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Variations in the concentration of plutonium, strontium-90 and total alpha-emitters in human teeth collected within the British Isles

R.G. O'Donnell^a, P.I. Mitchell^a, N.D. Priest^{b,*}, L. Strange^b, A. Fox^b,
D.L. Henshaw^c, S.C. Long^d

^aDepartment of Experimental Physics, University College Dublin, Belfield, Dublin 4, Ireland

^bBiomedical Research Department, AEA Technology, 551 Harwell, Oxon, OX1 0RA, UK

^cH.H. Wills Physics Laboratory, University of Bristol, Bristol BS8 1TL, UK

^dRadiological Protection Institute of Ireland, Clonskeagh, Dublin 14, Ireland

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Abstract

Concentrations of plutonium-239, plutonium-240, strontium-90 and total α -emitters have been measured in children's teeth collected throughout Great Britain and Ireland. The concentrations of plutonium and strontium-90 were measured in batched samples, each containing approximately 50 teeth, using low-background radiochemical methods. The concentrations of total α -emitters were determined in single teeth using α -sensitive plastic track detectors. The results showed that the average concentrations of total α -emitters and strontium-90 were approximately one to three orders of magnitude greater than the equivalent concentrations of plutonium-239,240. Regression analyses indicated that the concentrations of plutonium, but not strontium-90 or total α -emitters, decreased with increasing distance from the Sellafield nuclear fuel reprocessing plant — suggesting that this plant is a source of plutonium contamination in the wider population of the British Isles. Nevertheless, the measured absolute concentrations of plutonium (mean = 5 ± 4 mBq kg⁻¹ ash wt.) were so low that they are considered to present an insignificant radiological hazard. © 1997 Elsevier Science B.V.

Keywords: Plutonium; Strontium; α -Particles; Teeth; Bones; Geographical variations

* Corresponding author. Current address: School of Environmental Science, Middlesex University, Bounds Green Road, London N11 2NQ, UK. Tel.: +44 181 3625229; fax: +44 181 3611726; e-mail: n.priest@mdx.ac.uk

1. Introduction

In 1984 an advisory group, under the chairmanship of Sir Douglas Black, reported the results of an investigation into a claimed linkage between the incidence of childhood leukaemia in the Seascale area of West Cumbria and radionuclide discharges by British Nuclear Fuels Ltd, Sellafield (Black, 1984). This investigation failed to confirm a linkage, but recommended that more attention be concentrated on measuring radiation doses actually received by members of the public in West Cumbria and other relevant areas, including control areas.

In 1985 and 1988, Popplewell et al. published the results of their analysis of plutonium levels in a limited number of samples of human autopsy tissue collected in Great Britain. An elevated level was observed in samples from former residents of Cumbria. Later the results of a further study were reported (Popplewell et al., 1989). This employed isotopic determinations and provided strong circumstantial evidence that plutonium, of BNFL, Sellafield aerial discharge origin, had found its way into the local populace. However, the number of samples measured in these studies were low and the studies were not designed to measure the effect of distance from Sellafield on the concentration of skeletal plutonium in the British Isles. Moreover, legal considerations now preclude further widespread analyses of autopsy tissues, making it impossible to expand this study to gain such information. It follows that alternative methods must be employed.

Teeth are an extension of the skeleton and accumulate contaminating stable and radioactive bone-seeking metals that enter the body. Moreover, studies with radium-226 (Yamamoto et al., 1994), lead (Stack, 1990) and other metals have established that their levels in teeth are related to skeletal content/body burden and, while the collection of autopsy tissues is now difficult, teeth removed from adolescent children for orthodontic purposes are freely available. It follows that the analysis of teeth provides a viable alternative to the analysis of bone samples for the study of geographical variations in bone-seeking radionuclide contamination.

For the present study, teeth were collected from as many of the UK District Health Authorities as possible and from the Republic of Ireland. These were analysed for their anthropomorphic plutonium and radiostrontium, and largely natural total α -emitter content. The spatial distribution of measured concentrations with respect to the BNFL, Sellafield plant was determined. The collection of the teeth was undertaken by the University of Bristol, which also undertook the measurement of total α -emitters in selected teeth. Batches of approximately 50 teeth were then sent to either AEA Technology, Harwell or University College Dublin for plutonium/strontium-90 analysis. Project coordination and statistical analyses were completed at AEA Technology.

2. Materials and methods

2.1. Teeth

All the teeth used for the study were intact permanent teeth removed for orthodontic purposes. The teeth were collected by dentists, who also recorded the identification of each tooth, the age and sex of the donor and the date extracted. Most teeth collected were upper/lower, left/right first premolars (4's) from 13 to 14-year-old adolescents (38% male, 62% female) — these being the most common teeth removed by orthodontists. Teeth collected from West Cumbria were sent first to AEA Technology, Harwell, due to the presence of a preexisting arrangement between this region and the United Kingdom Atomic Energy Authority. Teeth were batched from each Area Health Authority as sufficient numbers of similar teeth became available and sent for analysis. At the end of the collection period, sufficient teeth for batch analysis had been collected from the following regions: Antrim; Armagh and Down; Avon; Barnet; Berkshire; Blackburn; Blackpool and Fylde; Bristol; Bromley; Central Scotland; Cheltenham; Cheshire; Cleveland; Clwyd; Croydon; Cumbria; Dewsbury; Barnstaple; Dorset; Dumfries and Galloway; Gloucestershire; Grampian; Durham; Exeter; Fife; Forth Valley; Gateshead; Glasgow; Greater London; Gwynedd; Hampshire; Hereford and Worcester; Hertford-

shire; Highlands; Humberside; Isle of Wight; Kent; Lancashire; Leicestershire; Lincolnshire; Lothian; Manchester; Merseyside; Northampton; North and South Tees; North Yorkshire; Northumberland; Norwich; Oxfordshire; Powys; Sheffield; South Cumbria; South Glamorgan; Staffordshire; Strathclyde; Sunderland; Surrey; Tyne and Wear; Warwickshire; Weston-super-Mare; West Cumbria; West Glamorgan; West Sussex; Wexford; Wiltshire; Winchester; Wolverhampton. Teeth from other areas were available for single analysis. In total, 67 batches of teeth were analysed for plutonium-239,240 (including 6 from Cumbria), 71 batches for strontium-90 (including 6 from Cumbria) and 53 individual teeth (1 from Cumbria) for their total α -emitter content.

2.2. Plutonium analyses

The plutonium-239,240 content of batched samples of teeth was determined by low background α -spectrometry. Each batch was dry ashed at 550°C in a muffle furnace for 1 week then weighed to derive ash wt. The ash was dissolved in hot, concentrated aqua regia (150 ml 12 M hydrochloric acid and 50 ml 15 M nitric acid) and known quantities of ^{242}Pu and ^{85}Sr yield tracers added. Plutonium was separated from strontium by a partial precipitation technique using 30 mg of Fe^{3+} to scavenge plutonium and 1 g of anhydrous strontium nitrate to act as a carrier for the ^{90}Sr present. After the addition of these agents the acidic solution was first diluted, then neutralised, to pH 4–4.5, by the addition of ammonium hydroxide. A precipitate formed which comprised iron hydroxide plus plutonium and the supernatant contained strontium (this was used for the strontium analysis). The precipitate was collected, dried, re-dissolved in 50 ml of 8 M nitric acid, then purified using an ion-exchange column (BIO-RAD AG 1-X4). Plutonium was eluted from the resin using 0.1 M NH_4I in conc. HCl, evaporated to dryness, redissolved in 3 M HCl and electroplated from a chloride plating solution (1A, 45 min) onto 25-mm diameter polished stainless steel discs. These were flamed at 500°C for 30 s to 'fix' the plutonium, then

counted for a minimum of 2×10^6 s using a Canberra Model 7404 Quad α -spectrometer equipped with low background, passivated, ion-implanted silicon detectors (PIPS). The quantity of plutonium-239,240 in each sample was determined from the measured ratio of $^{239,240}\text{Pu}:^{242}\text{Pu}$, by reference to the known quantity of the latter added to the sample. The detection limit of this technique was estimated to be 25–50 μBq .

2.3. Strontium-90 analyses

Following the removal of precipitate for plutonium analysis the above mentioned supernatant was further processed to determine ^{90}Sr . This was evaporated to dryness, then re-dissolved in a mixture (73:27%) of fuming nitric acid and distilled water to separate strontium from calcium. Subsequently, barium, radium and lead were removed using barium chromate according to the method of Sunderman and Meinke (1957) and the purification completed using an iron hydroxide scavenge as proposed by Glendenin (1945). After these purification steps sodium carbonate was added to the resultant solution to precipitate strontium as the carbonate. The precipitate was separated and treated with 73% fuming nitric acid to produce insoluble strontium nitrate. This final, almost instantaneous, precipitation removes residual traces of calcium and also the coloured chromate ion which interferes with Cerenkov counting. It also provided an unequivocal time at which there is no ingrown ^{90}Y present in the sample. This time was recorded and used as $t = 0$ in ^{90}Y ingrowth equations.

The strontium sample produced was assayed for ^{85}Sr using a low-background, high-resolution germanium detector to measure its 514 keV emission. The chemical recovery of strontium was calculated by comparison with a calibrated standard solution. Strontium-90/yttrium-90 was determined by Cerenkov counting using a LKB Quantulus 1220, low-background, liquid scintillation counter. Using these methods the minimum detection level for ^{90}Sr was calculated to be 15.3 ± 1.2 mBq.

2.4. Total alpha-assay

The total α -activity within single teeth was determined using α -sensitive plastic track detectors. Teeth selected for assay were first cut in the plane of their long axis (coronal), then the cut surface of each half was placed in contact with a small sheet of high purity plastic α -detector (PADC — Tastrack, Track Analysis Systems Ltd, Bristol, UK) and clamped in position. The assembly was sealed in a high density polyethylene bag (to prevent the influx of radon) at the storage temperature of -20°C for approximately 180 days. At the end of this period each assembly was taken apart and the plastic was removed. The sheets were then individually etched under controlled conditions in caustic solution (6.25 M sodium hydroxide at 75°C for approx. 6 h) to reveal the tracks produced by α -particles during their passage into the plastic detector. The number of α -tracks, per unit area of tooth, was then counted using an automatic image-analysis system. This was used to measure the values of up to 14 parameters for each recorded α -particle track. These measurements allowed α -spectroscopy of the recorded activity, although for the present work only an estimate of total α -activity was made. This method is described in detail elsewhere (Bondarenko et al., 1996).

2.5. Analysis of data

For each measurement of radioactivity, a distance of the domicile of the donors from the BNFL Sellafield reprocessing plant was allocated. In the case of a city, the approximate geographical centre of the city was used for this purpose. Where the source of teeth was a county, the distance from Sellafield to the major population centre within this county was allocated.

Simple linear regression models were fitted to the data using the PROC REG procedure of the SAS (Statistical Analysis System) statistical package, version 6.08 (SAS, 1990). Radionuclide concentrations were plotted against distance from Sellafield and regressions were performed using both the original values and the \log_e transformed

values for radionuclide concentrations, applying the model:

$$y \text{ (or } \log_e y) = mx + c$$

where y is the dependant variable (radionuclide concentration) and x is the independent variable (distance in miles from Sellafield). Where preliminary analyses of the data, together with examination of the residual variation indicated that the radionuclide distributions were log-normally distributed, the models based on the \log_e transformed data were used. In all cases where a 'less than' value was recorded for a measurement the result was halved, in accordance with common practice, prior to their inclusion in the analysis.

In addition to the above, the mean plutonium concentration for teeth samples collected at distances up to 50 miles from Sellafield, 50–150 miles from Sellafield and greater than 150 miles, respectively, was calculated.

3. Results

The mean concentrations of plutonium-239,240, strontium-90 and total α -emitters in the teeth analysed are presented in Table 1. The data show that the average plutonium concentrations in teeth collected from within the United Kingdom and Republic of Ireland (5 mBq kg^{-1} ash wt.) are more than three orders of magnitude lower than the levels of total α -activity and of strontium-90 — these being broadly similar at approximately 8 Bq kg^{-1} and 7 Bq kg^{-1} ash wt., respectively. Analysis of the data showed that the plutonium concentrations and strontium concentrations were log-normally distributed with each distribution biased towards lower radionuclide concentrations. It follows that the median concentrations of these, also presented in Table 1, were lower and that for most individuals, the ratio total- α :Pu- α would be in the region of 3,000:1.

The results of the regression analyses for plutonium-239,240, strontium-90 and total α -emitters plotted against distance from Sellafield are shown in Figs. 1–3, respectively. Fig. 1 shows that plutonium concentrations are strongly correlated (P

Table 1
Plutonium-239,240 and strontium-90 concentrations and total α -content of teeth collected within Great Britain and Ireland

Radionuclide	<i>n</i>	Mean content (\pm S.D.) (Bq kg ⁻¹ ash wt.) ^a	Median content (\pm S.D.) (Bq kg ⁻¹ ash wt.) ^a
^{239,240} Pu	67	0.005 \pm 0.004	0.002 \pm 0.003
⁹⁰ Sr	71	8 \pm 5	6 \pm 2
Total α -emitters	53	7 \pm 2	7 \pm 2

^aAll values given to one significant figure.

= 0.008) with distance from this nuclear plant, but the other figures suggest no such relationship exists for either strontium-90 or for total α -activity ($P = 0.97$ and 0.91 , respectively). In the case of plutonium the analyses demonstrated that the plutonium content of teeth (C) was related to distance in miles from Sellafield (D) according to the following expression:

$$\log_e C = 1.5314 + 0.0046D \text{ mBq kg}^{-1} \text{ ash wt.}$$

According to this expression, plutonium concentrations in teeth peak close to the Sellafield plant, where the predicted mean concentration is 5.8 mBq kg⁻¹ ash wt. (95% confidence limits 8.6 and 3.9 mBq); then fall to approx. 1.8 mBq kg⁻¹ ash wt. (95% confidence limits 2.8 and 1.2 mBq) at 300 miles from the plant. This prediction is consistent with the plutonium concentrations measured for teeth samples collected at different distances from Sellafield (Table 2).

4. Discussion

Given the detection limit for plutonium by low-level α -spectrometry it was not possible, at the time of the study, to analyse individual teeth for this element. Consequently, only batched samples comprising approximately 50 teeth were analysed. Even so, the plutonium content of some batches was at the detection limit. It follows, that

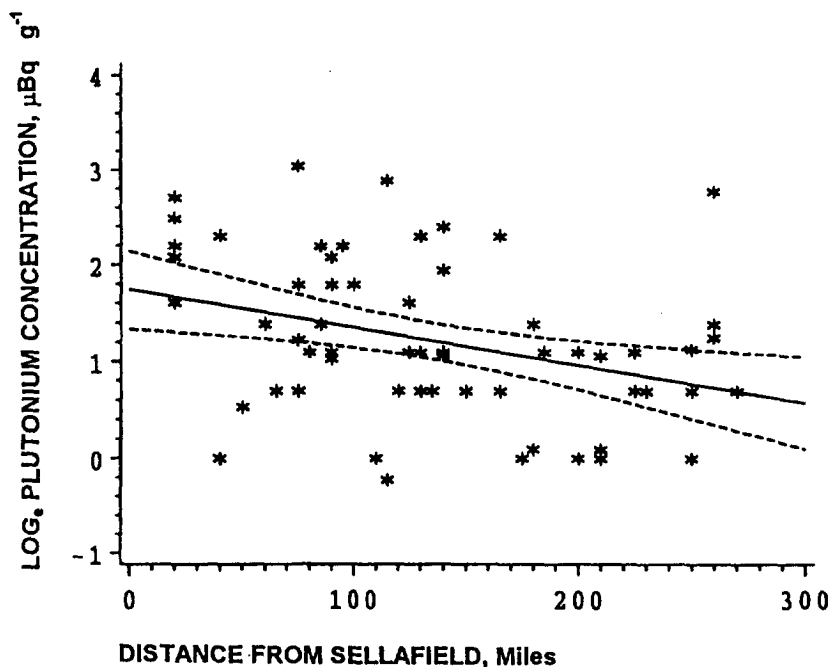


Fig. 1. \log_e ^{239,240}Pu content of children's teeth vs. distance from Sellafield. Regression equation: $\log_e [^{239,240}\text{Pu}] = 1.5314 - 0.0046 \times \text{Distance from Sellafield (miles)}$. Statistical significance: $P < 0.0081$.

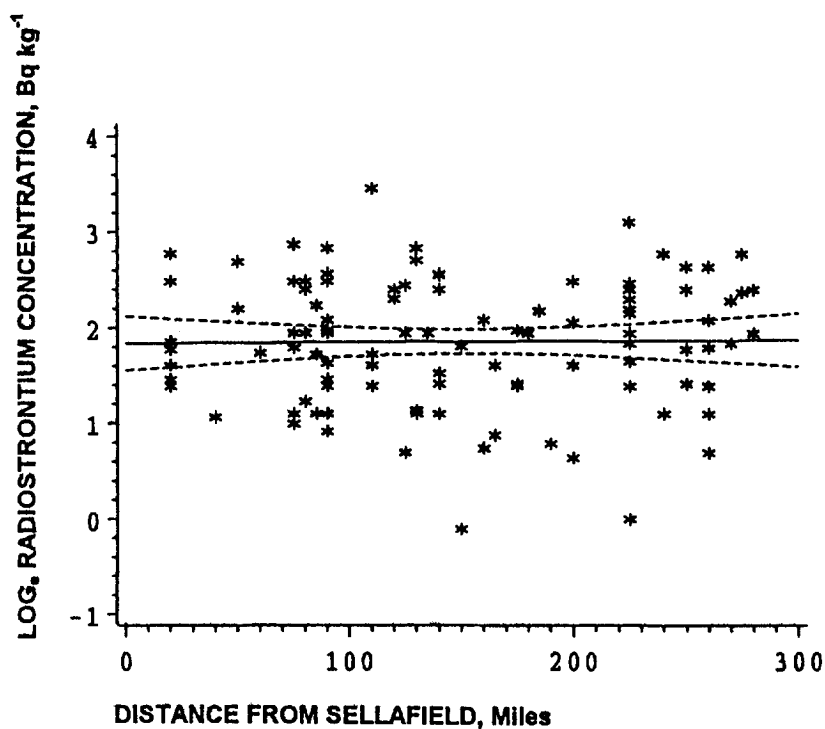


Fig. 2. \log_{10} ^{90}Sr content of children's teeth vs. distance from Sellafield. Regression equation: $\log_{10} [^{90}\text{Sr}] = 1.8338 + 0.00003 \times \text{Distance from Sellafield (miles)}$. Statistical significance: $P < 0.9741$.

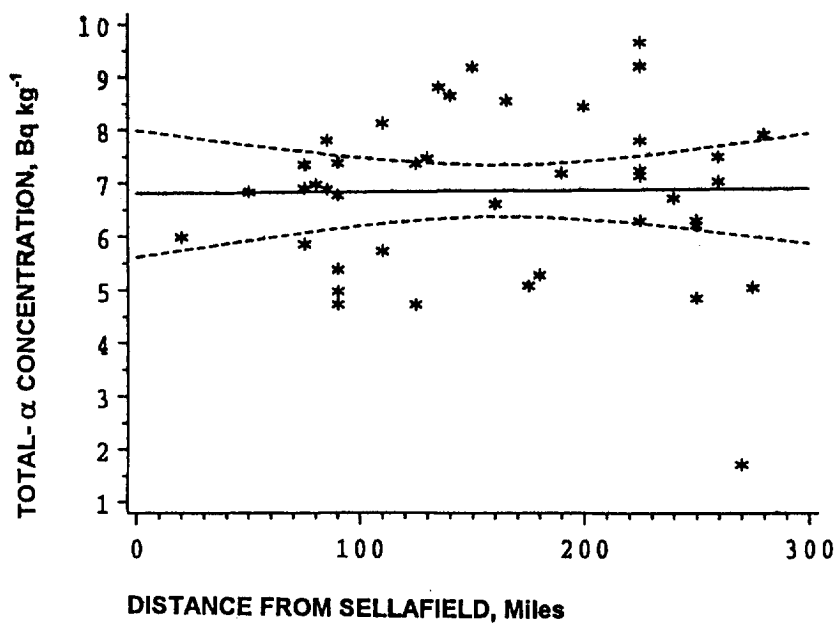


Fig. 3. Total α -activity of children's teeth vs. distance from Sellafield. Regression equation: $[\text{total } \alpha\text{-emitters}] = 6.8071 + 0.0004 \times \text{Distance from Sellafield (miles)}$. Statistical significance: $P < 0.9064$.

Table 2
Plutonium-239,240 concentration in teeth as a function of distance from Sellafield

Number of batches	Distance (miles)	Activity (mBq kg ⁻¹ ash wt.)
9	0–50	7.1 ± 4.1
34	50–150	5.0 ± 4.8
24	> 150	3.0 ± 2.7

if further studies are undertaken a more sensitive analytical technique would be of benefit. Recently, one of the authors (NDP) in association with the University of Manchester and the Australian National University, Canberra has developed accelerator mass spectrometry (AMS) for this purpose. Early results suggests that this technique is more sensitive than other mass spectrometry techniques and has a sufficiently low instrument detection limit (approx. 5×10^5 atoms equivalent to 0.5 μBq of ²³⁹Pu) to allow the measurement of the plutonium content of individual teeth (Fifield et al., 1996).

The levels of plutonium found in teeth are at the lower end of the range of the equivalent recorded bone values (Tables 1 and 3). Given the lower age (shorter period of exposure) of the children donating teeth than the adults at death,

Table 3
Comparison of the measured levels of plutonium in teeth with some levels previously reported in bone

Reference	Sample	Activity (mBq kg ⁻¹ ash wt.)	Origin of samples
Present study	Teeth	5	Great Britain and Ireland
Bunzl and Kracke, 1983	Bone	3–4	Germany
O'Donnell, 1993	Bone	3	Greenland
O'Donnell, 1993	Bone	14*	Ireland
Burkinshaw et al., 1987	Bone	5	Great Britain
Popplewell et al., 1985 Popplewell, 1986	Bone	3.5–6	Great Britain

* Hip bones from elderly patients.

from which bones were removed for analysis, our results give credence to the suggestion that plutonium levels in teeth are similar to those in bone and that teeth samples may be analysed to provide an index of variations in plutonium skeletal and hence body content. Moreover, the disparity between plutonium levels in teeth collected from West Cumbria compared with sites remote from this location is similar to that noted by Popplewell et al. (1985) (1988), who compared plutonium levels in bone collected from persons who had lived in West Cumbria with those that lived elsewhere in the UK. Popplewell, however, did not estimate the geographical extent of additional plutonium contamination resulting from Sellafield releases of this element. The present results suggest that this 'Sellafield effect' persists beyond the environs of the BNFL plant and the plutonium-contaminated coastal shores of West Cumbria — this finding is consistent with an aerial source of plutonium (Popplewell et al., 1989).

Despite the above, the results of the present study do not suggest that plutonium released by BNFL presents a significant additional radiation-induced cancer risk to the exposed population. This may be deduced from the very low levels of plutonium recorded in the teeth compared with their total α -activity — given that α -particles of all energies are of similar 'toxicity'. However, caution should be exercised as the extent of this disparity may not be as great as indicated by the ratio total- α activity:plutonium activity. This is because Henshaw et al. (1994) have shown that the spatial distribution of α -activity in teeth is highly non-uniform with a marked tendency for ²¹⁰Pb-supported ²¹⁰Po to concentrate in outer enamel, with little or no ²²⁶Ra present, while ²²⁶Ra itself concentrates preferentially around the pulp cavity of teeth. Nevertheless, it has been shown (Lovaas and Hursh, 1968) that in terms of calcium concentration, ²¹⁰Pb levels in teeth are highly correlated with those in bone at representative skeletal sites. Similarly for ²²⁶Ra, Yamamoto et al. (1994) have shown levels in teeth to be very similar (within a factor of two) to those found in bone samples removed from the same donors. Thus, the measured levels of ²¹⁰Po and ²²⁶Ra in teeth, the 500 mBq kg⁻¹ ash wt. of ²³²Th

and uranium isotopes also found (L. Strange, AEA Technology, Harwell, unpublished data), and the higher levels of strontium-90 (which, although a β -emitter, does irradiate bone), if representative of bone levels, show that the dose/risk increment to the skeleton from the few extra mBq of plutonium present is insignificant. This is particularly clear in the case of thorium which is distributed in bone with the same pattern, and hence irradiates the same cells as plutonium (Priest, 1990). A similar conclusion was reached by Taylor (1995) who calculated the radiation doses to bone resulting from environmental radionuclides (other than from ^{210}Po). His results suggest that the skeletal dose arising from plutonium isotopes is approx. 3% of that produced by radium, thorium and uranium isotopes.

The noted lack of correlation between ^{90}Sr levels and total α -emitter levels in teeth with distance from Sellafield was predicted. In the case of total- α , all the important radionuclides of lead (polonium), uranium and thorium are natural radionuclides, most of which (other than a small fraction of the total uranium) have no association with discharges from Sellafield — although other correlations with proximity to major roads and estuaries have been noted in the case of $^{210}\text{Pb}/^{210}\text{Po}$ (Henshaw et al., 1995). In the case of ^{90}Sr , which is released in the waste-stream of the Sellafield plant, it is likely that no correlation exists because the major route of uptake of this radionuclide is from milk, most of which is redistributed within the UK and mixed before consumption by the public.

In conclusion, the results of the present study clearly indicate that the BNFL Sellafield nuclear fuel reprocessing plant does contribute to plutonium body-burdens in the British population. However, the levels of this element seem too low to provide a significant increment in the risk of induced skeletal (including leukaemia) tumours above that arising from the intake of natural radionuclides.

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