Investigation of Strontium-90 intake in teeth of children living near Chernobyl

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Introduction

The catastrophe in reactor block no 4 of the nuclear power plant of Chernobyl resulted in a release of radioactive material. This fallout affected especially the region of Chernobyl, but also whole Europe and could be detected worldwide. Certain radioactive nuclides such as jodine (^{131}I, ^{129}I), cesium (^{137}Cs, ^{134}Cs) and strontium (^{90}Sr, ^{89}Sr) as well as plutonium and americium nuclides mainly contributed to the contamination of man and environment.

Even 20 years later experts dispute about the consequences of this radioactive exposition of man (IAEA report, IPPNW report). The effects of the radioactive iodine nuclides on an increased incidence of thyroidal cancers in exposed populations could be proven (Prof. Edmund Lengfelder of the Otto Hahn Strahleninstitut Munich). Longterm effects caused by ^{137}Cs and ^{90}Sr cannot be definitely estimated. Incorporated ^{137}Cs will spread throughout in the whole body. Whereas ^{90}Sr will be deposited mainly in teeth and bones, that close to the marrow, where blood formation can be disturbed (leukaemia).

Considerable activities can be detected in the affected areas even 20 years after the catastrophe and are ubiquitary spread in every compartement of the biosphere. Food is contaminated more or less according to type and origin. The gamma nuclide ^{137}Cs can be analysed easily and therefore is investigated intensively. Biological half-life and behaviour of cesium are well documented for every compartement and man (whole body countings). ^{90}Sr is a pure β-emitter and can be analysed only after chemical extraction procedures. As a consequence ^{90}Sr is analysed only rarely.

This lack of data leads to the general uncertainty of the exposed population and even of scientists. Strontium is much more mobile than cesium and shows a quite different fate in the biosphere. Analyses of primary teeth can illustrate the incorporated activity of ^{90}Sr in a more differentiated way. Particulary during the organogenesis of the foetus after 6 months up to 6 months after birth the child incorporates ^{90}Sr. Once incorporated the strontium remains in the body and is excreted only slowly. The biological half-life of ^{90}Sr is 11 years.

The intake of ^{90}Sr of the exposed population could be estimated with the help of suppositions on the basis of analytical data from food and environmental samples. In this report first results of a project of many years duration of ^{90}Sr analyses of primary teeth of children from the most affected regions around Chernobyl are presented. These original results are compared with analyses of teeth from Swiss children. A further outlook will be presented at the end.

Materials and methods

Samples

During the last 15 years many children from the surrounding area of Chernobyl received free sanitation of their teeth in Switzerland. Primary teeth from 64 children were collected during these dental treatments. The teeth were preserved with formaldehyde and sent to the lab. The children were born between 1977 and 1991 in villages in the region of Chernobyl (i.e. Luginy, at a distance of 80 km from Chernobyl). Their primary teeth were collected during the years 1991 to 1998.

Fig. 1 *Primary tooth samples from children of Luginy*

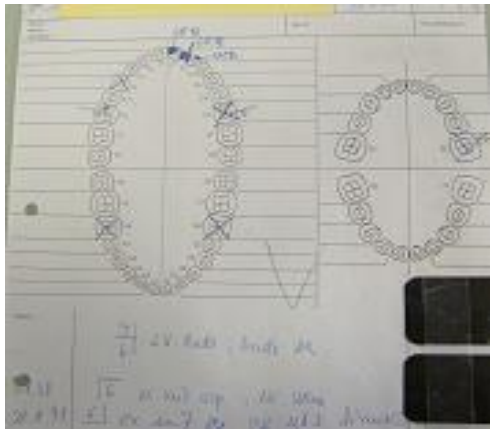


Fig. 2 *A protocol from a dental treatment*

Radioactivity of ^{90}Sr

The activity in radioactivity is defined as:

Activity $A = \frac{\ln 2 * m}{T_{1/2} * A_r}$

Whereas the specific activity is defined as the quotient of activity and mass:

$A_{spec} = \frac{\ln 2}{T_{1/2} * A_r}$ Ar: relative atomic mass (g), $T_{1/2}$: half live (s), m: mass weight of the pure carrier-free nuclide

Nuclides with short half lives show high specific acitivities but their masses are infinitely small. ^{90}Sr with a half live of 28.5 years has a mass of 1.94 µg per 10 MBq rsp. 1,94 pg/Bq. ^{90}Sr in the low Bq level corresponds to analytical concentrations in the pg/L range and therefore are difficult to detect with ICP-MS. Due to such very low concentration levels it is necessary to work with carrier nuclides (inactive isotope of the same element) to avoid substantial losses of nuclides by adsorption onto glass walls.

^{90}Sr decays to ^{90}Y which has a short half-life of 65 hours to produce the stable ^{90}Zr. Already after 19 days (seven half-life periods of the radiocuclide ^{90}Y) the secular equilibrium between ^{90}Y and its daughter nuclide ^{90}Y is reached.

Principle of the method

In biological systems the element strontium behaves quite similarly to calcium. The concentration of Ca in most of the biological tissues (especially bones and teeth) is remarkably constant. The ratio of Sr / Ca in the tissues is admittedly representative for this ratio in the diet. So the activity of ^{90}Sr is usually reported in relation to the ca-content (Bq ^{90}Sr / g Ca). Teeth are the only calcified biological tissue which can be ethically obtained because the primary teeth are expelled spontaneously or have to be extracted for health reasons.

The collected teeth are mineralised and both nuclides ^{90}Sr and ^{90}Y are extracted from the ashes by precipitation as oxalate salts. The nuclide ^{90}Y is then separated from ^{90}Sr by adding inactive ^{88}Y as a carrier and precipitation of the total yttrium as yttrium hydroxide. After redissolving and precipitation as yttrium oxalate the β-decay of the ^{90}Y is immediately measured with a gas proportional counting system. The ^{90}Sr activity of the precipitate is recalculated to the time of the last precipitation via decay curve of the ^{90}Y source. The original ^{90}Sr of the sample is recalculated considering the overall chemical recovery of the several precipitation steps,

counting efficiency of the gas proportional counter and calculated the activity back to the birth date.
Ca was determined with inductively coupled plasma mass spectrometry (ICP-MS).

Sample preparation
The teeth were washed carefully with distilled water and air dried at ambient temperature over night.

Mineralisation of the tooth samples
At least 0.5 g of tooth was necessary for the determination of the ^{90}Sr activity. In some cases teeth from children of the same year of birth had to be pooled to get enough material for the analyses. The material was weighed into a porcelain crucible and ashed for 20 hours at 600 °C. Remaining particles were pounded and ashed again for 3 hours at 600 °C. The final greyish-white ashes were dissolved in 20 ml hydrochloric acid (33% w/w) and the solution was filtered into a volumetric flask using a paper filter and then diluted with distilled water up to 50 ml. 10 ml of the sample solutions were used for the ICP-MS analyses.

First oxalate precipitation
5 ml of a ^{89}Y carrier solution (9 g/l yttrium chloride) were added to 40 mL of this solution and heated to boiling. 6 g of oxalic acid were added carefully to the solution. The oxalates of calcium, strontium and yttrium were precipitated by adding 150 ml of a solution of ammonium acetate (100 g/l) until a pH of 5 was reached. After adding further 15 ml of ammonium acetate the solution was cooled to room temperature.

Yttrium hydroxide precipitation
The precipitate was filtrated by means of a glass sinter filter and washed with ammonium oxalate solution (1g/l w/w). The filter and the precipitate were dried using an infrared lamp and then mineralised for 4 h at 600 °C in a muffle furnace. The ashes was dissolved in 10 ml hydrochloric acid solution (20 % w/w) and diluted with water to 100 ml. 5 ml of a ^{88}Sr carrier solution (50 g/l strontium chloride) and 4 drops of methyl orange indicator solution were added to the solution and then heated to boiling. Ammonium hydroxide solution (25 % w/w) was added until the colour changed to yellow (pH>5). A further ml of ammonium hydroxide solution was added and the solution boiled for 10 minutes. The precipitation was then filtrated through a glass sinter filter and washed with diluted ammonium hydroxide solution. Date and time of the precipitation was noted. At this moment the ^{90}Y activity corresponds to the ^{90}Sr activity in equilibrium and the decay of the isolated ^{90}Y starts.

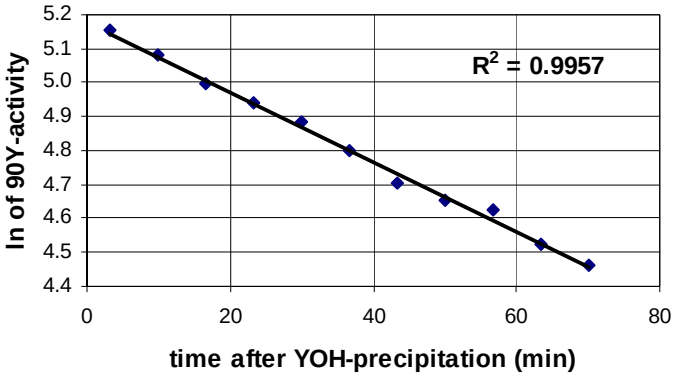
Yttrium oxalate precipitation
Then followed a second precipitation of the yttrium with oxalic acid to obtain a pure yttrium source. The precipitate was resolved by washing the filter with three portions of 3 ml of hydrochloric acid (20 % w/w) into a 200 ml Erlenmeyer flask. The filter was washed with distilled water and 5 drops of methyl orange indicator solution were added. Ammonium hydroxide solution (10 % w/w) was added to the solution until the colour changed to yellow (pH>5). By adding hydrochloric acid (20 % w/w) dropwise the pH of the solution was lowered to a pH of 1 to 2 (colour changed to red). The solution was heated to boiling and 8 ml of oxalic acid (20 g/l) were added. The precipitate was heated for a further 10 minutes and then cooled to room temperature. The precipitation was filtered off by use of a Millipore filter and washed with water, ethanol and diethyl ether. The yttrium oxalate was dried for 15 minutes at 105 °C and weighed. The weighing allowed the calculation of the overall chemical recovery of all precipitation steps. The YOH precipitation was finally fixed with collodium solution onto a metal disc for the β-analyses.

β-spectrometry
The β-analyses were started immediately after the preparation of the yttrium source. 4 samples were measured in parallel for at least 7 consecutive cycles of 400 minutes each. The original ^{90}Sr activity was recalculated by extrapolating graphically the β-activity to the final time of the

last precipitation and by taking into account detector efficiency and overall chemical recovery. Recoveries of the precipitation procedures were from 70 to 90 %. Teeth from children of Basel were spiked with ^{90}Sr and analysed. Total recovery was 103 % after correction of the chemical recovery. The results were expressed as quotient of ^{90}Sr in Bq/g Ca.

The β-counter used was a very low background multiple detector for low alpha/beta activities type MINI 20/41 from eurysis measures, Saint Quentin, Yvelines, France. 4 gas flow proportional counters permitted 4 simultaneous alpha and beta measurements. α/β amplitude discrimination of the pulses was applied to distinguish between α- and β-decays. The efficiency of the β-counter was 25%. The background counts were 0.22 counts per minute (cpm). With a counting time of 400 min the minimal detection limit was 0.04 cpm. The detection limit of the method was then about 0.08 cpm rsp. 15 mBq ^{90}Sr/g tooth or 45 mBq ^{90}Sr/g Ca. Total measurement uncertainty was about 30 % whereas the counting uncertainty was about 12 % at an activity level of 30 mBq/g Ca.

Fig. 3
Measured β-activity of a tooth sample. The reconstructed half-life of ^{90}Y (slope of the curve) was 67.2 hours and slightly higher than the theoretical value of 64.5 hours.



ICPMS analyses

10 mL of the hydrochloric acid solution of the mineralised tooth samples were analysed with ICP-MS at the Cantonal Laboratory Basel-Country. The analyses consisted in the determination of the non radioactive nuclides ^{40}Ca, ^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr. The ICP-MS system was from Perkin Elmer model Elan 6100. Total uncertainty for each nuclide was about 20 %.

Results

The ^{90}Sr activities were calculated back to the date of birth. These 64 results were expressed as Bq ^{90}Sr/g Ca.

Year of birth	Number of samples	Mean value (Bq ^{90}Sr/g Ca)	Standard deviation
1977	1	0.116	
1978	1	0.085	
1979	1	0.180	
1981	1	0.290	
1982	1	0.154	
1983	8	0.213	0.066
1984	10	0.193	0.061
1985	7	0.217	0.146
1986	5	0.229	0.015
1987	6	0.235	0.182
1988	18	0.232	0.009
1989	2	0.109	0.025
1991	2	1.919	0.757
1992	1	0.827	

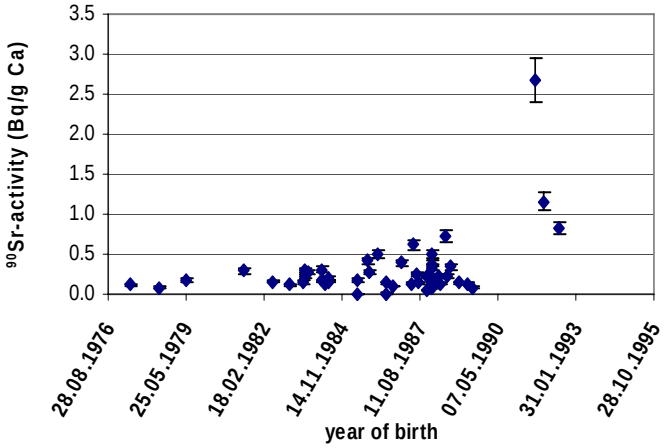


Fig. 4
Development of the ^{90}Sr activity in primary teeth of children from Luginy. The values are given as average of the year of birth and are calculated back to the year of birth.

Comparison with results from Swiss primary teeth

The following figure shows the ^{90}Sr activities in teeth of children from Chernobyl compared with results from Swiss children. The Swiss data are yearly published by the Swiss Federal Office of Public Health[1]. Primary teeth from Swiss children of the cantons of Waadt and Zurich show activities of 0.02 - 0.03 Bq/g Ca. The Swiss date from 1953 to 2002 show clearly a maximum in the 1960 years resulting from the fallout of atomic bomb tests. Activities in teeth of Swiss children reached 0.4 Bq/g Ca whereas the Chernobyl fallout caused a maximum in Switzerland in 1986 of only 0.06 Bq/g Ca.

The investigated Chernobyl teeth show about the ten fold of the Swiss values. An increase of the activity due to the accident at Chernobyl could not be observed.

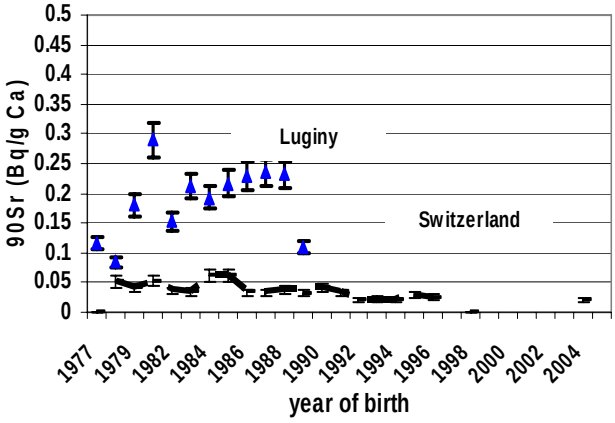


Fig. 5
Comparison of ^{90}Sr activity in primary teeth of children from Luginy and children from Switzerland. All values are mean values and calculated to the year of birth. Two high values in 1991 and 1992 are not shown (see figure 4).

Discussion

It is quite surprising that the activity of teeth from children in the region of Chernobyl born before 1986 is as high as the activity of the teeth of children born just after the catastrophe. Also if the environment had been continuously contaminated since long time, the input from the accident should produce a measurable augmentation of ^{90}Sr activity. At the moment we think that ^{90}Sr from the contaminated food is more effectively adsorbed by young people, even they are older than 6 years, than admitted so far.

During the sessions with the dentists, parents and children were asked about their eating habits. Half of the children were eating berries from the own garden and / or mushrooms collected in the woods. As the following figure shows there could not be found any correlation between ^{90}Sr activity in the teeth and the eating habits. Children not eating own-grown vegetables, berries or collected mushrooms showed even higher activities.

[1] Froidevaux P. et. al.: Mesures de ^{90}Sr dans les vertèbres et les dents de lait in: Environmental radioactivity and radiation exposure in Switzerland. Swiss Federal Office of Public Health, (2005 and further).

Similar high ^{90}Sr activities were found in baby teeth of children living in St. Louis and New York Metropolitan Area from 1957 - 1970[2]. The authors suggest the emissions from local nuclear power reactors as the main source. The observed large variations of the activities can be explained by differences in dietary intake of pregnant women. Temporal variations in reactor emissions from month to month will affect foetal concentrations of radioactivity.
Without a better understanding of the measured values it is not possible to calculate a risk assessment of the ^{90}Sr activity.

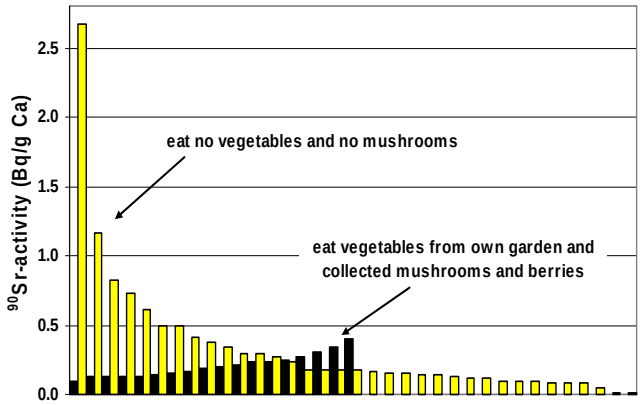


Fig. 6
Distribution of ^{90}Sr activities in primary teeth as a function of the eating habits of children from Luginy.

Perspectives
More investigations and discussions are needed to explain the present data. It seems very important to proceed with further measurements of ^{90}Sr activity in teeth of the population in the region of Chernobyl to follow the situation and to understand better the origin of the observed relatively high activity level existing there. The population has the right to know how much radioactive strontium they incorporate and has to be informed also about these aspects of the tragedy.

Acknowledgements
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[2] Jay M. Gould et. al.: Strontium-90 in baby teeth as a factor in early childhood cancer. Int. J. Health Services 30, 515-539 (2000).