Available online at www.sciencedirect.com
SCIENCE DIRECT
Science of the Total Environment 367 (2006) 596–605
Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Humankind
www.elsevier.com/locate/scitotenv

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

P. Froidevaux *, Jean-Jacques Geering, J.-F. Valley

Institute of Applied Radiophysics, University of Lausanne, Grand Pré 1, CH-1007 Lausanne, Switzerland

Received 1 February 2006; accepted 1 February 2006
Available online 20 March 2006

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

* Corresponding author. Tel.: +41 21 623 34 80; fax: +41 21 623 34 35.
E-mail address: pascal.froidevaux@inst.hospvd.ch (P. Froidevaux).

0048-9697/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2006.02.011

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).