ELSEVIER

The Science of the Total Environment 229 (1999) 165–182

the Science of the Total Environment
An International Journal for Scientific Research into the Environment and its Relationship with Man

Strontium-90 concentration measurements in human bones and teeth in Greece

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Received 4 November 1998; accepted 10 February 1999

Abstract

Strontium-90 concentration was measured in human bones and teeth collected in Greece during the period 1992–1996. One hundred and five bone samples, mainly cancellous bone, and 108 samples, taken from a total of 896 individual teeth were processed. Samples were classified according to the age and sex of the donors. Samples were chemically pre-treated according to a specially devised method to enable extraction of ^{90}Y, at equilibrium with ^{90}Sr in the original sample. Subsequently, ^{90}Y β activity was measured with a gas proportional counter. Radiostrontium concentration in bone samples showed small variations with respect to age or sex, with an average value of 30 mBq ^{90}Sr/g Ca. However, ^{90}Sr concentration measurements in teeth demonstrated 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 histological structure of skeletal bones and teeth, the later consisting mainly of compact bone. An age-dependent model for radiostrontium concentration in human bones and teeth is developed which is 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 foodstuffs in Greece for the past four decades. The results obtained in this study indicate that radiostrontium environmental contamination which resulted from the atmospheric nuclear weapons tests in the 1960s, exceed by far that caused by the Chernobyl accident.

Keywords: Radiostrontium; ^{90}Sr; Human bone; Human teeth

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0048-9697/99/$ - see front matter
PII: S0048-9697(99)00052-2

1. Introduction

Strontium-90 is an artificial radioactive isotope, produced by nuclear fission during the explosion of a nuclear device 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 (1994). 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 into the atmosphere (Christensen et al., 1975; Kritidis, 1989). Serious nuclear accidents also added radioactive strontium isotopes into the atmosphere, including the accident at Chernobyl which released about 10^{16} Bq of ^{90}Sr and 10^{18} Bq of ^{89}Sr. These releases of strontium isotopes, together with fallout from other fission products, were distributed world-wide.

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

Many investigations concerning ^{90}Sr concentration levels in bone and teeth have been conducted since the start of nuclear weapons tests in the 1950s. Mean values of ^{90}Sr concentration in bones throughout the world as a function of the calendar year are presented in Fig. 1. These val-

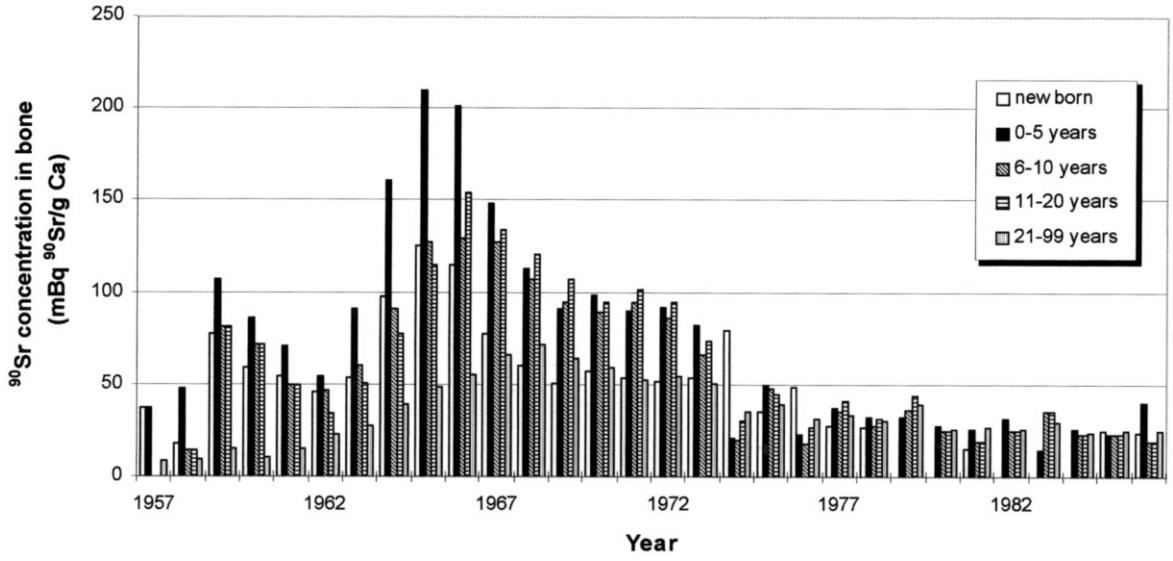

Fig. 1. World-wide mean annual values of ^{90}Sr concentration in bones grouped by age. A compilation of data from reports for the period 1957–1986 (Schulert et al., 1959; Bailey et al., 1960; Rosenthall et al., 1963, 1964; Jeanmaire and Patti, 1967, 1969, 1970, 1971, 1973, 1976, 1979; Christensen et al., 1975; Glowiak et al., 1977; Dehos and Kistner, 1980; Salonen, 1980; Klusek, 1984; Aarkrog et al., 1988).

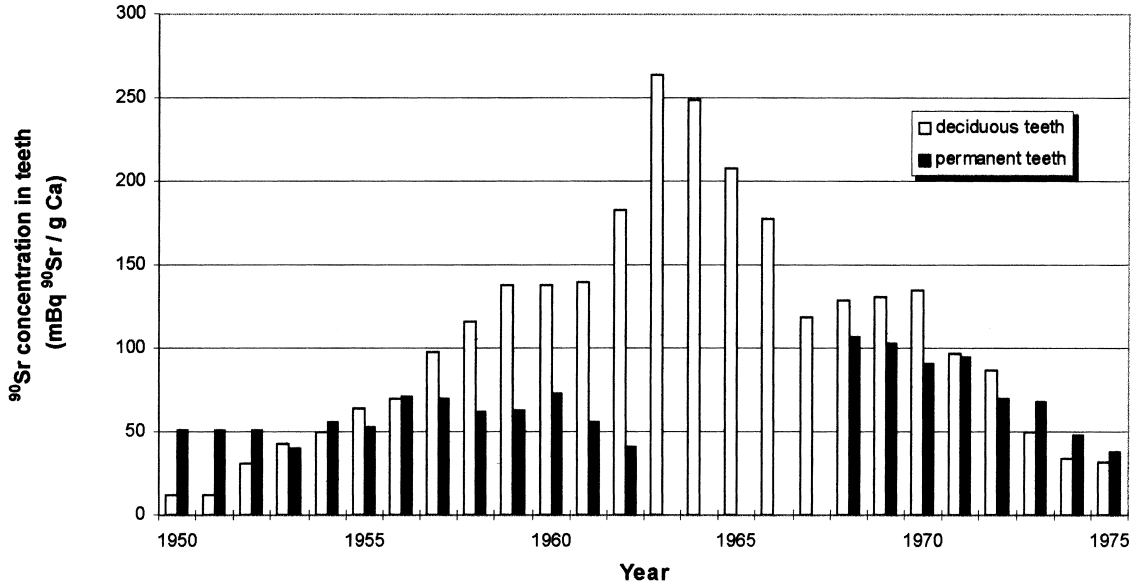

Fig. 2. World-wide mean annual values of ^{90}Sr concentration in teeth. A compilation of data from reports for the period 1950–1975 (Butler, 1961; Reiss, 1961; Rosenthall et al., 1963; Santholzer and Knaifl, 1966; Aarkrog, 1968, 1971; Lerch et al., 1969; Rytomaa, 1971, 1972; Nagai and Ishii, 1972; Prokof'yev et al., 1973; Kolehmainen and Rytomaa, 1975; Glowiak et al., 1977; Turai et al., 1988; Kulev et al., 1994).

ues have been compiled from more than 20 studies reported in the scientific literature. Similarly, mean annual values of ^{90}Sr concentration in teeth throughout the world, also compiled from a number of independent investigations, are presented in Fig. 2.

Only one report assesses levels of ^{90}Sr in bones from the Greek population (Mimikos and Duvoyiannis, 1970), covering the period 1962–1967. Considering the geographical proximity of Greece to the Ukraine, it was deemed important to assess current radiostrontium levels in human bones in Greece, especially after the Chernobyl accident.

A second reason for conducting the research presented here pertains to our earlier study on radiostrontium concentration in human teeth in Ukraine, 5 years after the Chernobyl accident (Kulev et al., 1994). An unexpected feature in the data from that investigation was observed; although the overall trend of the data was a gradual decrease of ^{90}Sr concentration with age, the 25–45 year age-groups showed abnormally high radiostrontium concentrations (see Fig. 3). The explanation offered for this anomaly at the time was that this age group (20–40 years of age at the time of the Chernobyl accident) contained a significant number of men (and women) who, either as part of the military or civilian staff of engineering firms, were mobilised immediately after the accident for the extensive clean-up operations within the 30-km zone around the damaged nuclear power plant. However, it was felt that this explanation should be tested by repeating the study for a population at a geographical position far from the site of the 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

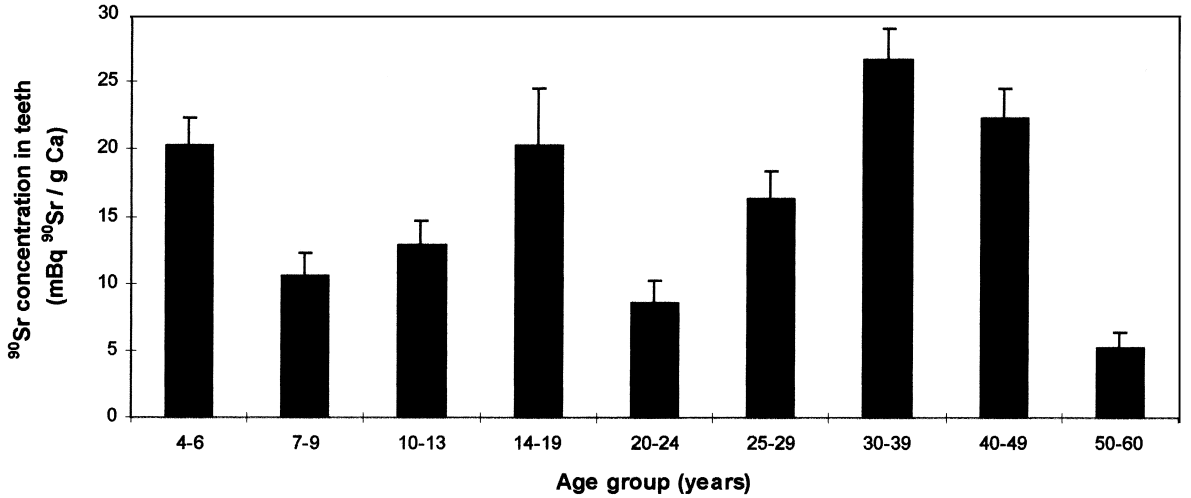

Fig. 3. Age dependence of ^{90}Sr concentration in human teeth in the male population of South Ukraine, 5 years after the Chernobyl accident (Adapted from Kulev et al., 1994).

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 a distance of 400 km. The texture of bone also varied although samples generally consisted of cancellous (spongy) bone. The samples collected included 35 sterna, 34 ribs, 24 arthroses, 18 femoral epiphyses, six knees, one clavicle and one vertebrae sample. The distribution of samples according to age and sex is shown in Fig. 4.

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

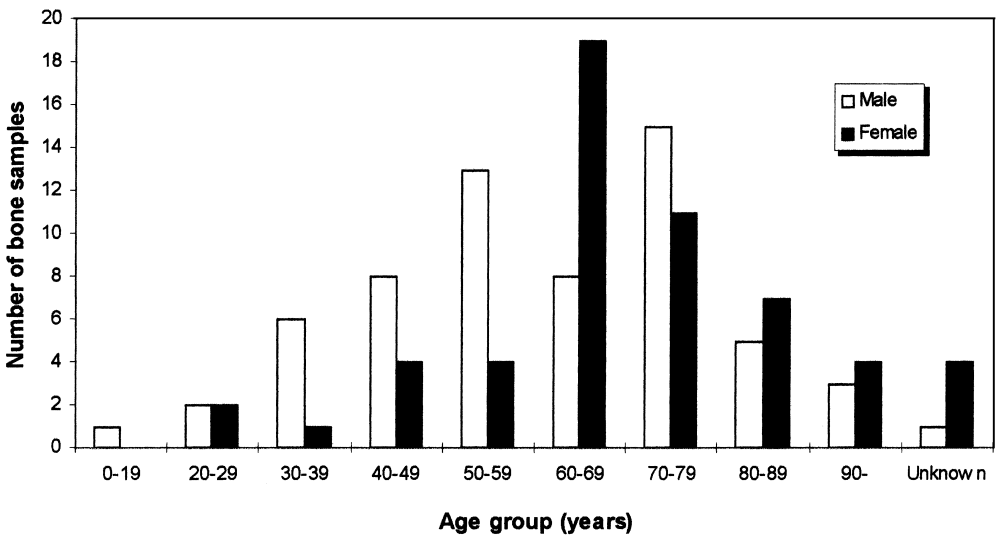

Fig. 4. Bone samples distribution according to age and sex. The last columns represent samples of unknown age.

the 'Hatzikosta' General Hospital at Ioannina. Teeth were grouped according to age and sex, with each group containing typically 8–10 teeth. A total of 896 teeth were processed and measured including, 225 molars, 203 premolars, 130 incisors, 45 canines and 246 roots of various teeth. The distribution of tooth samples according to sex and age is shown in Fig. 5.

2.2. Preparation of bone and teeth samples

Most of the bone samples were stored in a formalin solution for more than 1 year. Sternum and rib samples were stripped of remaining soft tissue and dried at 80–100°C to constant weight in order to remove formaldehyde from the sample. Femurs which were free of any soft tissue, were stored at −20°C. Typical dried mass of bone samples was 30 g.

Tooth samples were stored in plastic bottles at room temperature. A typical sample of dried tooth was about 10 g. All bone and teeth samples, after initial cleaning, were weighed and ashed in a muffle at 500–600°C for about 1 day. The ash was ground and blended to produce an homogenised 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 purpose of the study 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 (LANL, 1992). The method also measures the concentration of ^{89}Sr which is often present in the biological matrix.

The chemical method developed at the NPL is presented in detail in Appendix A of this paper. It 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 filter paper which is then subjected to measurements of activity.

2.4. Calculation of ^{90}Sr concentration

Yttrium-90 concentration in the sample was determined from the disintegration rate mea-

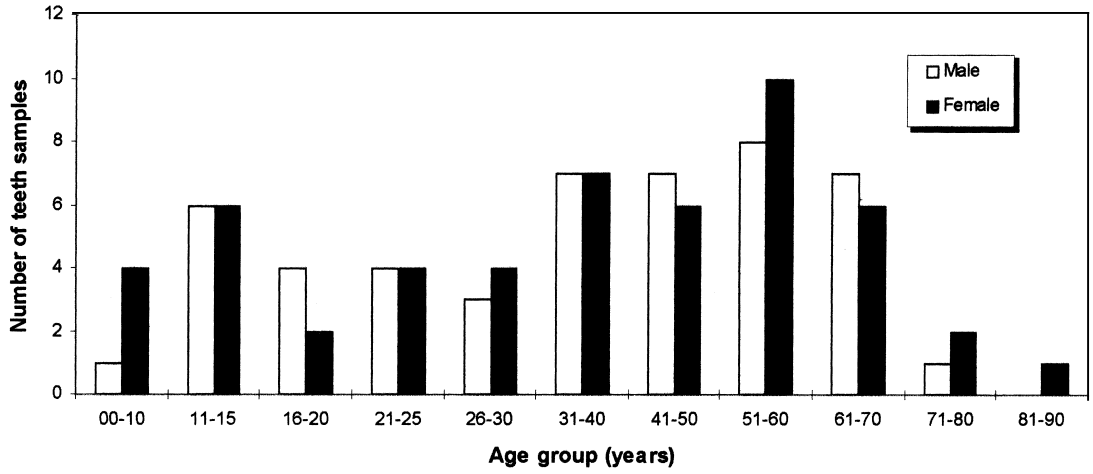

Fig. 5. Teeth samples distribution according to age and sex.

sured in 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 the calcium percentage of the bone or tooth ash is 40% (Bryant and Loutit, 1961; Glowiak et al., 1977).

All samples used in this study were measured using a Canberra 2404 α-, β-gas flow proportional counter (gas P10: Ar 90%-CH_4 10%) for at least 3 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. Background was determined from periodic measurements of a blank sample and averaged 0.69 ± 0.10 cpm (counts per minute).

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

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

in which:

Y_0 = Yttrium disintegration rate at time T_1 (in h) of ^{90}Sr removal from the sample (in cpm);
Y = Net yttrium (after subtraction of the background) disintegration rate at time T (in h), 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 h^{-1}).

The ^{90}Y activity A_Y, of the filter paper source was calculated using the expression

$$A_Y = \frac{Y_0}{60\, C_y E_Y}\,[\mathrm{Bq}] \quad (2)$$

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.

The ^{90}Sr concentration of the sample is expressed in mBq/g Ca of the sample and is calculated using the expression

$$A_S = \frac{1000 A_Y}{0.4M} \quad (3)$$

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 fraction in the ashed sample (40%).

3. Experimental results

3.1. Bone samples

Measurement of the mass of bone samples in wet and ash form yielded an estimate of the percentage of inorganic matter in each sample. The percentage of inorganic matter did not show statistically significant variations according to sex and age (confidence level = 0.01). However, statistically significant variations were evident with regard to type of bone. Samples taken from the sternum yielded $17 \pm 6\%$ inorganic matter, whereas ribs bones yielded $9 \pm 6\%$; femur epiphysis and patellae showed much higher inorganic matter content, measuring $28 \pm 7\%$.

Strontium-90 concentration in bone samples did not show statistically significant variations with regard to sex, age and residence of donor. The mean concentration of ^{90}Sr was 30 ± 13 mBq ^{90}Sr/g Ca. Fig. 6 shows ^{90}Sr bone concentrations according to age. The number in each column denotes the number of samples analysed per age group, while error bars represent one standard deviation (see also Table 1).

Statistically significant differences were found in ^{90}Sr concentration with regard to the type of bone sample measured. Patellae showed much lower values of 16 ± 7 mBq ^{90}Sr/g Ca in contrast

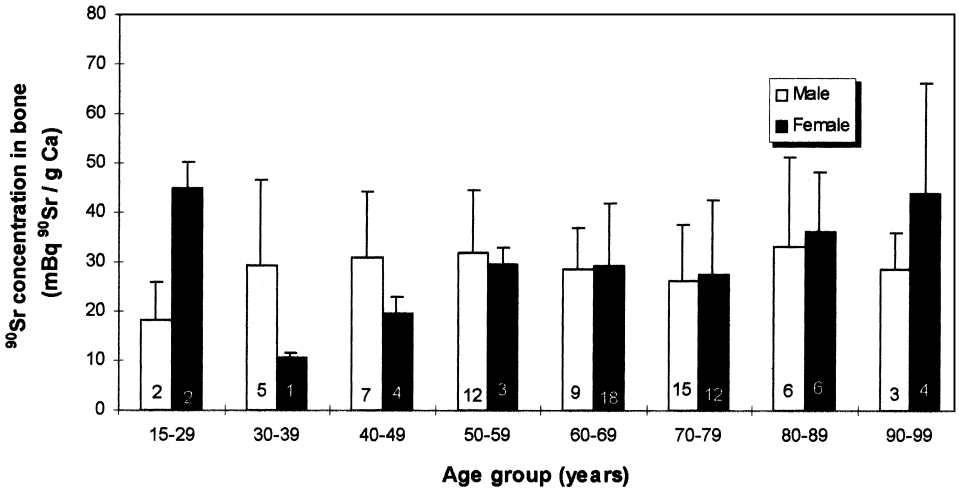

Fig. 6. ^{90}Sr concentration in bone samples according to age and sex. Each column represents the mean value of ^{90}Sr concentration for the samples in the group; error bars represent 1 S.D. Numbers in each column give the number of samples analysed per group.

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).

3.2. Tooth samples

Inorganic matter in tooth samples did not vary statistically with sex and tooth category; the mean value for all categories of teeth and for both sexes was $71 \pm 4\%$. Inorganic matter in the roots of teeth was found to be lower ($64 \pm 3\%$ for roots in women and $62 \pm 2\%$ for roots in men).

With regard to age, the inorganic matter of teeth was found, in general, to decrease for both sexes. Inorganic matter of deciduous teeth from donors with ages up to 15 years was $79 \pm 3\%$ for male donors and $76 \pm 2\%$ for female donors. In contrast, inorganic matter in teeth from donors with age 51–80 years was about 12% and 9% lower, respectively for the two sexes.

In contrast to the results from bone samples, ^{90}Sr concentration in teeth exhibited an interesting variation with regard to age. Fig. 7 shows radiostrontium concentration activities in teeth from the various age groups up to 80 years. The numbers in the columns denote the number of samples analysed per group, while error bars rep-

Table 1
Radiostrontium concentration in male and female bone samples[a]

Age (years)	Male		Female	
	(mBq ^{90}Sr/g Ca)	N	(mBq ^{90}Sr/g Ca)	N
15–29	18 ± 8	2	45 ± 5	2
30–39	29 ± 17	5	11 ± 1	1
40–49	31 ± 13	7	20 ± 3	4
50–59	32 ± 13	12	30 ± 3	3
60–69	29 ± 8	9	29 ± 12	18
70–79	26 ± 11	15	28 ± 15	12
80–89	33 ± 18	6	36 ± 12	6
90–99	29 ± 7	3	44 ± 23	4

[a] Mean values of each group with the respective 1 S.D. and the number N of samples analysed per group, are presented

Table 2
Radiostrontium concentration in bone samples according to type of bone[a]

Kind of bone	^{90}Sr concentration (mBq ^{90}Sr/g Ca)	*N*
Patella	16 ± 7	7
Rib	29 ± 10	35
Sternum	33 ± 16	40
Sternum and rib	31 ± 11	11
Femoral heads	30 ± 12	17

[a] Mean values with the respective S.D. and the number *N* of samples analysed per group are presented

resent 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 at the age of 20 years. For ages 21–40 years, ^{90}Sr concentrations increased further from 19 ± 8 mBq ^{90}Sr/g Ca to 64 ± 21 mBq ^{90}Sr/g Ca; thereafter levels decreased steadily to 4 ± 1 mBq ^{90}Sr/g Ca for the 71–80 age group.

Similar, but not as sharp variations, are observed 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 and a further increase from 24 ± 9 mBq ^{90}Sr/g Ca to 48 ± 24 mBq ^{90}Sr/g Ca up to 40 years. After the age of 40 years, ^{90}Sr concentrations decreased steadily to 6 ± 1 mBq ^{90}Sr/g Ca for the 71–80 age group. Statistically significant differences were observed between different age groups of the same sex, but not between the sexes of the same age group (see also Table 3). There were no statistically significant differences between the different types of teeth for the same age group (for either male or female).

4. Modelling 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. The large quantity of data collected for ^{90}Sr concentration in bones has been used in the development of several age-dependent models. Leggett et al. (1982) proposed a metabolic model that applies from birth through to adulthood. This model utilises compartments which corre-

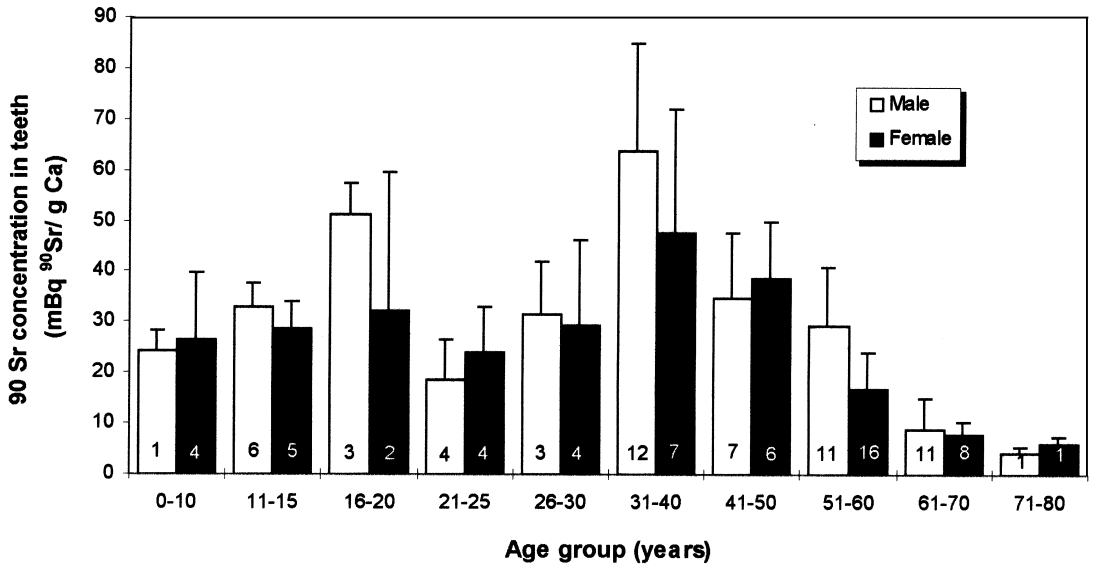

Fig. 7. ^{90}Sr concentration in teeth samples according to age and sex. Each column represents the mean value of ^{90}Sr concentration for the samples in the group; error bars represent 1 S.D. Numbers in each column give the number of samples analysed per group.

Table 3
Mean values of ^{90}Sr concentration in tooth samples for each age and sex group[a]

Age (years)	Male		Female	
	(mBq ^{90}Sr/g Ca)	N	(mBq ^{90}Sr/g Ca)	N
0–10	24 ± 4	1	26 ± 13	4
11–15	33 ± 5	6	29 ± 5	5
16–20	51 ± 6	3	32 ± 27	2
21–25	19 ± 8	4	24 ± 9	4
26–30	32 ± 10	3	29 ± 17	4
31–40	64 ± 21	12	48 ± 24	7
41–50	35 ± 13	7	39 ± 11	6
51–60	30 ± 12	11	17 ± 7	16
61–70	9 ± 6	11	8 ± 2	8
71–80	4 ± 1	1	6 ± 1	1

[a]One standard deviation and the number N of samples analysed per group are also presented.

spond to physical processes or subsections of the skeleton and examines the behaviour of ^{90}Sr in terms of the behaviour 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 these two categories, the skeleton may be viewed as consisting of three compartments, with two compartments associated with structural bone (cancellous and compact bone) and the third associated with metabolic bone (bone surface). Cancellous and compact bones are differentiated in terms of their surface to volume ratio. Surface to volume ratios may vary significantly within bone types and the distinction between the two categories may not be readily evident at some points in the skeleton and at some stages of development (Leggett et al., 1982). Thus the classification of bone as cancellous or compact represents a simplification introduced for the purposes of the model.

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

$$\frac{dQ(t)}{dt} = A(t)B(t)F(T+t) - L(t)X(t) - \lambda Q(t) \quad (4)$$

in which

- $A(t)$ is the annual amount of calcium intake as a function of age t (g Ca y^{-1}),
- $B(t)$ is the percentage of ^{90}Sr contained in food which is taken up by the skeletal compartment as a function of age t,
- $F(T+t)$ is the mean ^{90}Sr concentration in food during calendar year $t+T$ (Bq/g Ca),
- $L(t)$ is the annual rate of bone turnover which is assumed to be equal to the annual removal rate of ^{90}Sr from the bones (kg y^{-1}); $L(t)$ depends on the type of bone involved in the exchange process described by Eq. (4))
- $X(t) = \frac{Q(t)}{M(t)}$ is the concentration of ^{90}Sr (Bq/kg) in the skeletal; compartment of total mass $M(t)$; and
- λ is the radiological decay constant of ^{90}Sr (y^{-1}).

The integration of Eq. (4) requires knowledge of the functions $A(t)$,$B(t)$, $F(T)$, $L(t)$ and $M(t)$. With the exception of the level of ^{90}Sr contami-

nation in food $F(T)$ 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 in Appendix B.

There is scant data concerning radiostrontium contamination of foodstuffs during the past 40 years in Greece. Some information is provided by measurements of 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 (Bakacs-Polgar and Kurcs-Csiky, 1964) was adopted as the percentage of ^{90}Sr in the total β fallout. The ratio of ^{90}Sr in all foodstuffs to that in milk was taken as 1.5 (Leggett et al., 1982). The function $F(T)$ obtained in this fashion, albeit with considerable uncertainty, is plotted in Fig. 10 (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).

4.3. Model estimates of ^{90}Sr levels in skeletal compartments

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 cancellous bone were used in the case of bone samples and for compact bone in the case of teeth. The predictions of the model, by employing the response functions proposed (see Appendix B) by Leggett et al. (1982) are contained in Figs. 8 and 9.

Although a straightforward application of the model gave a satisfactory reproduction of the overall trend observed in the data of Figs. 6 and 7, it was realised that considerable improvement could be obtained by removing some of the uncertainties associated with the response functions in Eq. (4).

As already noted, the major uncertainties in the model arise from:

1. Incomplete knowledge of the levels of ^{90}Sr in foodstuffs in Greece during the past four decades [function $F(T)$].

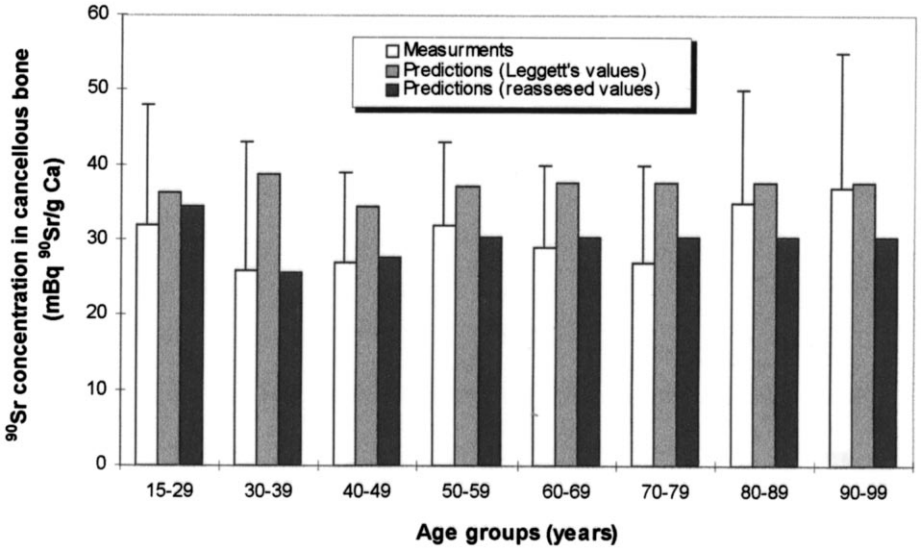

Fig. 8. Mean age-group values of ^{90}Sr concentration in bone samples and predictions of the NPL model. White bars represent experimental data, grey bars predictions of the model by using the response function suggested by Leggett et al. (1982) and dark bars predictions obtained by using reassessed values of the parameters of response functions (cancellous bone turnover rates and ^{90}Sr concentration in food in Greece).

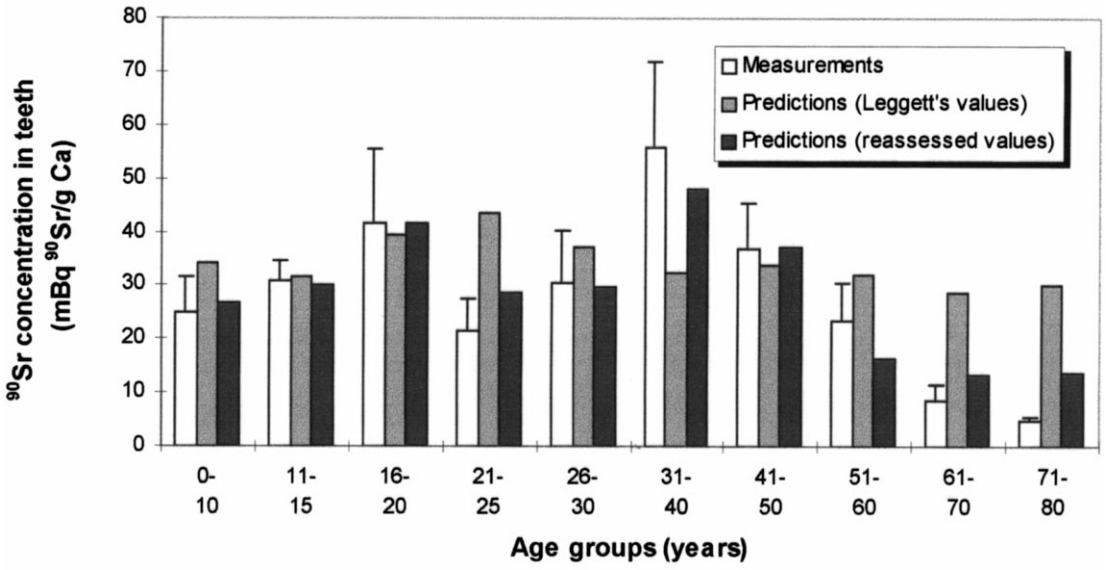

Fig. 9. Mean age-group values of ^{90}Sr concentration in teeth samples and predictions of the NPL model. White bars represent experimental data, grey bars predictions of the model by using the response function suggested by Leggett et al. (1982) and dark bars predictions obtained by using reassessed values of the parameters of response functions (teeth turnover rates and ^{90}Sr concentration in food in Greece).

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 functions 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 cancellous 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 in Figs. 8 and 9. Details with regard to modifications of the response functions are given in Appendix B. Predictions of the model with the adjusted response functions appear to reproduce the data very well.

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 behaviour of the effect it represents.

The functions $F(t)$ and $L(t)$, as determined from the fitting process, are contained in Figs. 10–12. The main modification arising from the fitting process with regard to function $F(t)$ seems to be the adjustment of the relative impact of the Chernobyl accident in comparison with that of the atmospheric nuclear weapons tests period. As seen in Fig. 10, the effect of the latter on the contamination of foodstuffs was more intense and lasted longer. Reassessed values of the turnover response function for cancellous bone, although they do not show any drastic change in structure, are predicted to be much higher than the ones used in the original calculations, especially for adults over 45 years of age (see Fig. 11). There is a dramatic difference between reassessed and original values of function $L(t)$ for compact teeth, which are represented in a logarithmic scale in Fig. 12. The very high excursion at around age 8 years, introduced by the fitting process, corresponds to the formation of permanent teeth.

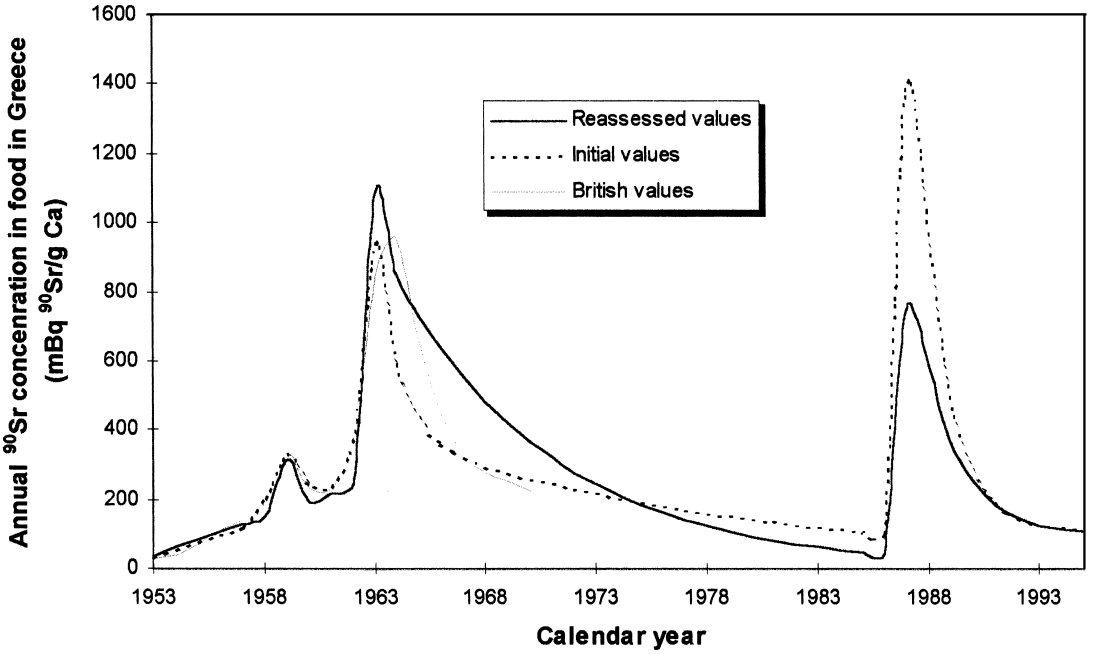

Fig. 10. Mean annual values of ^{90}Sr concentration in food in Greece for the period 1953–1994 (dashed line; K. Stamoulis, 1998) and assessed values derived by fitting model predictions to measurements of bone and teeth samples (solid line). The heavier line extending to 1970 represents values of ^{90}Sr concentration in foodstuffs in the UK.

5. Discussion and conclusions

The human skeleton carries with it the history of radiostrontium environmental contamination during the period that the individual was alive. As shown by the data in Fig. 1, any increase in ^{90}Sr in the environment affects within a very short period 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 passes 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 cancellous 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 tissue.

Human bone samples from Greece measured in this study 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 cancellous 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, data from human teeth show a very interesting structure. In the Ukrainian results presented in Fig. 3, the data, obtained 5 years after the Chernobyl accident, contain three prominent peaks with centroids at the ages of 5.0 ± 0.2, 14.2 ± 0.4 and 35.4 ± 0.5 years. The first two correspond to children who were born or were shedding their

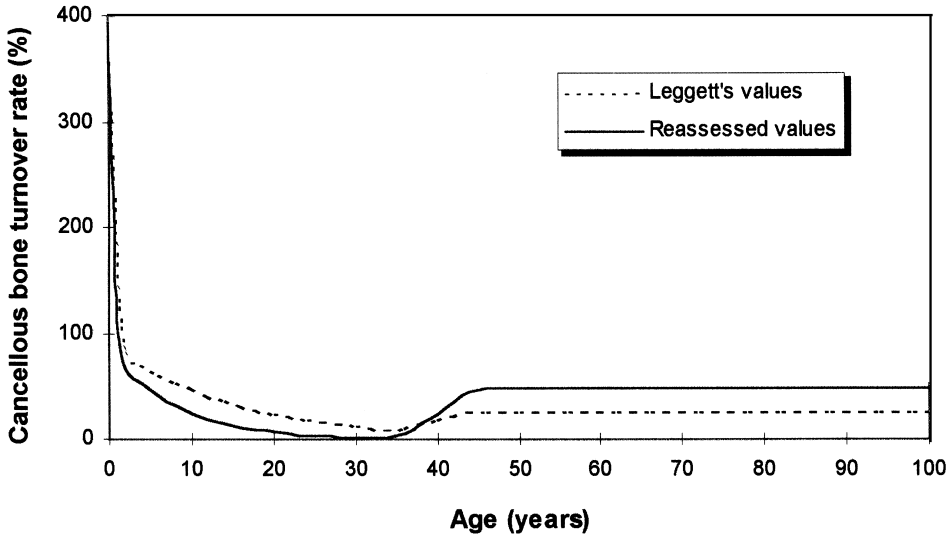

Fig. 11. Cancellous bone turnover rates (dashed line) and response function derived by fitting model predictions to measurements of bone samples.

deciduous teeth at the time of the Chernobyl accident. The third broader peak corresponds to individuals who were growing up during the period of the atmospheric nuclear weapons tests during the 1960s. In the Greek data which were collected about 3 years later and are presented in Fig. 9, the first peak observed in the Ukrainian data (expected in the age group 0–10 years of age) is missing. However, the later two peaks, with centroids at the age of 17.5 ± 0.4 and 38.5 ± 1.2 years are evident. This shift of about 3.5 years coincides with the time elapsed between the collection of the two sets of data.

As seen in Figs. 8 and 9, the model developed

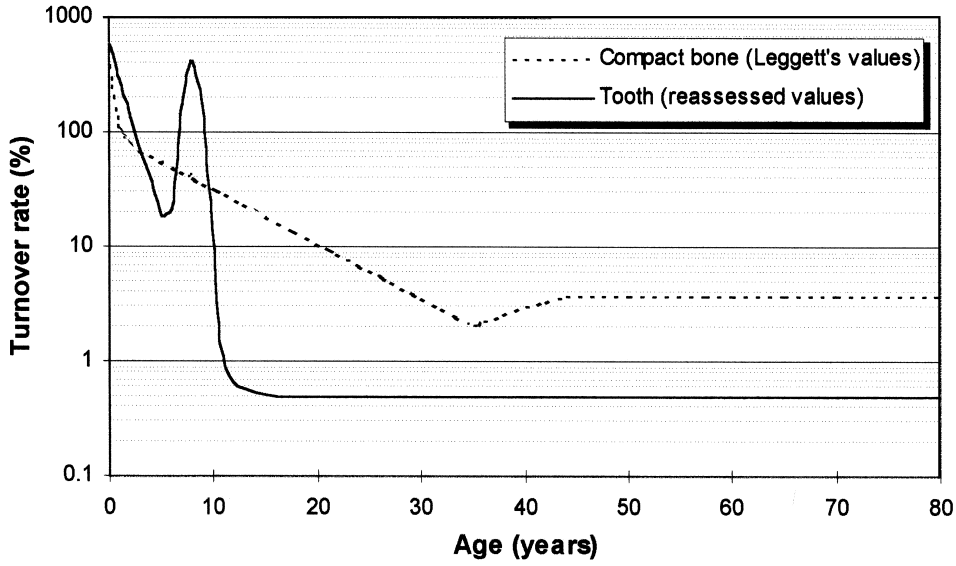

Fig. 12. Compact bone turnover rates (dashed line) and response function derived by fitting model predictions to measurements of teeth samples.

at the NPL was 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 foodstuffs in Greece for the past 40 years.

The results reported here clearly indicate that the contaminating effects of the atmospheric nuclear weapons testing in the 1960s exceed by far those of the Chernobyl accident. From both Figs. 3 and 9, 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 individuals from the 1960s. This is corroborated by the findings presented in Fig. 10, which demonstrate that contamination of foodstuffs in the 1960s far exceeded that caused by the Chernobyl accident both in intensity and duration.

Appendix A: The NPL method for chemical treatment of bone and teeth samples

Bone and tooth samples were treated chemically prior to radiostrontium concentration measurement according to the following method.

A pre-weighed quantity of bone or tooth ash (4.0–10.0 g) was placed in a 150-ml glass beaker and a 2 M hydrochloric acid solution was added (40–100 ml). Using magnetic stirring the ash was diluted completely and 1.0 ml of Y^{3+} carrier was added to the solution. By adding a small quantity of hydrochloric acid solution (2 M) or ammonia solution (25% w/v) the pH was adjusted to about pH ≈ 1. The volume of the solution was recorded and the solution was transferred into a separating funnel (125 ml or more). Solution of BEHHP in dodecane, 5% (v/v), in the amount of one-quarter of the previously recorded volume, was added to the separation funnel and the mixture was shaken vigorously for about 3 min. Time T_1 of yttrium separation from the inorganic solution was recorded for subsequent calculation of the ^{90}Y disintegration. The organic and aqueous phases were allowed to separate for about 30 min.

The aqueous (lower) phase was discarded and 25 ml of 6 M nitric acid solution was added to the organic (upper) phase. The separating funnel was shaken vigorously for about 3 min and the phases were allowed to separate for 30 min. Subsequently, the aqueous phase was transferred to a 50-ml centrifuge tube. The procedure was repeated and the aqueous phase was transferred to yet another centrifuge tube.

A quantity of 25 ml ammonia solution (25% w/v) was added to the centrifuge tubes in which, after stirring, yttrium hydroxide gel formed. The centrifuge tubes were allowed to cool at room temperature and were centrifuged at 3500 rev./min for about 5 min. The supernate was discarded and 5 ml of 2 M hydrochloric solution were added under stirring to the precipitate of yttrium hydroxide. Distilled water in the amount of 20 ml was added to the solution of each centrifuge tube and the pH was adjusted to about 1.7–1.8 with hydrochloric acid or ammonia solution.

The centrifuge tubes were placed in a water bath at 90°C for about 30 min and 5 ml of saturated solution of oxalic acid was added to each tube under stirring. White precipitate of yttrium oxalate was thus formed. A filter paper (Whatman No. 42) was weighed and placed in a vacuum filtering apparatus. Solutions with the precipitates were allowed to cool at room temperature and were filtered trough the filter paper. A few ml of distilled water was added to the centrifuge tubes and was also filtered through the same paper.

The filter paper was dried in an oven at 80°C for about 20 min and allowed to cool at room temperature. The filter was weighed and yttrium yield was calculated from the yttrium oxalate (in the form of $[(COO)_2]_3Y_2 \cdot 9H_2O$) on the filter. The filter was then transferred to the β-counter apparatus and measured for activity.

Appendix B: Physiological semi-empirical formulas employed by the NPL model

The physiological response functions needed in

the evaluation of the basic model equation Eq. (4) were derived from data in the scientific literature.

Annual ^{90}Sr concentration in food in Greece was initially estimated using available data on total β fallout, and scatter data on ^{90}Sr concentration in milk during the period 1961–1971 and 1986–1993. Estimates were calculated by fitting data on total β fallout and milk ^{90}Sr concentration with the UNSCEAR model (Lassey, 1979), which predicts milk ^{90}Sr concentration from ^{90}Sr fallout data. The percentage of ^{90}Sr in total β fallout was taken as 0.24%, which was calculated from data in total β activity and ^{90}Sr concentration in soils given by Bakacs-Polgar (Bakacs-Polgar and Kurcs-Csiky, 1964). The function $F(T)$ derived in this way is given by

$$F(T) = \begin{cases} f_1\exp[f_2(T-1952)] + f_3\exp[-f_4(T-1959)^2] & \\ \quad + f_5(T-1952) + f_6 & 1953 \le T \le 1963 \\ f_7\exp[-f_8(T-1962)] & \\ \quad + f_9\exp[-f_{10}(T-1962)] & 1963 \le T \le 1986 \\ f_{11}\exp[-f_{12}(T-1985)] + f_{13} & 1986 \le T \le 1995 \end{cases}$$

Table 4
Values of the parameters of the function of annual ^{90}Sr concentration in food in Greece $F(T)$[a]

Parameters	Initial values	Reassessed values
f_1	$2.21\cdot10^{-5}$	$1.14\cdot10^{-5}$
f_2	1.57	1.60
f_3	171	191
f_4	0.95	1.16
f_5	21.2	22.3
f_6	11.2	17.7
f_7	1370	1316
f_8	0.92	1.93
f_9	418	814
f_{10}	0.06	0.12
f_{11}	2846	1331
f_{12}	0.8	0.54
f_{13}	112	98.7

[a]The second column contains values derived from the initial estimate of function $F(T)$, while in the third column contains reassessed values of the parameters obtained by fitting the model to experimental data.

where T is the calendar year. Values of the parameters f_k, $k = 1,2,...,13$ are given in the second column of Table 4. In the third column of the same table the reassessed values of the parameters derived by fitting model to the experimental data of ^{90}Sr concentration in bones and teeth are also shown.

Bone and teeth turnover rates were reassessed also by fitting the model to respective ^{90}Sr concentrations. The fitting was performed independently using the initial estimates for ^{90}Sr intake presented in the second column of Table 5.

According to Leggett et al. (1982), bone turnover rate as a function of age t is given by the function

$$L(t) = \begin{cases} \dfrac{l_1}{C(t)} & 0 \le t \le 1.5 \\ l_2\exp(-l_3 t) & 1.5 \le t \le 35 \\ l_4 t - l_5 & 35 \le t \le 44 \\ l_6 & 44 \le t \end{cases}$$

where $C(t)$ is the total amount of calcium in the skeleton at age t. Values of the parameters l_k $k = 1,2,...,6$ are given in the second and third column of Table 5. These values were used by our model to predict ^{90}Sr concentration in teeth and cancellous bone respectively. In the fourth column of the same table the reassessed values for the parameters used for cancellous bone are presented, derived by fitting the model to the ^{90}Sr concentration measurements for cancellous bone.

In order to obtain better agreement for the ^{90}Sr concentration in teeth a different function was used in this case

$$L_{\text{teeth}}(t) = l_{t1}\exp[-l_{t2}t] + l_{t3}\exp\Big[-l_{t4}(t-l_{t5})^2\Big] + l_{t6}$$

in which t is the age of the donor. The use of the gaussian factor in the proposed function is based on the fact that deciduous teeth fall-out during the period 6–10 years of age, with a maximum occurring at the age of 8 years. During this period, turnover rate increases rapidly and takes values of the same order as during the calcifica-

Table 5
Values of the parameters of the function of compact and cancellous bone turnover rate $L(t)$[a]

Parameters	Leggett's values compact bone	Leggett's values cancellous bone	Reassessed values cancellous bone
l_1	104.3	104.3	110.3
l_2	0.975	0.912	0.871
l_3	0.11	0.064	0.128
l_4	$1.8\cdot10^{-3}$	$1.75\cdot10^{-2}$	$3.06\cdot10^{-2}$
l_5	$4.24\cdot10^{-2}$	$51.7\cdot10^{-2}$	$51.5\cdot10^{-2}$
l_6	0.037	0.255	0.493

[a]The second and third column contain Leggett's values, while in the fourth column contains reassessed values of the parameters obtained by fitting the model to ^{90}Sr concentration measurements in cancellous bone

Table 6
Values of the parameters of the function of teeth turnover rate $L_{teeth}(t)$[a]

Parameters	Assessed values teeth
l_{t1}	6.95
l_{t2}	0.75
l_{t3}	4.3
l_{t4}	0.9
l_{t5}	8.1
l_{t6}	$0.9\cdot10^{-2}$

[a]The second column contains reassessed values of the parameters obtained by fitting the model to ^{90}Sr concentration measurements in teeth

tion of the deciduous teeth. The constant factor l_{t6} was used in order to determine the long term but low turnover rate in teeth during adulthood. Assessed values of the parameters $l_{tk}k = 1,2,...,6$, derived by fitting the model to ^{90}Sr concentration measurements in teeth, are given in Table 6. It should be noted that the assessed value for parameter l_{t5} is consistent with the assumption which lead to the introduction of the gaussian term.. Similarly, the reassessed value for constant turnover rate during adulthood, l_{t5}, is reasonable compared to the very low turnover rates of the compact bone proposed in the literature (approx. 1%, for the age period 20–60 years, ICRP Publication 23, 1974).

The calibration of the model described above was performed in the following order: First, the parameters l_k and l_{tk} for cancellous bone and teeth turnover rates respectively, were allowed to vary, using the initial estimated values of ^{90}Sr concentration in food in Greece (second column of Table 4). Using the reassessed values of the parameters l_k and l_{tk}, values of the parameters of the function $F(T)$ were reassessed by again fitting the model to ^{90}Sr concentration measurements in cancellous bone and teeth; mean values of the parameters f_k obtained from the two separate fits are presented in the third column of Table 4, while the function $F(T)$ thus determined is contained in Fig. 10. Fig. 10 also contains graphically the initial estimate of function $F(T)$ and the corresponding values of annual ^{90}Sr concentration in food in the UK (Leggett et al., 1982).

Predictions of ^{90}Sr concentration in cancellous bone using Leggett's values and the reassessed values for turnover rates are presented in Fig. 8, while predictions of ^{90}Sr concentration in teeth using Leggett's values for compact bone and the assessed values for teeth, are presented in Fig. 9. In Figs. 11 and 12 turnover rates for cancellous bone and teeth are, as obtained from the reassessed parameter values in Tables 5 and 6, compared to the original functions proposed by Leggett.

During the fitting process, free parameters were initially allowed to vary within 30% of the values found in the literature. However, some of the parameters yielded numerical values which were at the limit of their allowed range. For these parameters the range was extended and minimum χ^2 was reached for numerical values which, in some cases (see Tables 4 and 5) differed by as much as 100% from initial estimates. The stability and uniqueness of the solution reached was tested

by repeating the fitting process from different sets of *ad hoc* initial values.

Finally, the mass of each skeletal compartment $M(t)$ was taken as the percentage (20% cancellous and 80% compact bone) of the total skeleton mass $M_{\text{tot}}(t)$, which was calculated based on measurements for the Greek population (Mantzagrioti-Meimaridi et al., 1981).

$$M_{tot}(t) = \begin{cases} -3.99\times10^{-6}t^5 - 1.61\times10^{-4}t^4 + 9.55\times10^{-3}t^3 \\ \quad -0.119t^2 + 0.769t + 0.57 & 0 \le t \le 20 \\ 9.5 & 20 \le t \le 30 \\ -1.96\times10^{-4}t^2 - 3.94\times10^{-2}t \\ \quad +11.374 & 30 \le t \end{cases}$$

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