleton and other calcified tissues (Leggett et al., 1982). Teeth are an extension of the skeleton and accumulate contaminant stable and radioactive bone-seeking metals that enter the body. The ^{90}Sr activity in deciduous teeth (milk teeth) reflects the activity in food at around the time of the child's birth and in

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the mother's diet during pregnancy (Webb et al., 2005). Attention has been focused on ^{90}Sr for several decades because it is believed to be the most potentially hazardous of the long-lived fission products that had started to accumulate as a result of nuclear weapon tests.

The level of ^{90}Sr in bones and teeth has been monitored since the beginning of nuclear weapon tests. There were two peaks in the concentration of ^{90}Sr versus year of birth (milk teeth) or death (bone) in 1958 and 1965, followed by an exponential decrease after the entry into force of the Nuclear Test Ban Treaty (Aarkrog, 1971; Hanson and Thomas, 1982; Stamoulis et al., 1999). Recently Mangano et al. (2003) reported on an unexpected rise during the 1990s in ^{90}Sr in milk teeth from children in the United States. They attribute the increase to nuclear power reactors.

In Switzerland, a monitoring program of ^{90}Sr in milk teeth and vertebrae was initiated by the Federal Office of Public Health in the late fifties. Since this time, milk teeth have been collected annually from three regions of Switzerland. Of all the European countries, Switzerland was one of the most contaminated by global fallout due to the geomorphology of the country (Froidevaux et al., 2004). Plutonium deposition can reach 300 Bq m^{-2}, and ^{90}Sr 3500 Bq m^{-2}. The Chernobyl accident added numerous short and long-lived radionuclides, of which only ^{137}Cs is still measurable in the Swiss environment. In the Italian-speaking part of Switzerland (south of the Alps), ^{137}Cs deposition from Chernobyl is as high as 60 000Bq m^{-2} in small areas. Since the beginning of the seventies there has also been a mere potential contribution from nuclear power plants. Switzerland has five reactors producing a total of 3350 MWe.

In this paper we report the results of ^{90}Sr measurements made on milk teeth extracted in Switzerland from 1963 to 2002. We describe briefly the method used until 1971 and in more details the one used from 1972 until now, which permit the determination of strontium activity at very low levels by means of complex but very robust protocols. Our results are compared with the results of Mangano et al. (2003) and other authors (Kulev et al., 1994; O'Donnell et al., 1997; Stamoulis et al., 1999). Differences in between our method and the method of Mangano et al., and the possible impact of these differences on our results are discussed.

2. Method

2.1. Reagents and equipment

All reagents used were analytical grade (Merck, Darmstadt, Germany or Fluka, Buchs, Switzerland). Chromatographic separations were carried out on a Bio-Rad AG 50W-x8 (100–200 mesh) cationic exchanger. Solutions were loaded on to the chromatographic columns (50 ml, 8 ml respectively) by a peristaltic pump (Ismatec IPS-12) with automatic control of the delivered volume, elution rate and the end of elution (bubble detector). Yttrium oxalate sources were counted on a Tennelec LB 4100 low background proportional counter. Background was 0.004 cps (counts per second). The source was typically measured for 100 h with intermediate results recorded automatically every 4 h to check the purity of ^{90}Y. Using this method, a detection limit of $5{\cdot}10^{-3}$ Bq/g Ca can be achieved. Yttrium yield was determined by atomic absorption on the dissolved sources with a Perkin Elmer 4100 apparatus, using a N_2O/acethylene flame.

2.2. Sampling

From 1963 to 2002, milk teeth were collected annually by dentists in three regions of Switzerland. The first region (ZH) includes the county of Zürich and a large part of the Swiss German lowland. The Gösgen, Beznau and Leibstadt nuclear power plants are in this region. The second region is the French-speaking part of Switzerland, mainly the Vaud county (VD). There is a potential influence from the Mühleberg nuclear power plant in this region. The third region is the Italian-speaking part of Switzerland (TI), which is separated from the Swiss nuclear power plants by the Alps.

Each tooth was sent to our laboratory in a plastic bag labeled with the place and date of birth of the child, and the place and date of extraction of the tooth. Only teeth from children born in the specified area (ZH, VD, TI) were used. It was assumed that the mother lived in the area during the pregnancy.

2.2.1. ^{90}Sr measurements

Method used until 1971: After dissolution of the ash in hydrochloric acid, Sr carrier was added, then the alkaline earth were separated as oxalate, heated at 600° to destroy oxalate. After dissolution of the alkaline earth in nitric acid, strontium was specifically separated as nitrate by two precipitations with fuming nitric acid. Strontium was further purified by washing with ethanol followed by iron scavenging. After a partial ingrowth of ^{90}Y, this radionuclide was separated by two successive precipitations as hydroxide, then as oxalate (Lerch et al., 1965). After filtration, the yttrium oxalate precipitate was measured using in-house built low background Geiger counter described by Lerch and Bercier (1964).

Method used from 1972 to 2004: A minimum of 5 teeth from the same region and year of birth were used for each measurement. If necessary, fillings were removed mechanically by crushing the tooth in a mortar. The samples were ashed by heating at 600 °C for 24 h to oxidize the organic matter. Then 5 ml of 0.2 M Sr carrier solution was added and the ashes were dissolved in 8 M nitric acid. After dilution to 250 ml and filtration (if necessary), an aliquot (3 ml) was taken for calcium determination by atomic absorption. Then 4 M ammonium carbamate was added until the pH of the solution reached 8–9. After centrifuging, the carbonate precipitate was dissolved in 8 M nitric acid. The solution was diluted to 500 ml and 125 ml of 0.5 M EDTA was added. The pH of the solution was carefully adjusted to 4.6 and the solution was pumped onto the cationic exchanger (AG 50W-x8, 50 ml). Ca and Mg were eluted from the column with 0.09 M CyDTA (*trans*-1,2-cyclohexylenedinitrilo tetraacetic acid) (pH 5.5). Strontium was eluted with 300 ml of 0.075 M CyDTA (pH 6.5). The solution was adjusted to pH 4.2 and pumped onto the cationic exchanger (AG 50W-x8, 8 ml). The column was then washed with 80 ml of water. Sr was eluted into a flask containing 5 ml of a 0.0225 M Y carrier with 100 ml of 0.2 M sodium citrate (pH 7.5). This solution was left in the refrigerator for at least 12 days until the equilibrium between ^{90}Sr and its daughter product, ^{90}Y, was partially reached. The solution was adjusted to pH 2 and then pumped onto the 8 ml column of the cationic exchanger (AG 50W-x8). The column was washed with 0.75 M NaCl until the pH of the effluent reached 4. Yttrium was eluted with 100 ml of a 0.25 M sodium malonate solution. When half of the solution had passed through the column, the time was recorded to permit a correction for the decrease in ^{90}Y. Strontium was eluted with 125 ml of a 4 M NaCl solution for determination of the strontium chemical yield by atomic absorption. Yttrium oxalate was precipitated from the malonate solution by adding oxalic acid, filtered on a Millipore GSWOP 02400 filter, and then counted in a low background proportional gas counter (Tennelec LB 4100). Counting efficiency, enhanced by backscattering, was 50.0±0.5%. After counting, the yttrium source was dissolved in hydrochloric acid and the chemical yield of yttrium determined by atomic absorption (Perkin Elmer 4100). Using this method, the detection limit is 5 mBq g^{-1} Ca.

The method is described in detail by Geering et al. (1990). Unfortunately, no interlaboratories comparison samples were available to assess the quality of the analytical method for the determination of ^{90}Sr in milk teeth. However, many interlaboratory comparisons and IAEA proficiency tests have been done for quality assurance of the same method for ^{90}Sr measurements on other materials: ^{90}Sr in urine (average bias 3%, PROCORAD 2000, France), ^{90}Sr in soil samples (average bias 7.5%, IAEA ALMERA proficiency test, 2001, Austria), ^{90}Sr in a mineral matrix (average bias 8%, IAEA proficiency test, 1999), ^{90}Sr in water (average bias 5%, Bundesamt für Strahlenschutz, Ringversuch 1/1998, Germany), ^{90}Sr in water (average bias 3%, Bundesamt für Strahlenschutz, Ringversuch 1/2000, Germany), ^{90}Sr in milk powder (average bias 3%, IAEA-152, 1988, Austria), ^{90}Sr in whey powder (average bias 3%, IAEA-154, 1988, Austria). Results show that the method is perfectly designed for the ^{90}Sr determination in various kinds of samples, even with complex matrices.

3. Results

In this study, activities are calculated at, and results are presented as a function of the year of birth. However, milk teeth from children born during the same year can be extracted at different ages. Therefore it is possible that, in the case of an abrupt increase in the level of ^{90}Sr in the food chain, the milk teeth of children born at the time of the event but extracted at different ages will have different ^{90}Sr activities due to the continuous exchange of calcium between the blood plasma and the tooth root zone (Shishkina et al., 2001). Thus milk teeth extracted from older children might have a lower ^{90}Sr content when contamination in the food chain is decreasing, and a higher content when it is increasing. This was checked using the data collected in Vaud county since 1963. For each year of birth, milk teeth extracted in at least five different years were collected. Table 1 shows the results of a statistical analysis of at least 10 (but usually 20 to 30) activity determinations for each extraction period. A *t*-test on the data gives the parameter P, which represents the probability that the correlation between the measured activity in a given birth year and the year of extraction is due to random chance. Fig. 1 shows that the probability P is less than 5% only for the years 1962–1964, when activities peaked at their highest values and there were strong variations in activity over short periods of time. For the other periods, P is too high to conclude that the extraction year has an effect on the ^{90}Sr activity of a given birth year. When the contamination level of the food chain does not change significantly, there is no significant difference between teeth from the same year of birth that have been extracted in different years. For example, the reduced centered variables $(x-x_m/\sigma)$ for the year of birth 1983 and the period of extraction [1992–2000] are within the

Table 1
Probability that the correlation observed between the ^{90}Sr activity of teeth for a given year of birth and the year of extraction is due to random chance, calculated as a Student's law (t)

Year of birth	Years of extraction	N	t	P	Slope
1955–1956	1964–1968	29	0.473	0.650	−0.001
1956–1957	1964–1969	40	0.042	0.967	0.0002
1958–1959	1966–1971	24	0.409	0.69	−0.0010
1960	1966–1973	29	−0.529	0.61	−0.0016
1961	1968–1975	29	2.867	0.017	0.0167
1962	1969–1975	30	3.933	0.0034	0.0337
1963	1971–1976	33	4.989	0.0011	0.0643
1964	1969–1976	21	2.938	0.0148	0.0140
1965	1970–1976	18	3.121	0.0168	0.0120
1966–1967	1972–1976	17	1.893	0.088	0.01522
1967–1968	1972–1976	10	0.419	0.685	0.0027
1978–1979	1988–1991	7	0.050	0.962	−0.0003
1980–1981	1988–1991	11	0.447	0.66	−0.00092
1982–1983	1988–1999	12	1.424	0.1778	−0.00092
1984–1985	1993–2000	10	1.665	0.130	−0.00128
1986–1987	1996–2000	11	0.873	0.401	−0.00146
1988–1990	1997–2001	10	1.703	0.120	0.00203

N is the number of measurements.

intervals [−1.3; 1.3] for Zürich county, [−1.2; 0.13] for the French-speaking part of Switzerland and [−1.5; 1.3] for Tessin county. No outlier was detected, which means that within $\pm 2\sigma$, the distribution is not changing significantly. Table 1 and Fig. 1 show that the slope of ^{90}Sr activity for a given year of birth as a function of different years of extraction is only significantly different from zero for the corresponding low probabilities P. Also, the slopes are shallow, indicating that the accumulation of ^{90}Sr in milk teeth during childhood is low. This means that milk teeth from different collection years can be pooled to give a larger sample size with more statistically significant measurement results for each birth year. Table 2 and Fig. 2 show the results for the three regions. There is a peak in ^{90}Sr activity in milk teeth in 1964 and a sharp decline from then until now. A shoulder associated with the main peak corresponds to the years 1958–1959, when the United States and the Soviet Union suspended atmospheric nuclear weapon tests. The sharp increase in ^{90}Sr activity in milk teeth at the beginning of the sixties is the consequence of the

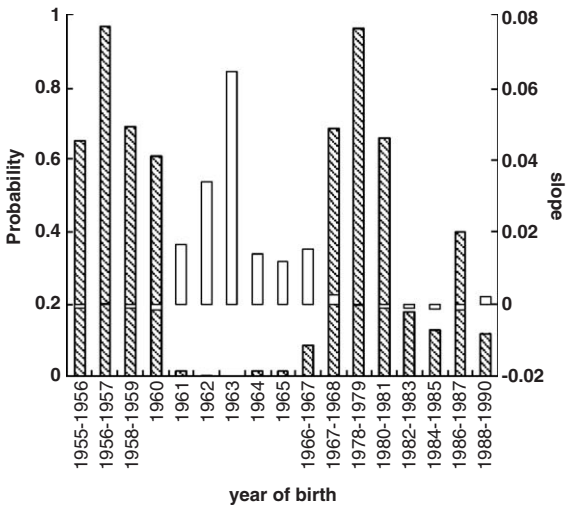

Fig. 1. Probability (striped bars) that the correlation between the ^{90}Sr activity of milk teeth for a given year of birth and different years of extraction is due to random chance, calculated as a Student's law and corresponding slopes (white bars).

Table 2
^{90}Sr activity (Bq g^{-1} Ca) in milk teeth collected in three different regions of Switzerland as a function of the year of birth

Year of birth	Vaud county		Tessin county		Zürich county	
	N	Bq g^{-1} Ca	N	Bq g^{-1} Ca	N	Bq g^{-1} Ca
1951					3	0.020±0.003
1952					4	0.026±0.005
1953	5	0.059±0.003			3	0.034±0.001
1954	13	0.089±0.010			3	0.051±0.005
1955	11	0.109±0.010			2	0.086±0.006
1956	18	0.116±0.007			2	0.093±0.020
1957	22	0.146±0.010			1	0.114±0.008
1958	24	0.161±0.012			7	0.142±0.007
1959	23	0.158±0.012			7	0.141±0.004
1960	32	0.155±0.010	2	0.138±0.02	18	0.141±0.005
1961	27	0.177±0.009	1	0.184±0.012	28	0.161±0.010
1962	30	0.279±0.009	4	0.208±0.005	34	0.253±0.010
1963	33	0.421±0.015	6	0.357±0.007	44	0.353±0.015
1964	21	0.388±0.014	5	0.324±0.002	42	0.309±0.013
1965	18	0.294±0.011	5	0.270±0.005	44	0.257±0.014
1966	9	0.238±0.024	3	0.227±0.004	31	0.185±0.008
1967	6	0.181±0.013	2	0.202±0.007	20	0.161±0.009
1968	3	0.166±0.017			9	0.146±0.015
1969	1	0.138±0.005			3	0.154±0.013
1977			2	0.066±0.019		
1978	2	0.077±0.010	4	0.058±0.012	2	0.050±0.013
1979	3	0.064±0.008	4	0.062±0.009	4	0.059±0.020
1980	5	0.069±0.008	6	0.070±0.009	4	0.043±0.005
1981	6	0.054±0.003	5	0.064±0.010	10	0.045±0.003
1982	8	0.044±0.003	4	0.052±0.006	6	0.040±0.003
1983	5	0.039±0.004	8	0.045±0.004	8	0.042±0.003
1984	5	0.043±0.002	5	0.043±0.004	9	0.043±0.007
1985	4	0.037±0.002	6	0.049±0.004	7	0.038±0.002
1986	7	0.043±0.006	6	0.050±0.003	7	0.038±0.001
1987	5	0.031±0.003	4	0.052±0.004	5	0.034±0.002
1988	5	0.037±0.003	3	0.044±0.004	3	0.031±0.003
1989	3	0.035±0.003	1	0.040±0.005	4	0.024±0.003
1990	3	0.029±0.003	1	0.032±0.003	3	0.023±0.003
1991	2	0.028±0.003	2	0.027±0.003	1	0.020±0.003
1992			2	0.028±0.003	2	0.025±0.003
1993	1	0.017±0.003				
1994			2	0.020±0.004	2	0.020±0.003
1996	2	0.025±0.003			2	0.028±0.003

N is the number of measurements with at least 5 teeth used for each measurement.

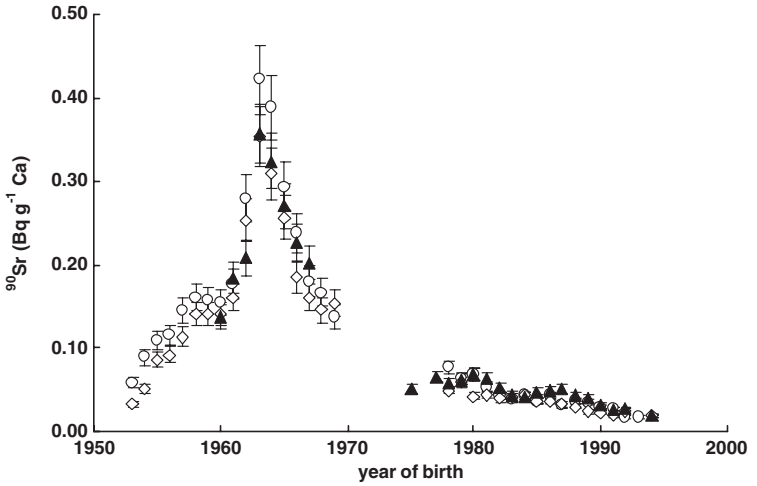

Fig. 2. ^{90}Sr activities (Bq g^{-1} Ca) in milk teeth extracted from Swiss children from 1963 to 2001, for three different regions: Zürich county (lozange), Vaud county (circle) and Tessin county (triangle).

detonation of many very powerful devices. After the entry into force of the Partial Nuclear Test Ban Treaty that ended American, British and Soviet atmospheric nuclear weapon tests, a large and regular decrease in ^{90}Sr activitiy in milk teeth is observed. Unfortunately, budget restrictions prohibited the collection of milk teeth for children born between 1970 and 1977. The program was restarted after the Chernobyl nuclear power plant accident and teeth have been collected for children born from 1978 up until now. A fit of the data from 1964 to 1996 with an exponential curve, gives an apparent half-life for ^{90}Sr in milk teeth of 9.8±3 years. This is close to the half-life of 9±2 years in grass and soil and 13±3 years in milk as determined by Friedli et al. (1991) in Switzerland.

An examination of the period from 1975–1994 reveals several interesting features (Fig. 3). The Chernobyl nuclear power plant accident occurred in 1986. At close examination, the Chernobyl contribution is visible. Following the Chernobyl accident, special attention was given to milk and milk products in Switzerland. Typically, values for milk increased temporarily by a factor of 2, passing from 0.1 Bq kg^{-1} to 0.2 Bq kg^{-1} in some regions of Switzerland (Geering et al., 1990). Interestingly, this Chernobyl ^{90}Sr has vanished rapidly from the upper layer of the soil, indicating that its chemical form was different from the one deposited during the sixties. Thus, milk activity returned to the pre-Chernobyl value within a few months (Friedli et al., 1991). In spite of the increase in milk activity for the few months following the accident, only a very slight increase in ^{90}Sr in milk teeth was observed for children born in 1986 and 1987. In Switzerland, the increase was largest in Tessin county where a higher deposition of ^{137}Cs from the Chernobyl cloud also occurred (Fig. 3).

Froidevaux et al. (2004) showed that ^{90}Sr activity in cheeses produced in Western Europe are strongly correlated with grazing altitude. When cows graze between 100 and 400 m in altitude, cheeses have ^{90}Sr activities between 0.028 and 0.036 Bq g Ca^{-1}. When cheeses are produced at higher altitudes (>700 m) activities of up to 0.12 Bq g Ca^{-1} are observed. This is mainly due to the higher deposition of radionuclides in mountainous areas, often correlated with precipitation rates (Wright et al., 1999; Hölgye and Filgas, 1995). This leads to different ^{90}Sr body burdens for people living in different regions.

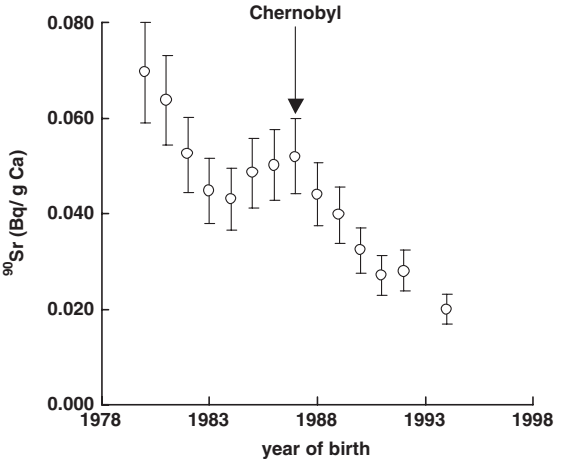

Fig. 3. Activities in milk teeth of Tessin county children born between 1980 and 1994. The arrow highlights the effect of the Chernobyl accident.

4. Discussion

4.1. Analytical method

The chemical separation method used in this work for the determination of ^{90}Sr in milk teeth gives results with a very low detection limit (5 mBq g^{-1} Ca), low measurement uncertainties (±15% at 95% of confidence level) and an efficient means of checking the source purity by measuring the half-life, which should be close to 64 h for a pure ^{90}Y source (Fig. 4). This method has been used in our laboratory to measure ^{90}Sr in milk teeth and human vertebrae, as well as in milk, grass, wheat, vegetables and soil since 1972 (Geering et al., 1990). Prior to 1972, a strontium specific precipitation by fuming nitric acid followed by iron scavenging was used. After a partial ingrowth of ^{90}Y, this radionuclide was separated by two successive precipitations as hydroxide, then as oxalate (Lerch et al., 1965). This method was replaced in 1972 by the chromatographic separation of strontium and yttrium described above. No difference was observed between the results obtained from the two methods. This is as expected since the results depend only on the primary ^{90}Y source used to calibrate the proportional counters used to measure source activity. Nevertheless the chromatographic method is preferred because it can be partly automated and is less hazardous for the operator than the fuming nitric acid method. Using a combination of extractions by TBP and precipitation steps, Neuzil and Dysart (1984) demonstrated that ^{90}Sr and ^{228}Ra can be determined in teeth samples, due to the fact that ^{228}Ac follows yttrium in the chemical separation scheme. They concluded that the radium isotope accumulates from the decay of its parent ^{232}Th which appears to be deposited in teeth as a phosphate in calcium-bearing tissue. Meanwhile the ^{228}Ra activity is equal to the ^{90}Sr activity in the teeth of children from 0–10 years of age, at a level of 0.030 Bq g^{-1} Ca. Thus if the chemical separation does not allow a complete purification of the dosed radionuclide (^{90}Sr or ^{90}Y), a large overestimation of the ^{90}Sr activity is possible. Altzitzoglou et al. (1998) made similar observations for the measurement of ^{90}Sr in bone ash. Stamoulis et al. (1999) and O'Donnell et al. (1997) used a complete separation of either ^{90}Y or ^{90}Sr to assess the level of ^{90}Sr in milk and permanent teeth.

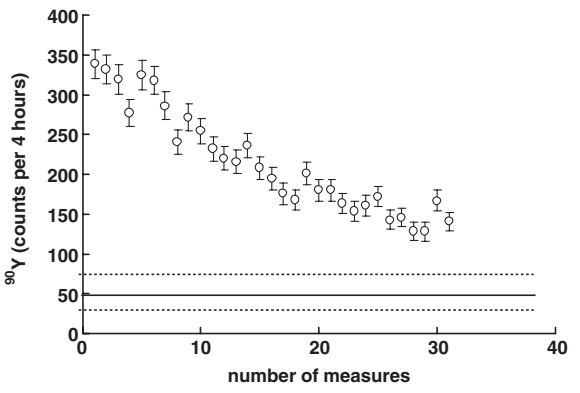

Fig. 4. Activity (counts per 4 h) determined for a yttrium oxalate source obtained from 3 g of ashed milk teeth from Tessin county children born in 1992 (teeth extracted in 2002). Black line represents the background. Dotted lines are the upper and lower limits of the confidence interval for the background (2σ).

In the method used by Mangano et al. (2003), it is likely that radium and actinium are not completely separated from thorium. Because these two radionuclides are in secular equilibrium with their parent, ^{232}Th, this will add about 100 mBq g^{-1} Ca to the ^{90}Sr activity. At current levels of activity (e.g. 30 mBq g^{-1} Ca), this may be sufficient to account for the increase in milk teeth activity reported by the authors. In the method of Mangano et al. it is also possible that eventually other natural radionuclides (e.g. lead isotopes) will follow strontium. Also, the liquid scintillation counting method used for activity determination is not specific enough to the ^{90}Sr/^{90}Y pair. Any other beta emitters present in the flask will also be counted. If Mangano's method might be satisfactory to evaluate ^{90}Sr in case of a large increase in milk teeth activity, it is compulsory to use a method that allows a total decontamination of the ^{90}Sr or ^{90}Y sources when measuring them in milk teeth at present time ^{90}Sr activity. Altzitzoglou et al. (1998) emphasized the necessity of a complete chemical separation scheme and the importance of using a sufficient quantity of material when undergoing a rigorous certification process to separate ^{232}Th, natural U, $^{239/240}$Pu, ^{241}Am, ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{210}Po and ^{90}Sr in bone ash samples.

Problems can arise from the use of a very small sample. Mangano et al. used 100 mg of tooth ash. In our study, a usual amount of 3–5 g of teeth ash is used (pool of at least 5 teeth from children from the same region, born during the same year), leading to a ^{90}Y signal about 4 times higher than the background at the year 2002 level of activity, the background signal being 0.004 cps (Fig. 4). Using 100 mg of tooth ash would not allow us to quantify the tooth activity. Stamoulis et al. (1999) also emphasized the necessity of pooling teeth to obtain larger samples with enough ash (4–10 g) for ^{90}Sr analysis, while O'Donnell et al. (1997) used batches of at least 50 permanent teeth. Aarkrog (1971) used a smaller quantity of tooth ash during the sixties due to

higher levels of activity, but not less than 2–5 g. As a quality control assessment, Mangano et al. measured milk teeth of children from the Philippines born in 1991 and 1992. They obtained a value of 75 mBq g^{-1} Ca. In Switzerland, this is the value obtained for children born during the 1970s. The Philippine Archipelago is situated between 8°N and 20°N while Switzerland is situated at around 46°N. UNSCEAR (1977) indicated an average integrated ^{90}Sr deposition density of 3×10^{9} Bq km^{-2} for the latitude 40–50°N. Deposition between 10 and 20°N is three times lower. Thus it is doubtful that Philippine children born in the early 1990s would have had such a high level of ^{90}Sr activity in their milk teeth. Using a method that allowed a total decontamination of the measured ^{90}Y source, Stamoulis et al. (1999) measured the ^{90}Sr activity in milk teeth from Greek children of 0–10 years of age born during the nineties and obtained a value close to 0.030 Bq g^{-1} Ca. This is similar to the value obtained in Switzerland, even though Greece was more heavily contaminated by the Chernobyl accident. Activities as high as those reported by Mangano et al. have not been measured elsewhere, even in places where the ^{90}Sr deposition from the Chernobyl accident was measurable.

4.2. Radioecological considerations

Switzerland is characterized by large variations in geomorphology, with three distinct regions: the Swiss lowland, the Jura mountains, and the Alps. The highest ^{137}Cs, ^{90}Sr and plutonium depositions were in the Alps and the Jura Mountains (Froidevaux et al., 2003). Part of Tessin county was also more contaminated than the Swiss lowland due to topography and high rainfall. Higher ^{137}Cs deposition from the Chernobyl accident was also detected there. On the other hand, Tessin county is separated from the Swiss nuclear power plants by the Alps, excluding any potential effects of the nuclear power plants on the level of ^{90}Sr in milk teeth. Meanwhile there is no difference in the level of ^{90}Sr in the milk teeth of children born in Tessin county compared with children born in lowland Switzerland; the maximum value is 0.360 Bq g^{-1} Ca in Tessin county, 0.421 Bq g^{-1} Ca in Vaud county and 0.353 Bq g^{-1} Ca in Zürich county. Variation in ^{90}Sr content from place to place and sample to sample is the rule rather than the exception. High inhomogeneity has been observed for the Chernobyl and fallout depositions (Schimmack et al., 2003; Pourcelot et al., 2003). Transfer of radioactivity from the environment to the food chain varies between regions and depends on the altitude of production of dairy products (Froidevaux et al., 2004). Natural radioactivity is also highly inhomogeneous in the environment. The radium content of well, mineral and tap water can vary greatly, giving different activity levels in the skeleton and teeth, depending on consumption habits. Consumption habits for milk and diary products are of particular importance when attributing the variation in ^{90}Sr activity in milk teeth to different factors, including the presence of a nuclear reactor in the vicinity. An assessment of the ^{90}Sr content of milk is essential. In our opinion it is not possible to explain an increase in milk teeth activity, without observing the same increase in milk samples. An evaluation of the average ^{90}Sr deposition and content in milk is necessary for assessing the body contamination in a given region. In Switzerland, higher activities are present in milk produced in upland Switzerland (Alps) than in lowland Switzerland. No effect of the nuclear power plants on milk activity has been observed (e.g. Geering et al., 1997).

Fig. 5 shows a comparison of the ^{90}Sr activity in milk teeth from children from Vaud and Zürich counties between 1953 and 1992. The ^{90}Sr activities are strongly correlated (r=0.99), indicating the same evolution of contamination of the food chain in both regions. Yet, the value of the slope of the linear regression is 0.84. This means that milk teeth from Zürich county contained on average 16% less ^{90}Sr than milk teeth from the French-speaking part of Switzerland between 1953 and 1992. One explanation for this difference is the different average deposition of ^{90}Sr fallout from nuclear tests, leading to a different level of contamination of diary products. An alternative explanation is a difference in consumption habits. In fact, the Zürich county is an urban region where the consumption of dairy products may be less than in Vaud county, which includes large farming areas. Note that Zürich county is within 50 km

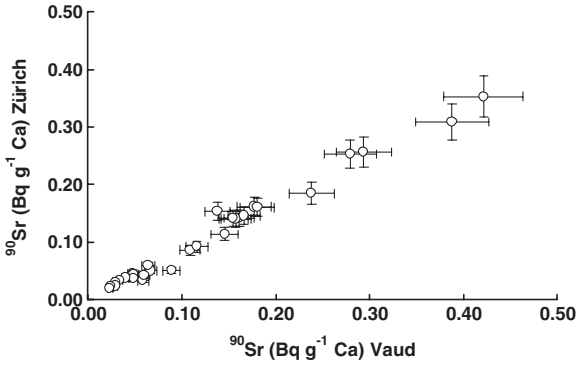

Fig. 5. Activity of ^{90}Sr in milk teeth (Bq g^{-1} Ca) from Zürich county as a function of ^{90}Sr activity in milk teeth from Vaud county, for the same year of birth (from 1953 to 1990).

of four nuclear reactors (3000 MWe), while Vaud county is 150 km from a reactor, and Tessin county is separated from the nuclear power plants by the Alps. Still, the ^{90}Sr activity in milk teeth of children born in Zürich county is currently the lowest of the three regions. The activity in milk teeth from Tessin county is slightly higher than the other regions due to a higher deposition of ^{90}Sr from the global fallout and the Chernobyl accident in that part of Switzerland (Fig. 3). ^{90}Sr activities in food samples (e.g. milk and wheat) and bones or teeth in Switzerland since the start of the year 2000 show evidence of an equilibrium situation where activities expressed as Bq g Ca^{-1} are almost identical. The activity in milk produced in the Swiss lowland is between 0.04 and 0.06 Bq g Ca^{-1}. For wheat it is between 0.1 and 0.15 Bq g Ca^{-1} (Völkle et al., 2003). The average activity of vertebrae sampled from people deceased during 2003 was 0.035 ± 0.010 Bq g Ca^{-1}, a value very close to the value reported for milk. Milk teeth measured during 2003 for children born between 1987 and 1992 averaged 0.030 ± 0.010 (Froidevaux et al., 2003). Thus the ^{90}Sr activity in milk, milk teeth and vertebrae has reached an equilibrium value of about 0.030 Bq g Ca^{-1} throughout lowland Switzerland. The trend in activities from the Zürich, Tessin and Vaud areas towards the same value of about 0.030 mBq g Ca^{-1} indicates a downward trend in the transfer factor from soil-to-milk due to the migration of ^{90}Sr to depth in the soil and to irreversible adsorption mechanisms in soil. As well, modern dairy farming practices involve the mixing of milk from numerous farms before redistribution around the country. As a consequence, the activity in milk and in milk teeth is averaged out. Stamoulis et al. (1999) observed that the ^{90}Sr profile for milk teeth is similar to that for human vertebrae. This is also true for Switzerland. Since the beginning of the 1980s milk teeth and human vertebrae have had almost the same ^{90}Sr activity, decreasing steadily down to 0.030 Bq g^{-1} Ca in 2001 (Geering et al., 2001). Any suspected important increase in ^{90}Sr activity in milk teeth should be verifiable by measurements of ^{90}Sr in milk and in human vertebrae of adults from the same area, using a radiochemical method that allows for a total decontamination of the strontium (or yttrium) source from natural radionuclides. O'Donnell et al. (1997) did not observe any increase in ^{90}Sr in the milk teeth of children living in the proximity of the Sellafield reprocessing plant. They reported an average activity of 8 Bq kg^{-1} ash wt. Using a calcium content of 0.370 kg Ca per kg of ash (average of 130 milk teeth from Swiss children born between 1988 and 1997) we calculate an activity of 0.021 Bq g Ca^{-1} for O'Donnell's study. In a study carried out in South Ukraine between 1990 and 1991, Kulev et al. (1994) observed that measured ^{90}Sr concentrations of human teeth ($N=1000$) were lower by a factor of 10 compared with measurements made in the mid-1960s and mid-1970s. The effect of the Chernobyl accident was measurable as a 3-fold increase in activity in the teeth of the male population aged 25–45 years, possibly due to the mobilization of this age group for clean-up operations within the 30-km zone around the damaged reactor. The maximum value reported by Kulev et al. was less than 50 mBq g Ca^{-1}. This is much lower than the 100–180 mBq g Ca^{-1} reported by Mangano et al. (2003). The maximum activities reported for milk teeth during the peak period from 1962–1964 were 0.350 Bq g Ca^{-1} in the United States (Mangano et al., 2003), 0.250 Bq g Ca^{-1} as a world-wide average (Stamoulis et al., 1999), 0.240 Bq g Ca^{-1} in Denmark (Aarkrog, 1971) and 0.421 Bq g Ca^{-1} in Switzerland (this work). Thus neither our study, nor Stamoulis's, nor Kulev's nor O'Donnell's studies confirm Mangano's observations for ^{90}Sr.

4.3. Limitations of the scope and method

This report represents a long-term study of in vivo ^{90}Sr levels in Switzerland that has been carried out under the direction of the Federal Office of Public Health. Another aspect of this study, the yearly sampling of vertebrae from deceased people by different pathology institutes in Switzerland, is not discussed here. This program of the survey of the radiation exposure of the Swiss population is complemented by an analysis of ^{90}Sr in milk sampled all over the country. One limitation of this work concerns the extrapolation of the results to the entire Swiss population. Milk teeth were collected mainly from the Swiss lowland, while it was observed that dairy products from mountainous regions were more contaminated by ^{90}Sr than lowland dairy products (Froidevaux et al., 2004). This could introduce variations in ^{90}Sr exposure that are not taken into account in this work. It is also worth noting that farming habits changed from the sixties to the nineties from small independent production units to larger farms with centralized milk collection and processing. This makes it difficult to compare different regions in Switzerland when, for example, the pooling of milk between the Zürich and Vaud regions may be responsible for the convergence of activities in milk and milk teeth in these two regions in the eighties and nineties.

The Federal Office of Public Health restarted the milk teeth program following the Chernobyl nuclear power plant accident, recognizing the importance of the information obtained from ^{90}Sr determination in

these samples. Today the milk teeth program, and also the determination of ^{90}Sr in vertebrae, is part of an ongoing national program of environmental radioactivity monitoring.

5. Conclusions

The Swiss experience monitoring ^{90}Sr activity in milk teeth shows a sharp increase in activity that is attributable to atmospheric nuclear weapon tests that were carried out by the Soviet Union, the United States and, to a lesser extent, England and France during the 1950s and the 1960s. Since the entry into force of the Partial Nuclear Test Ban Treaty that ended atmospheric tests in 1963, the ^{90}Sr activity in the milk teeth of Swiss children has decreased rapidly, with an apparent half-life of about 10 years. The Chernobyl nuclear power plant accident doubled the ^{90}Sr activity in milk for a period of a few months, but the resultant increase in activity in the milk teeth of children born in 1986 was barely measurable. In contrast with the work of Mangano et al. (2003), no increase in ^{90}Sr has been observed in the milk teeth of Swiss children living close to nuclear power plants during the 1990s. Milk teeth activities determined in Switzerland during the nineties are similar (within the limits of uncertainty) to the activities reported by Kulev et al. (1994), O'Donnell et al. (1997) and Stamoulis et al. (1999), but not to the milk teeth activities reported by Mangano et al. (2003).

The method used for ^{90}Sr determination in milk teeth in Switzerland permits a complete separation of ^{90}Sr/^{90}Y from other natural and artificially occurring radionuclides, and allows the purity of the measured source to be checked. A very low detection limit is obtained by using a large quantity of tooth ash. Statistical analysis of the data shows no correlation between the activity of the teeth and the year of extraction, with the exception of teeth from children born during the period 1962–1964, when large changes in the ^{90}Sr activity in the environment and food chain were observed. Measurements of milk teeth collected in Vaud county show 16% more activity than those collected in Zürich county. This work demonstrates the importance of applying a very careful chemical separation scheme with high decontamination factors for ^{90}Sr or ^{90}Y from natural uranium and thorium series when measuring ^{90}Sr in milk teeth or other samples. It is likely that the absence of a high purity chemical separation of strontium or yttrium from natural uranium and thorium series, and the use of only a very small amount of tooth ash, gave rise to the elevated activities reported for milk teeth in the United States by Mangano et al. compared with previous measurements.

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References

Aarkrog A. Prediction models for ^{90}Sr in shed deciduous teeth and infant bone. Health Phys 1971;21:803–9.

Altzitzoglou T, Larosa JJ, Nicholl C. Measurement of ^{90}Sr in bone ash. Appl Radiat Isotopes 1998;49:1313–7.

Apostoaei AI. Absorption of strontium from the gastrointestinal tract into plasma in healthy human adults. Health Phys 2002; 83:56–65.

Christensen GC, Alstad J, Evale E, Pappas AC. ^{90}Sr in human bone in Norway 1956–1972. Health Phys 1975;28:677–84.

Friedli C, Geering JJ, Lerch P. Some aspects of the behaviour of ^{90}Sr in the environment. Radiochim Acta 1991;52:237–40.

Froidevaux P, Friedrich-Bénet K, Barraud F, Schmittler T, Valley JF. Mesures de ^{90}Sr dans les vertèbres et les dents de lait. Environmental radioactivity and radiation exposure in Switzerland; 2003. chap. 6.2, SFOPH, 1700 Fribourg 2003, ISBN3-905235-44-7.

Froidevaux P, Geering JJ, Pillonel L, Bosset JO, Valley JF. ^{90}Sr, ^{238}U, ^{234}U, ^{137}Cs, ^{40}K and $^{239/240}$Pu in Emmental type cheese produced in different regions of Western Europe. J Environ Radioact 2004; 72:287–98.

Geering JJ, Friedli C, Lerch P. Method of determination of ^{90}Sr in the environment. J Trace Microprobe Tech 1990;8:211–30.

Geering JJ, Froidevaux P, Valley JF, Burger M, Holzer R, Figueiredo V, et al. Mesures de strontium-90 dans le sol, l'herbe, le lait et le froment. Environmental radioactivity and radiation exposure in Switzerland; 1997. chap. 7.2, SFOPH, 1700 Fribourg 1997, ISBN3-905235-28-5.

Geering JJ, Barraud F, Froidevaux P, Valley JF. Mesure de ^{90}Sr dans les vertèbres et les dents. Environmental radioactivity and radiation exposure in Switzerland; 2002. chap. 6.2, SFOPH, 1700 Fribourg 2001, ISBN3-905235-40-4.

Hanson WC, Thomas JM. Prediction of ^{90}Sr body burdens and radiation dose in Anaktuvuk Pass Alaska Esquimos due to fallout. Health Phys 1982;43:323–33.

Hardy EP, Krey PW, Volchok HL. Global inventory and distribution of fallout plutonium. Nature 1973;241:444–5.

Hölgye Z, Filgas R. Inventory of ^{238}Pu and $^{239/240}$Pu in the soil of Czechoslovakia in 1990. J Environ Radioact 1995;27:181–9.

Kulev YD, Polikarpova GG, Prigodeyb EV, Assimakopoulos PA. Strontium-90 concentration in human teeth in South Ukraine, 5 years after the Chernobyl accident. Sci Total Environ 1994; 155:215–9.

Leggett RW, Eckerman KF, Williams LR. Strontium-90 in bone: a case study in age-dependent dosimetric modeling. Health Phys 1982;43:307–22.

Lerch P, Bercier P. Nouvelle installation de comptage à bas-niveau pour la mesure du strontium-90 dans les os humains. Chimia 1964;18:16–20.

Lerch P, Geering J-J, Delay A. Méthode radiochimique d'analyse du strontium-90 dans les os en présence de contaminants radioactifs. Chimia 1965;19:43–4.

Mangano JJ, Gould JM, Sternglass EJ, Sherman JD, McDonnell W. An unexpected rise in strontium-90 in deciduous teeth in the 1990. Sci Total Environ 2003;317:37–51.

Neuzil EF, Dysart ME. The determination of ^{90}Sr and ^{228}Ra in human teeth by age groups and in other substances. Int J Appl Radiat Isot 1984;35:1113–5.

O'Donnell RG, Mitchell PI, Priest ND, Strange L, Fox A, Henshaw DL, et al. Variation in the concentration of plutonium, strontium-90 and total alpha-emitters in human teeth collected within the British Isles. Sci Total Environ 1997;201:235–43.

Pourcelot L, Louvat D, Gauthier-Lafaye F, Stille P. Formation of radioactivity enriched soils in mountain areas. J Environ Radioact 2003;8:215–33.

Schimmack W, Kracke W, Sommer M. Spatial variability of fallout-^{90}Sr in soil and vegetation of an alpine pasture. J Environ Radioact 2003;65:281–96.

Shishkina EA, Lyubashevskii NM, Tolstykh EI, Ignatiev EA, Betenekova TA, Nikiforov SV. A mathematical model for calculation of ^{90}Sr absorbed dose in dental tissues: elaboration and comparison to EPR measurements. Appl Radiat Isotopes 2001;55:363–74.

Stamoulis KC, Assimakopoulos PA, Ioannides KG, Johnson E, Soucacos PN. Strontium-90 concentration measurements in human bones and teeth in Greece. Sci Total Environ 1999; 229:165–82.

UNSCEAR. Sources and Effects of Ionizing Radiation. United Nation Scientific Committee on the Effects of Atomic Radiation 1977 report to the General assembly, vol. 40 (A/32/40). New York; 1977.

Völkle H, Andrey JL, Estier S, Ferreri G, Gurtner A, Jungck M, et al. Environmental Radioactivity and Radiation Exposure in Switzerland; 2003. chap. 5, SFOPH, 1700 Fribourg 2003, ISBN3-905235-44-7.

Webb E, Amarasiriwardena D, Tauch S, Green EF, Jones J, Goodman AH. Inductively coupled plasma-mass (ICP-MS) and atomic emission spectrometry (ICP-AES): versatile analytical techniques to identify the archived elemental information in human teeth. Microchemical Journal 2005;81:201–8.

Wright SM, Howard BJ, Strand P, Nylén T, Sickel MAK. Prediction of ^{137}Cs deposition from atmospheric nuclear weapons tests within the Arctic. Environ Pollut 1999;104:131–43.