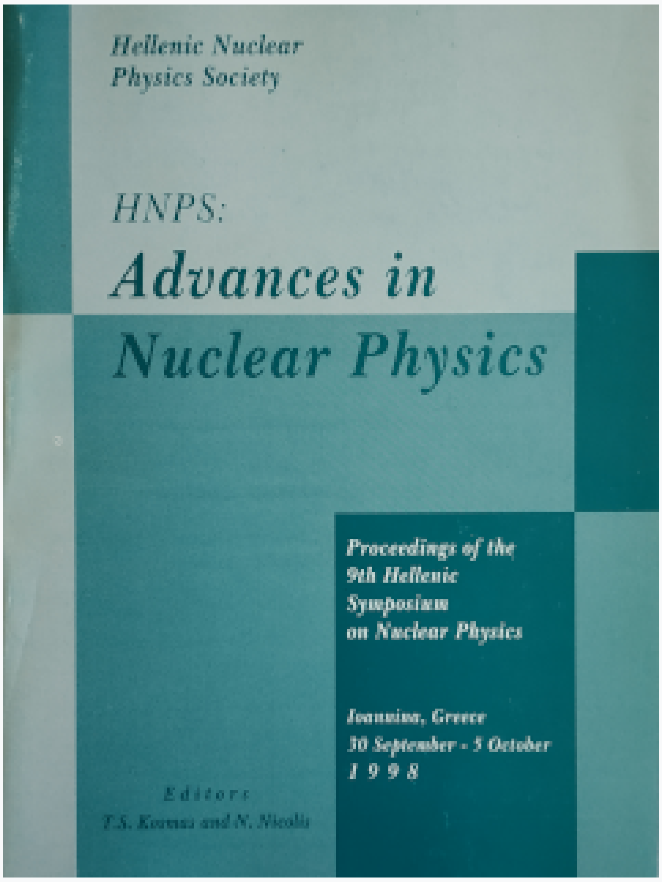
HNPS Advances in Nuclear Physics

Vol 9 (1998)

HNPS1998



Strontium-90 in human bones and teeth in Greece: Measurements and predictions using an age-dependent model

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doi: [10.12681/hnps.2796](https://doi.org/10.12681/hnps.2796)

Strontium-90 in human bones and teeth in Greece: Measurements and predictions using an age-dependent model

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Abstract

Strontium-90 concentration in human bones and teeth, collected in Greece during the period 1992-1996, was measured. One hundred and five bone samples, mainly cancelous (spongy) bone, and 108 samples, involving a total of 896 individual teeth, were processed. Samples were classified according to age and sex of the donors.

Radiostrontium concentration in bone samples showed small variations with regard to age or sex, yielding an average value of 30 mBq ^{90}Sr / g Ca. However, ^{90}Sr concentration measurements in teeth evinced a pronounced structure, which clearly reflects contamination from the 1960s atmospheric nuclear weapons tests and the more recent Chernobyl accident. This difference is attributed to the different bone texture of skeletal bones and teeth, the later consisting mainly of compact bone.

An age-dependent model for radiostrontium concentration in human bones and teeth was developed, which was able to successfully reproduce the experimental data. Through a fitting process, the model also yielded calcium turnover rates for compact bone as a function of age, as well as an estimate of radiostrontium contamination of foodstaffs in Greece for the past four decades.

1 Introduction

Strontium-90 is an artificial radioactive isotope, produced by nuclear fission in explosions of nuclear devices and in nuclear energy plants. A detailed table of nuclear explosions for the period 1945-1994, including date, site and country responsible for the explosion is given by Lawson [18]. During this period 881 nuclear weapons' tests were performed above ground and in the atmosphere and 878 tests underground. From tests during the first half of this period (1945 - 1965) it is estimated that about 10^{18} Bq of ^{90}Sr and 1.3×10^{20} Bq of ^{89}Sr were released in the atmosphere ([8],[16]). Serious nuclear accidents also added radioactive strontium isotopes into he atmosphere. For example, the nuclear accident at Chernobyl released about 10^{16} Bq of ^{90}Sr and 10^{18} Bq of ^{89}Sr. These releases, together with fallout from other fission products, soon spread world-wide.

Strontium is an element with chemical behavior similar to that of calcium. As the human skeleton consists of about 40 % calcium, ^{90}Sr is a bone-seeking nuclide. Radiostrontium enters into the food chain and finally into the human body, where it is deposited to bone and teeth. The longest-lived of these radioactive isotopes, ^{90}Sr ($T_{1/2}$ = 28.0 y), disintegrates by electron emission to ^{90}Y, also a radioactive nucleus, which further beta-decays to the stable nucleus ^{90}Zr with a half-life $T_{1/2}$ = 64.1 h. The two successive electrons deposit their energy within a very small volume in the vicinity of the disintegrating nuclei. This energy is absorbed by bone or teeth tissue and the biological effect is related to the total energy released and the rate at which the energy is absorbed in living cells. The damage to the cells may be irreversible leading to leukemia and bone neoplasm. Thus, knowledge of radiostrontium concentration in bone tissue is necessary for estimating risk to public health.

Many investigations with regard to ^{90}Sr concentration levels in bone ([3,5,8–10,12,13,26,27,30,32]) and teeth ([1,2,7,10,14,15,20,23–25,27–29,31,35]) have been conducted since the start of nuclear weapons tests in the 1950s.

There is only one report concerning concentration levels of ^{90}Sr in bones for the Greek population [22], covering the period 1962-1967. A need therefore was felt for assessing current radiostrontium levels in human bones in Greece, especially after the Chernobyl accident.

2 Materials and methods

2.1 Sampling

One hundred and five human bone samples were collected during the period 1993-1995. Bone samples were obtained from the Athens State Morgue and the University of Ioannina General Hospital. Although all donors of the samples were residents of Greece, the geographical location of residence of the donors within the country varied significantly. Eighty-one samples were from the greater Athens area, 19 samples from the district of Epirus (about 300 km North-West of Athens) with the rest of the samples from various other regions of Greece. However, since Greece is a small country, the geographical area from which all samples were obtained did not exceed 400 km in diameter. The texture of bone also varied but samples generally consisted of cancelous (spongy) bone. Anatomically, the samples collected involved 35 sternum, 34 rib, 24 sternum-rib, 18 femur epiphysis, 6 knee, 1 clavicle and 1 vertebrae sample.

One hundred and eight teeth samples were collected by dentists in Athens, the city of Ioannina and the town of Metsovo, as well as at the dental clinics of the University of Athens Hospital and the 'Hatzikosta' General Hospital at Ioannina. Teeth samples were grouped according to age and sex. Each sample contained typically 8-10 teeth. Thus, a total of 896 teeth were processed and measured involving, 225 molars, 203 premolars, 130 incisors, 45 canines and 246 roots of various teeth.

2.2 Preparation of bone and teeth samples

Most of the bone samples were stored in formalin for more than one year. Sternum and rib samples were cleaned from remains of adhering soft tissue and dried at 80-100 C to constant weight in order to remove formaldehyde from the sample. Femur bone samples, which were free of any soft tissue, were stored at -20 C. Typical dried mass of bone samples was 30 g.

Teeth samples were stored in plastic bottles at room temperature. Typical dried mass for teeth samples was 10 g. All bone and teeth samples, after initial cleaning, were weighted and ashed in muffle at 500-600 C for about one day. The ash was ground and blended to produce an homogenized powder. Some samples were grouped in order to obtain larger samples with enough ash quantity for ^{90}Sr analysis.

2.3 Chemical treatment of the samples

For the purposes of the research reported here a new method of chemical treatment of samples prior to ^{90}Sr measurement was developed at the Nuclear Physics Laboratory (NPL) of the University of Ioannina. The chemical method is based on the preferential chelation of ^{90}Y, the daughter nucleus of ^{90}Sr disintegration. The chelating agent is an organic solution of Bis (2 - ethyl - hexyl) hydrogen phosphate (BEHHP) diluted in dodecane. This is similar to a chemical separation method used by the Los Alamos National Laboratory [17], which also takes into account ^{89}Sr, often present in the biological matrix.

The chemical method developed at the NPL, involves the chemical extraction of all ^{90}Y which at some recorded time T_1 is at equilibrium with ^{90}Sr in the original sample. The extracted ^{90}Y is deposited on a paper filter which is then subjected to activity measurements.

2.4 Calculation of ^{90}Sr concentration

Yttrium-90 concentration of the sample was determined from the disintegration rate of the filter, while ^{90}Sr concentration was calculated (in mBq / g Ca) on the assumption that ^{90}Y and ^{90}Sr are in equilibrium at time T_1 of the separation of the two isotopes and that calcium percentage of the bone or tooth ash is 40 % ([6], [10]).

All samples used in this study were measured in a Camberra 2404 alpha-, beta-spectrometer using a gas flow proportional counter (gas P10: Ar 90 % - CH_4 10%) for at least three times during a period of 7 days. These measurements were necessary in order to follow ^{90}Y disintegration and assess whether ^{90}Sr impurities were present in the sample. The duration of each measurement was typically 60 min. The detector background was determined from periodic measurements of a blank sample and was found with a mean value of 0.69 $\pm$ 0.10 cpm (counts per minute).

Successive measurements (three or more) of each sample were fitted to the function

$$Y = Y_0 \cdot \exp\{-\lambda(T - T_1)\} \tag{1}$$

in which Y_0 = Yttrium disintegration rate (at time T_1) of ^{90}Sr removal from the sample (in cpm), Y = Net Yttrium (after subtraction of the background) disintegration rate at time T, which is the mid time for the time interval of the measurement (in cpm) and λ = a constant equal to $\ln 2 / T_{1/2,Y}$ where $T_{1/2,Y}$ = 64.1 h is the half-life of ^{90}Y (in s).

Yttrium activity A_Y, of the filter paper source was calculated using the expression

$$A_Y = \frac{Y_o}{C_Y \cdot E_Y} \tag{2}$$

in which E_Y = ^{90}Y efficiency of the counter, which was estimated as 0.26 and C_Y = Yttrium yield of the sample chemical processing. Strontium-90 concentration of the sample is expressed in mBq / g Ca of the sample and is calculated using the expression

$$A_S = \frac{1000 \cdot A_Y}{0.4 \cdot 60 \cdot M} \tag{3}$$

where A_S = ^{90}Sr activity of the sample, expressed (in mBq / g Ca), M = mass of the ashed sample (in g) and 0.4 = constant for the calcium percentage in the ashed sample (40 %).

3 Experimental results

3.1 Bone samples

Strontium-90 concentration in bone samples did not show statistically significant variations with regard to sex, age and residence of donor. Mean value of ^{90}Sr concentration was found 30 $\pm$ 13 mBq ^{90}Sr / g Ca. Fig. 1 shows ^{90}Sr concentration measured in bones according to age. The number in each column denotes the number of samples analyzed per age group, while error bars represent one standard deviation (see also Table 1).

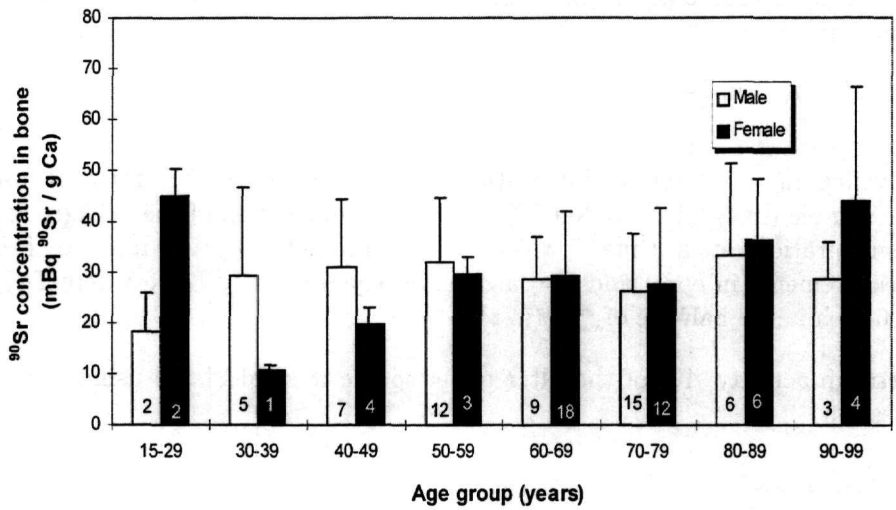

Fig. 1. ^{90}Sr concentration in bone samples according to age and sex. Each column represents the mean value of ^{90}Sr concentration for the samples in the group; error bars represent one standard deviation. Numbers in each column give the number of samples analyzed per group.

Statistically significant (t test confidence level 0.01) differences were found in ^{90}Sr concentration with regard to the kind of bone sample measured. Patella bone showed much lower values of 16 $\pm$ 7 mBq ^{90}Sr / g Ca in contrast with rib, sternum and femur epiphysis samples, where activity was almost double with a mean value of about 31 mBq ^{90}Sr / g Ca (see also Table 2).

Table 1
Radiostrontium concentration in male and female bone samples Mean values of each group with the respective one standard deviation (error bars) and number of samples analyzed per group, are presented. Results of t test for mean values between two sexes are also presented. "No" means that there is no statistically significant difference between mean values of ^{90}Sr concentration in bone samples from both sexes. (confidence level 0.01).

	Radiostrontium concentration in bone samples							
	Male		**Female**		**Mean values test**			
						Confidence level 0.01		
Age	*mBq $^{90}Sr/g$ Ca (m.v. ±ó)*	*I*	*mBq $^{90}Sr/g$ Ca (m.v. ±ó)*	*I*	*t test*	*Critical value*	*Degrees of freedom*	*Result*
15-29	18±8	2	45±5	2	−2.92	9.92	2	No
30-39	29±17	5	11±1	1	0.88	4.6	4	No
40-49	31±13	7	20±3	4	1.49	3.25	9	No
50-59	32±13	12	30±3	3	0.31	3.01	13	No
60-69	29±8	9	29±12	18	−0.18	2.79	25	No
70-79	26±11	15	28±15	12	−0.25	2.79	25	No
80-89	33±18	6	36±12	6	−0.31	3.17	10	No
90-99	29±7	3	44±23	4	−0.94	4.03	5	No

Table 2
Radiostrontium concentration in bone samples according to kind of bone. Mean values with the respective standard error and number of samples analyzed per group are presented.

	^{90}Sr in bone according to kind	
Kind of bone	**^{90}Sr concentration** *mBq $^{90}Sr/g$ Ca (m.v. ±ó)*	**Number of samples** *I*
Knee	16±7	7
Rib	29±10	35
Sternum	33±16	40
Sternum and rib	31±11	11
Femur head	30±12	17

3.2 Teeth samples

In contrast to the results from bone samples, ^{90}Sr concentration in teeth evinced an interesting variation with regard to age. Fig. 2 shows radiostrontium concentration activity of teeth for the various age groups up to 80 years. The numbers in the columns denote the number of samples analyzed per group, while error bars represent one standard deviation (see also Table 3). The data in this figure show an increase of ^{90}Sr concentration in teeth from 24 ± 4 mBq ^{90}Sr / g Ca at birth to 51 ± 6 mBq ^{90}Sr / g Ca for the age of 20 years. For ages 21 to 40 years old ^{90}Sr concentration increases further from 19 ± 8 mBq ^{90}Sr / g Ca to 64 ± 21 mBq ^{90}Sr / g Ca; thereafter it decreases steadily to the value of 4 ± 1 mBq ^{90}Sr / g Ca for the 71-80 age group. Similar but not

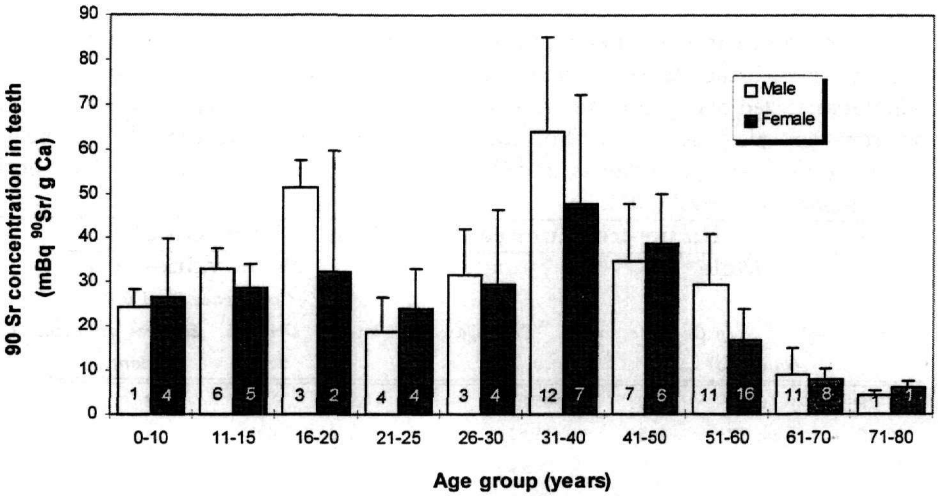

Fig. 2. ^{90}Sr concentration in teeth samples according to age and sex. Each column represents the mean value of ^{90}Sr concentration for the samples in the group; error bars represent one standard deviation. Numbers in each column give the number of samples analyzed per group.

Table 3
Mean values of ^{90}Sr concentration in teeth samples for each age group for both sexes. One standard error and the number of samples analyzed per group are also presented. The results of t test for the mean values between sexes for the same age group are presented in the last column. "No" means that there are no statistically significant differences between the mean values of the groups and "Yes" means that there are differences (confidence level 0.01)

Age	Male		Female		
	Mean value ±ó mBq ^{90}Sr/g Ca	*I*	*Mean value ±ó mBq ^{90}Sr/g Ca*	*I*	*t test results*
0-10	24±4	1	26±13	4	No
11-15	33±5	6	29±5	5	No
16-20	51±6	3	32±27	2	No
21-25	19±8	4	24±9	4	No
26-30	32±10	3	29±17	4	No
31-40	64±21	12	48±24	7	No
41-50	35±13	7	39±11	6	No
51-60	30±12	11	17±7	16	Yes
61-70	9±6	11	8±2	8	No
71-80	4±1	1	6±1	1	No

so sharp variations are noticed in female teeth. There is an increase from 26 ± 13 mBq ^{90}Sr / g Ca to 32 ± 27 mBq ^{90}Sr / g Ca for ages up to 20 years old and a further increase from 24 ± 9 mBq ^{90}Sr / g Ca to 48 ± 24 mBq ^{90}Sr / g Ca up to 40 years old. After that age, ^{90}Sr concentration decreases steadily to the value of 6 ± 1 mBq ^{90}Sr / g Ca for the 71-80 age group. Statistically significant differences are observed between different age groups of the same sex but not between the same groups of different sexes (see also Table 3). There are no statistically significant differences between different kinds of teeth for the same age group (male and female).

4 Modeling of ^{90}Sr concentration in human bones

The rate of uptake and removal of a radionuclide by the body can vary significantly with age. This is well documented in the case of ^{90}Sr, which has been measured in a large number of human skeletons. Thus, the large quantity of data collected for ^{90}Sr concentration in bones has been used in the development of several age-dependent models. Leggett [19] have proposed a metabolic model that applies from birth through adulthood. This model considers compartments which correspond to physical processes or subsections of the skeleton and examines the behavior of ^{90}Sr in terms of the behavior of calcium. Retaining certain features of this model, a new age-dependent model was developed at the NPL.

4.1 Skeletal Compartments

Bone is often divided into two categories, *structural bone* which refers to the mechanical function of the skeleton and *metabolic bone* which refers to the function of the skeleton in the regulation of extracellular calcium levels. Based on this two categories, skeleton may be viewed as consisting of 3 compartments, with 2 compartments associated with structural bone (cancelous and compact bone) and the third associated with metabolic bone (bone surface). Cancelous and compact bones are differentiated in terms of their surface to volume ratio. This distinction represents an average for the entire skeleton and bones can be defined in average as cancelous or compact.

4.2 ^{90}Sr kinetics in the skeletal compartments

For an individual born at time T (calendar year), the amount $Q(t)$ of ^{90}Sr in a skeletal compartment at age t is described by the differential equation

$$\frac{dQ(t)}{dt} = A(t)B(t)M(T+t) - L(t)X(t) - \lambda Q(t) \tag{1}$$

in which
$A(t)$ is the annual amount of calcium intake as a function of age t (g Ca y^{-1}),

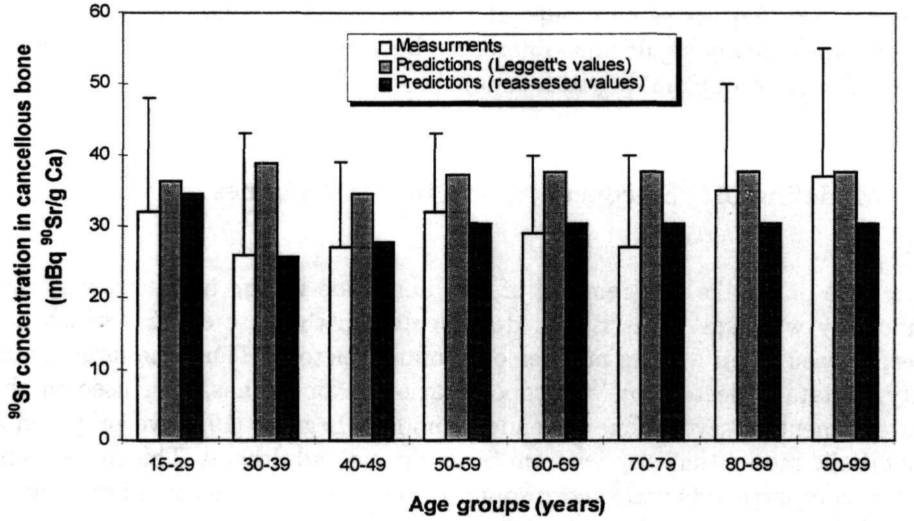

Fig. 1. Mean age-group values of ^{90}Sr concentration in bone samples and predictions using the NPL model with Leggett's values (grey bars) and reassessed values (black bars). Reassessed values of the parameters of response functions (cancelous bone turnover rates and ^{90}Sr concentration in food in Greece) were calculated by fitting the NPL model to measurements.

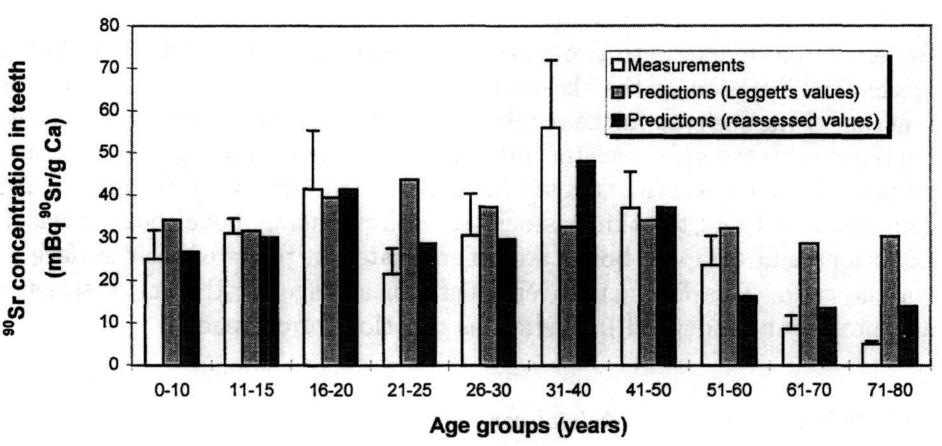

Fig. 2. Mean age-group values of ^{90}Sr concentration in bone samples and predictions using the NPL model with Leggett's values (grey bars) and reassessed values (black bars). Reassessed values of the parameters of response functions (cancelous bone turnover rates and ^{90}Sr concentration in food in Greece) were calculated by fitting the NPL model to measurements.

$B(t)$ is the percentage of ^{90}Sr contained in food which is uptaken by the skeletal compartment as a function of age t,
$M(T+t)$ is the mean ^{90}Sr concentration in food during calendar year $t+T$ (Bq / g Ca),

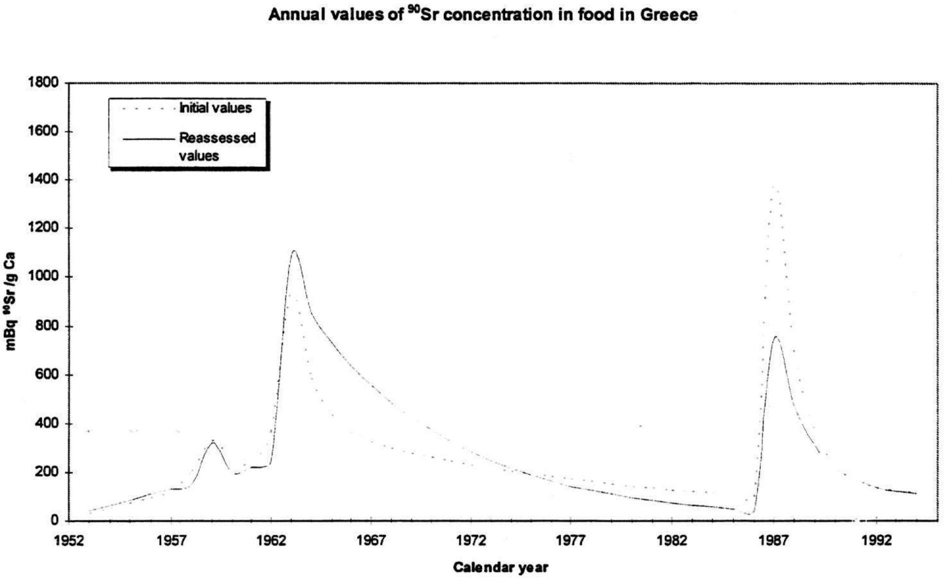

Fig. 3. Mean annual values of ^{90}Sr concentration in food in Greece for the period 1953-1994 (dashed line; K. Stamoulis, 1998) and assessed values derived by fitting model predictions to measurements of bone and teeth samples (solid line).

$L(t)$ is the annual bone turnover rate which is assumed to be equal to the annual removal rate of ^{90}Sr from the bones (kg y^{-1}),
$X(t) = Q(t)/V(t)$ is the concentration of ^{90}Sr (Bq / kg) in the skeletal compartment of volume $V(t)$ and
λ is the radiological decay constant of ^{90}Sr (y^{-1}).

The integration of eqn. (4) requires knowledge of the functions $A(t)$, $B(t)$, $M(T)$, $L(t)$ and $V(t)$. With the exception of the level of ^{90}Sr contamination in food $M(T)$ which is a function of calendar year T and depends on geographical location, the rest of the functions are derived from physiological considerations and data. Information on these functions and analytic expressions in the form of semi-empirical formulas are given elsewhere [34].

There is scant data concerning radiostrontium contamination of foodstaffs during the past forty years in Greece. Some information is provided by measurements for milk during the periods 1962-1971 and 1986-1994 and for total β radiation fallout in Greece during the period 1961-1983, performed by the Greek Atomic Energy Commission. In reducing these data, the value 0.24% proposed by Bakacs-Polgar [4], was adopted as the percentage of ^{90}Sr in the total β fallout. The ratio of ^{90}Sr in all foodstaffs to that in milk was taken as 1.5 [19]. The function $M(T)$ obtained in this fashion, albeit with considerable uncertainty, is plotted in Fig. 5 (dashed line). The two large excursions in this function correspond to the period of intensive atmospheric nuclear weapons testing (1963-1965) and the Chernobyl accident (1986).

Predictions of the NPL model were compared to the measurements obtained in the study presented here. For this purpose the data for both sexes were averaged within the corresponding age groups. Parameters for cancelous bone were used in the case of bone samples and for compact bone in the case of teeth. Although a straightforward application of the model gave a satisfactory reproduction of the overall trend, as can be seen by comparing data (white bars) and model predictions using Leggett's values (grey bars) in Figs 3 and 4, it was realized that considerable improvement could be obtained by removing some of the uncertainties associated with the response functions in eqn. (4).

As already noted, the major uncertainties in the model arise from
1. Incomplete knowledge of the levels of ^{90}Sr in foodstaffs in Greece during the past four decades [function $M(T)$].
2. Incomplete knowledge of turnover rates [function $L(t)$], in particular for teeth for which, in the absence of specific data, compact bone parameters were used.
It was thus decided to allow some of the parameters of the corresponding response to vary within their range of uncertainty so as to obtain the best values through a fitting process against the data for ^{90}Sr concentration. Some of the parameters of function $L(t)$ for cancelous bone were also allowed to vary. The fitting was performed with a modified version of code MINUIT (James and Ross, 1975) with the results presented also in Figs. 3 and 4 (black bars). Predictions of the model with the adjusted response functions are seen to reproduce the data rather well.

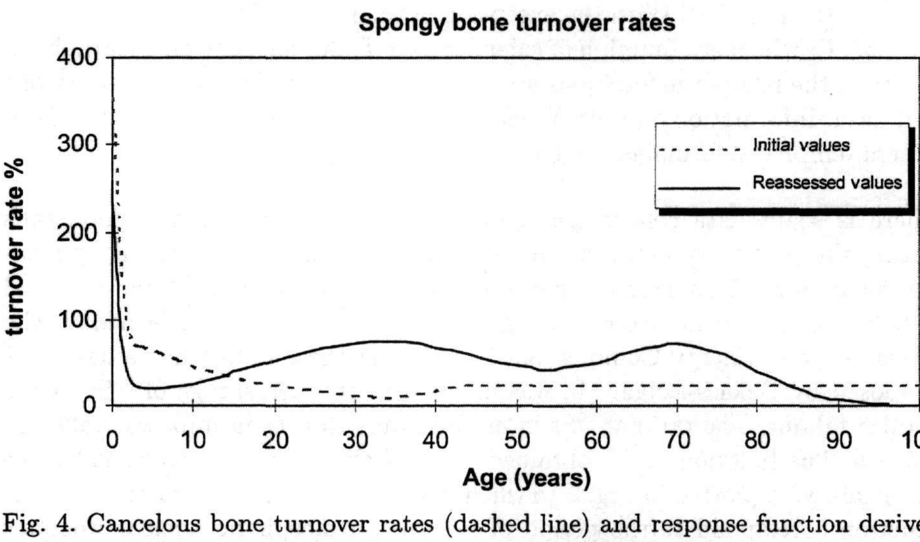

Fig. 4. Cancelous bone turnover rates (dashed line) and response function derived by fitting model predictions to measurements of bone samples.

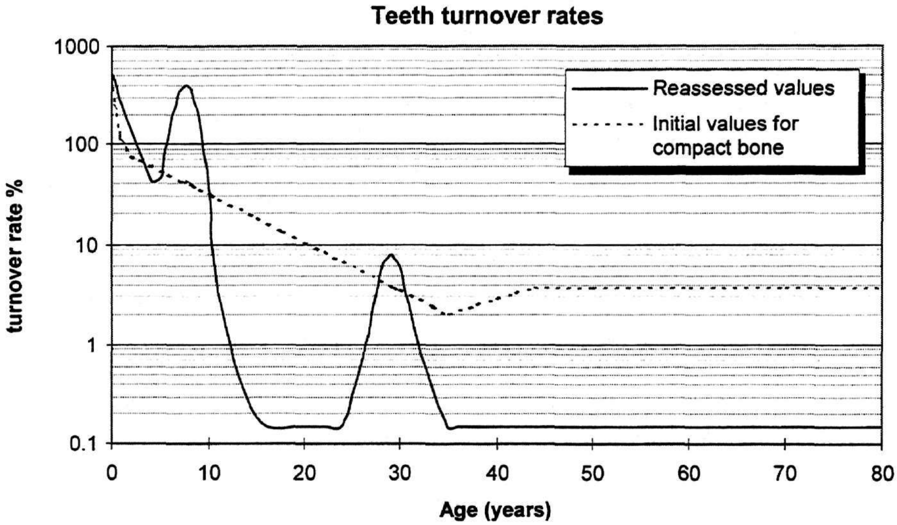

Fig. 5. Compact bone turnover rates (dashed line) and response function derived by fitting model predictions to measurements of teeth samples.

Adopting a different point of view, we may consider the adjustment of the response functions through the fitting of bone and teeth contamination data as constituting an indirect measurement of these functions. Clearly, a response function that yields a better fit to experimental data should reflect more accurately the behavior of the effect it represents. The functions $L(t)$ and $M(t)$, as determined from the fitting process, are presented in Figs. 5 - 7 with solid lines, while initial values of the functions are presented with dashed lines.

5 Discussion and conclusions

The human skeleton carries with it the history of radiostrontium environmental contamination during the period that the organism was alive. Any increase in ^{90}Sr in the environment affects within a very short time the younger age groups, which in their formative years accumulate calcium in their bones at high rates. Once acquired, the concomitant high concentration of radiostrontium, due to the slow turnover rates of calcium later in life, remains in the skeleton and as time evolves is reflected in higher age groups. The effect is more pronounced for compact bone, which after the age of 10 years has a turnover rate of less than 0.2 %, as compared to cancelous bone with a turnover rate which, although it falls rapidly after infancy, remains at a level of 10 - 20 % throughout adulthood. Thus a more pronounced record of past environmental radiostrontium contamination is expected from measurements of ^{90}Sr concentrations in teeth, which are primarily composed of compact bone.

Human bone samples from Greece measured in this research yielded an average of about 30 mBq ^{90}Sr / g Ca, with no pronounced structure as a function of age. Since the samples considered here were primarily composed of cancelous bone, it is surmised in view of the above discussion that any effects of the high ^{90}Sr contamination period of the 1960s, which should affect the 35 - 50 age groups, have been already washed out. Average levels of ^{90}Sr concentration in human bones in Greece for all ages have now been reduced to pre-1960s levels.

Contrary to the results obtained from bones, human teeth data show a very interesting structure. Two peaks, with centroids at 17.5 $\pm$ 0.4 and 38.5 $\pm$ 1.2 years are evident. The first correspond to children who were shedding their deciduous teeth at the time of Chernobyl. The second broader peak corresponds to individuals who were growing up during the period of the atmospheric nuclear weapons tests during the 1960s.

As seen in Figs. 3 and 4, the model developed at the NPL was very successful in reproducing ^{90}Sr concentration measurements in bone and teeth. In addition, through a fitting process, application of the model yielded estimates of calcium turnover rates in teeth and an estimate for radiostrontium contamination of foodstaffs in Greece for the past forty years.

The results of the research reported here clearly indicate that the effects of the atmospheric nuclear weapons tests in the 1960s exceed by far those of the Chernobyl accident. Comparing results with respective results it is apparent that the contamination from Chernobyl, which at present affects the teenage population, is considerably less than that still remaining in the middle-aged groups from the 1960s. This is corroborated by the findings in Fig. 5, which demonstrate that contamination of foodstaffs in the 1960s far exceeded that caused by the Chernobyl accident both in intensity and duration.

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