Review of Autopsy, In Vivo and Bioassay Measurements on Members of the Public in the UK

G J Ham, S A Hodgson, M J Youngman, G Etherington, and G N Stradling

ABSTRACT

This report reviews for the UK population, experimental data on whole body or partial body monitoring, urine assay, the assay of teeth, autopsy and fetal data for members of the public who have not been exposed occupationally to radionuclides. Most attention has been devoted to artificial radionuclides formed in the nuclear fuel cycle, in particular isotopes of plutonium, americium, caesium, strontium and iodine. Studies concerning enhanced levels of polonium have been included. Measurements of uranium and thorium have also been addressed since they provide important baseline data for making judgements on public exposures as a consequence of accidental releases from nuclear or commercial facilities.

The report targets principally persons who live, or have lived, in the vicinity of nuclear sites. However, it also includes data from individuals resident in other parts of the UK, as well as measurements made in the aftermath of the Chernobyl accident. When UK data are unavailable, information obtained from studies in other countries has been included.

All identified measurements made since 1957, the year of the Windscale accident, to the present day have been considered.

It is stressed that this report should be considered as a source document which has identified accessible publications. Hence, assessments of intake or dose have not been undertaken by the authors, nor reference made to publications where others may have done so.

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Page number 14 Title of Table 2.6.1 changed from 'mBq kg^{-1}' 'to Bq kg^{-1}'

Page number 61, last line '(6 Bq g^{-1} ± 2 (SD))' changed to '(6 Bq kg^{-1} ± 2 (SD))'

EXECUTIVE SUMMARY

This report reviews for the UK population, experimental data on whole body or partial body monitoring, urine assay, the assay of teeth, autopsy and fetal data for members of the public who have not been exposed occupationally to radionuclides. Most attention has been devoted to artificial radionuclides formed in the nuclear fuel cycle, in particular isotopes of plutonium, americium, caesium, strontium and iodine. Studies concerning enhanced levels of polonium have been included. Measurements of uranium and thorium have also be addressed since they provide important baseline data for making judgements on public exposures as a consequence of accidental releases from nuclear or commercial facilities.

The report targets persons who live, or have lived, in the vicinity of the nuclear sites. However, it also includes data from individuals resident in other parts of the UK, and includes measurements made in the aftermath of the Chernobyl accident. When UK data are unavailable, information obtained from studies in other countries has been included.

All identified measurements made since 1957, the year of the Windscale accident to the present day, have been considered.

Section 1 describes the objectives and scope of the review as outlined above.

Section 2 presents information on autopsy measurements. This includes reference to the considerable amount of data on ^{90}Sr published in the reports of the Medical Research Council. The published autopsy data on plutonium shows that the variation in tissue concentrations can be appreciable. In general, the median values for individuals who had lived in different regions of the UK were within an order of magnitude; the data were also consistent with those obtained in the US. However, the results of one study led the authors to conclude that the relatively high values in the livers of West Cumbrians may have resulted from early operations of the Sellafield Works. Owing to the lack of autopsy data on natural uranium and thorium in the UK, and hence baseline data on tissue distribution, information from detailed studies in the United States has been included. Measurements on lead and plutonium isotopes in tissues from UK residents were in good agreement with US data. There is only a paucity of data for caesium and iodine isotopes.

Section 3 reviews fetal and placental tissue measurements. Plutonium-239 has not been positively identified and the reported values essentially reflect the considerable variability in the limits of detection achieved, by mass or alpha spectrometry. The amount of plutonium reaching the fetus is therefore extremely small, even 5 mg of house dust would contain more ^{239}Pu than the reported limits of detection. For ^{238}U and ^{232}Th, the ranges of concentration were similar in the fetus and placenta; there appeared to be no evidence for a placental barrier. As expected, ^{210}Po and ^{210}Pb show wide variations in fetal and placental samples. There appeared to be no correlation between the concentrations of the elements within the same sample, but some correlation occurred with fetal age. No published work was identified for isotopes of strontium, caesium or iodine.

Section 4 is concerned with *in-vivo* measurements. Results for Southern England, Northern England, Scotland and Wales are described separately, mainly as a

consequence of the regional variations which occurred after the Chernobyl accident. In Southern England, extensive measurements have been made for ^{137}Cs and to a lesser extent for ^{134}Cs since the 1950's. Interestingly, in 1957 the levels for residents near Windscale were about three times those for residents of Oxfordshire and Berkshire; after the Windscale accident these levels increased about four-fold. Until the Chernobyl accident, the trend was steadily downwards. After the Chernobyl accident, the amounts in some individuals increased about 20-fold, but have now returned to pre-Chernobyl values. In Northern England, Scotland and Wales the amounts of ^{137}Cs measured in the body after Chernobyl were consistently higher than in the South reflecting the prevailing weather patterns and rainfall. In broad terms, regional variations in the ^{131}I contents of the thyroid in the immediate aftermath of the Chernobyl accident were similar to those described for ^{137}Cs. In 1990/1991 measurements on ^{241}Am in the skull were made on 66 residents of Caithness who lived in proximity to the Dounreay Nuclear Establishment; none was detected. It was calculated that the skeletal content was less than 28 Bq in all cases, and that previous intakes of ^{241}Am, if any, were unlikely to have increased the incidence of cancer. No in-vivo measurements of plutonium, uranium or thorium isotopes have been identified in this review.

Section 5 is concerned with the measurement of radioactivity in teeth. An advantage of measuring teeth is that they can be collected easily from orthodontists and be used as a basis for investigating regional variations in uptakes of actinides and other bone seeking radionuclides. In some studies the data for ^{90}Sr correlated with global fall-out. Concentrations of ^{239}Pu in teeth can vary widely. Whilst those in the vicinity of Sellafield tend to higher than for most regions of the UK, they are not the highest. It is unclear from the different publications whether or not there is a correlation between concentration in teeth and distance from Sellafield. No such correlation was identified for total alpha emitter concentrations. Considering low concentrations of ^{239}Pu in teeth and the reasonable consistency between ^{239}Pu concentrations in teeth and bone, it would appear that any increase in risk of leukaemia would be very small compared with intakes of naturally occurring radionuclides. Concentration measurements of ^{238}U, ^{210}Po and ^{226}Ra have also been included in this section.

Section 6 summarises urinary excretion measurements. Compared with the other sections, few studies have been reported, although this does not diminish their importance. One study on the urinary excretion rate of ^{90}Sr in adults and children living in the Dounreay area suggested that the results did not support this radionuclide as being the cause of increased childhood leukaemia. Another study involving the assay of ^{239} Pu in the urine of residents of Dounreay and from areas remote from nuclear facilities drew similar conclusions. Attempts to correlate the urinary excretion rate of ^{137}Cs with body content were unconvincing, possibly because the variations observed reflected differences in retention half-times.

In conclusion, it is stressed that this report should be considered as a source document which has identified and summarised accessible publications on measurements of radionuclides in members of the public. Hence, assessments of intake or dose have not been undertaken by the authors, nor reference made to publications where others may have done so.

CONTENTS

1 INTRODUCTION

The purpose of this report is to review for the UK population, experimental data on whole body or partial body monitoring, urine assay, the assay of teeth, autopsy and fetal data for members of the public who have not been exposed occupationally to radionuclides. Most attention has been devoted to artificial radionuclides formed in the nuclear fuel cycle, in particular isotopes of plutonium, americium, caesium, strontium and iodine. In addition studies concerning enhanced levels of polonium have been included. Measurements of uranium and thorium have also be addressed since they provide important baseline data for making judgements on public exposures as a consequence of accidental releases from nuclear or commercial facilities.

The report is targeted principally at persons who live, or have lived, in the vicinity of the nuclear sites at Sellafield, Springfields and Dounreay. However it also includes data from individuals resident in other parts of the UK, including measurements made in the aftermath of the Chernobyl accident. When UK data are unavailable, information obtained from studies in other countries has been included.

The report has considered all identified measurements made since 1957, the year of the Windscale accident, to the present day.

2 AUTOPSY MEASUREMENTS

Measurements on autopsy samples present administrative and legal difficulties. The legal requirement in the UK is that unless the measurements are to determine the cause of death, specific permission must be given before samples are taken. This results in only limited data being available, particularly for non-occupationally exposed persons. Moreover, autopsy measurements are also usually limited to radionuclides which have relatively long biological and physical half lives, so that the measurements after death can be used to estimate lifetime exposure.

Due to the relative scarcity of UK data on measurements of radionuclides in non-exposed human autopsy samples, data from the USA have been included in this review. The one exception to this is for ^{90}Sr where there is an extensive UK database.

Except for caesium and uranium isotopes all values quoted in this review have been converted to mBq kg^{-1}. Caesium isotopes are reported as mBq g^{-1} due to their higher concentration in body tissues, and uranium values are given as µg kg^{-1} due to its low specific activity. All measurements are relative to mass of wet tissue.

2.1 Strontium isotopes

2.1.1 Introduction

Strontium is a close analogue of calcium, and as such it is well known to concentrate in bone. Hence, this has been the organ of interest for virtually all studies of ^{90}Sr in autopsy samples.

2.1.2 Literature review

Strontium-90 was regularly measured in human bone samples between about 1955 and 1970. The Medical Research Council published yearly monitoring reports from 1959 to 1970. The last report (MRC, 1973) not only gave the measurements for that year but summarised the data from 1959 to 1970.

Figure 2.1.1 summarises the MRC data graphically. They clearly demonstrate that the peak activity measured in 1964-66 was attributable to the 1961-62 atmospheric weapon tests carried out by the USSR and USA. Following that peak, the measured concentrations have steadily decreased. In 1970, it was decided by the MRC that routine monitoring of human bone samples was not cost effective because the monitoring of air, vegetation and milk was sufficient to estimate the levels in humans.

There has been a considerable amount of work undertaken to evaluate the relationship between strontium in diet and the concentration in bones. Mole (1965) published work on the geographical and age distribution of stable strontium in comparison with diet. He concluded that the strontium content of bone (measured as the Sr:Ca ratio) varied by over a factor of two within the UK but that extreme values were found in locations as close as 40 miles apart. He also concluded that there was little turnover of strontium in adult bone, so that the adult concentrations were strongly effected by the diet and environment during childhood. He noted that even by the mid-1960's the food distribution system made it very difficult to determine local food concentrations. Similar conclusions on the importance of the first few years of life were drawn by Bryant and Loutit (1964). The authors reviewed measurement data from 1955 to 1961. They concluded that in the first year of life, the discrimination between strontium and calcium found in adults was not fully developed. They also suggested that the annual rate of replacement of bone in adults ranged from 2% for long bones to 8% for vertebrae. A study of the increased concentrations due to the weapon tests in 1961 and 1962 (Fletcher et al, 1966), concluded that the maximum concentration within any age group occurred in the first year of life, and that the maximum was reached at about 4 months. The authors also noted that fractional loss of ^{90}Sr was about 0.5 of the body content per year until the age of 8, falling to 0.1 to 0.2 in adolescence. They calculated that the fractional retention of dietary ^{90}Sr was about 0.07 per year throughout childhood and adolescence.

Two papers describe both the distribution of ^{90}Sr in bone and the uptake from diet (Papworth and Vennart, 1973; Bryant and Loutit, 1964). They show that the ^{90}Sr concentration in bone can be related to the concentration in diet. The historical data as shown in Figure 2.1.1 closely mirror the reported concentrations measured in diet shown in Figure 2.1.2, confirming the correlation.

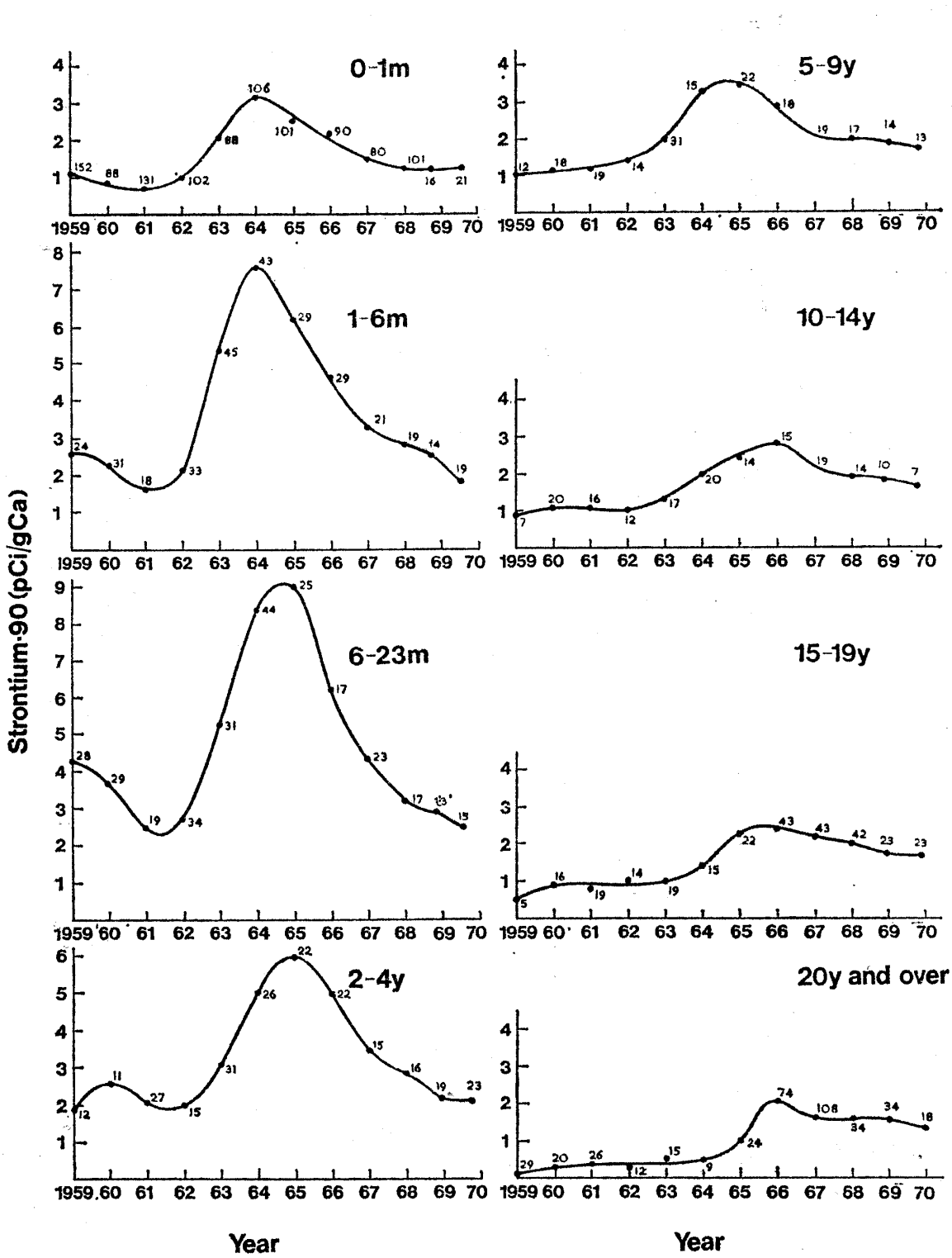

FIGURE 2.1.1 Strontium in Human Bone 1959-1970. The figures adjacent to the dots indicate the number of samples on which means are based.

(Reproduced from MRC, 1973) NB. 1 pCi = 37 mBq

2.1.3 Summary

In summary there are considerable historical data published in MRC reports. This monitoring programme ceased in 1970, but sufficient evidence had been gathered to allow empirical models relating bone concentration to dietary intake to be developed. The continuing decline in the published data on ^{90}Sr in diet confirmed the expectation in the 1970 MRC report that ^{90}Sr concentrations in human bone would continue to fall. The latest published measurement of ^{90}Sr in milk (Hammond et al, 2001) is equivalent to a maximum of 20 mBq g^{-1} Ca. This is approximately a quarter of the final value shown in Figure 2.1.2.

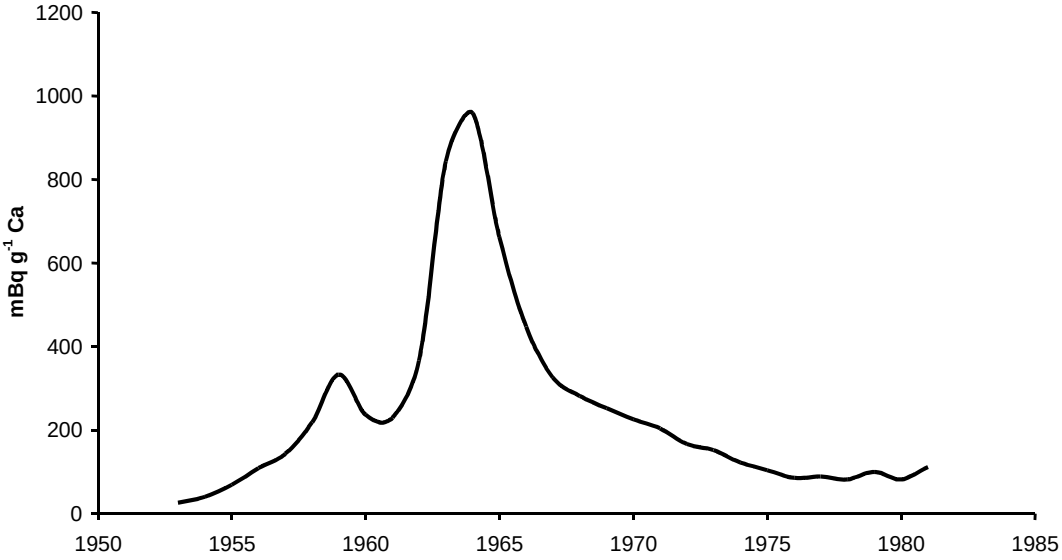

FIGURE 2.1.2 Strontium-90 in the average UK diet.

Data from Papworth and Vennart (1984).

2.2 Caesium isotopes

2.2.1 Introduction

There are very few data published on measurements of caesium concentration in autopsy tissue from members of the public. This is probably because caesium isotopes are easily measured by direct external counting of the living person, and they have relatively short biological, and for ^{134}Cs physical, half-lives.

2.2.2 Literature review

Only two publications reporting caesium measurements were identified. Both measured caesium isotopes in tissues originally collected for plutonium measurement. Ide and McInroy (1975) published ^{134}Cs measurements on liver

and lung samples from throughout the USA with median values of 5.7 and 3 mBq g^{-1} respectively. Similarly, Popplewell et al (1988) published ^{137}Cs measurements in liver. These are summarised in Table 2.2.1. It should be noted that the original publication had a typographical error; the values are in mBq g^{-1} and not mBq kg^{-1} (private communication from author).

TABLE 2.2.1 137Cs Content of Liver samples (Dates of death 1980-1984), mBq g^{-1}

Location	Oxfordshire	Central Scotland	W Cumbria
Median	0.3	0.6	1.2
Range	0.2 - 0.7	0.2 - 1.1	0.2 - 7.6
No in Group	14	14	15

(Popplewell et al, 1988)

2.2.3 Summary

The value of autopsy measurements for caesium is limited. The relatively rapid decrease in Cs body content after Chernobyl (Section 4.1.2) shows that the element is not retained in the body for a long time. The short half-life of ^{134}Cs (2 y) means that autopsy measurements can not be used as a measure of exposure during life. Whilst Popplewell et al made no comment on their results, the higher median value for persons in West Cumbria is possibly due to the increased rainfall in that area, although some contribution from Sellafield marine discharges can not be ruled out.

2.3 Iodine isotopes

2.3.1 Introduction

Very few data have been published on radio-iodine measurements in autopsy tissues. Again this is probably because the commonly occurring radioisotopes are easily measured in vivo, and have short physical half-lives, while iodine also has a short biological half-life.

2.3.2 Literature review

Three papers have been identified in this review. A study by Visalli (1964) measured the short-lived ^{131}I ($T_{½}$ 8 days) in thyroid samples from children in New England. Most measurements made before September 1962 were below the limit of detection. There was a cluster of positive results following atmospheric weapon tests in September and October 1962. As expected, the number of positive results then rapidly decreased. A study by Bowlt (1987) measured the ^{131}I content of thyroids from adults in Britain who died during the 9 weeks after the Chernobyl accident. Peak specific activities were up to 5 Bq g^{-1} dry weight, which then fell to about 0.3 Bq g^{-1} with a half-life of 11–16 days. A further study by Bowlt (1989) investigated the ^{129}I content of thyroids in members of the public in Cumbria. He found a relationship between the distance from Sellafield

and the ^{129}I content, although there was considerable variation in the data for similar distances. All activities were low, most between 0.1 and 0.2 Bq g^{-1} dry mass. The same study could find no evidence of an increase in the incidence of thyroid cancer.

2.3.3 Summary

Due to the short half-life of ^{131}I and the short biological half-life of all iodine isotopes, the published data are of little relevance to the current UK situation. These short half-lives also limit the usefulness of autopsy data for this element.

2.4 Plutonium isotopes

2.4.1 Introduction

Measurement of plutonium in non-occupationally exposed autopsy tissue presents many difficulties. Concentrations are low and so there are likely to be significant uncertainties on the measured concentrations. Where individual data are reported, it is apparent that ranges of an order of magnitude or more in the measured concentrations are typical. The authors of this report have identified no published work since the early 1990's. This is possibly due to the large sample sizes needed and the difficulty of obtaining permission to collect samples.

2.4.2 Literature review

Table 2.4.1 lists the published median values for selected organs. Only three publications (Osborne, 1963b; Bahreyni-Toossi, 1982; Popplewell et al, 1988) have reported measurements of plutonium in autopsy samples from the UK. Osborne compared the concentration in lung samples with a value calculated from the concentration in atmospheric dust using ICRP (International Commission on Radiological Protection) models. The agreement was good, although many parameters were estimated rather than measured. The lung concentrations measured by Bahreyni-Tossi and Popplewell are a little lower than those measured by Osborne but considering the range of individual results, all the UK measurements are in reasonable agreement with each other and with the US data in Table 2.4.1. The work published by Bahreyni-Toossi (1982) is also the subject of a paper by Burkinshaw et al, (1987). In the only paper found by this review investigating ^{241}Pu in autopsy samples, (Bunzl et al, 1987) measured the decay product ^{241}Am in livers from the Federal Republic of Germany. They compared the measured ^{241}Am and $^{239+240}Pu$ concentrations, and found a very good correlation with a mean Am:Pu ratio of 0.133. They also calculated expected concentrations using an assumed ^{241}Pu ingestion and the ICRP Publication 30 retention model (ICRP, 1979). The measured values were within the range of those calculated. However the calculated best estimate was about twice the measured mean, as was the Am:Pu ratio.

TABLE 2.4.1 Median (and range, where given) of $^{239+240}$Pu concentrations in mBq kg^{-1}

Place and date of collection	Lung	TBLN*	Liver	Vertebrae	Reference
S England, 1962	6 (4-8)				Osborne, 1963b
Yorkshire, 1982	2 (0.1-4)		14 (1-38)	8 (3-14)	Bahreyni-Toossi, 1982
Scotland, NE England and Oxfordshire, 1980-85	2 (0.4-16)		26 (5-60)	5 (2-11)	Popplewell et al, 1988
W Cumbria, 1981-85	7 (2-33)		52 (15-160)	7 (3-11)	
USA 1959-77	8	90	26	11	Fox et al, 1980
USA 1960-77	7 (0.3-116)	96 (0.5-7,400)	26 (3-157)	11 (0.7-500)	McInroy et al, 1979
New York 1973-74	2 (0.4-22)	4 (0.4-600)	14 (4-600)	10 (4-500)	Fisenne et al, 1980
Washington state, 1970-75	6 (0.5-50)	40 (0-90)	31 (0-130)	7 (0-220)	Nelson et al, 1993

* Tracheo-bronchial lymph nodes

Popplewell found a small but significant increase in the concentrations measured in samples from West Cumbria compared to those from the rest of the UK. This contrasts with work in Colorado (Ibrahim et al, 2002) which found no statistically significant relationship between distance from the Rocky Flats works and plutonium bone concentration (Ibrahim et al, 2002). In a later paper, mass spectrometry was used to measure the $^{239+240}$Pu and $^{241+239}$Pu ratios in samples from both Oxfordshire and Cumbria (Popplewell et al, 1989). The ratios from the Oxfordshire samples were in good agreement with those published for worldwide fall-out. However those from the Cumbrian samples indicated that the enhanced plutonium concentrations in the livers was from a source with a low ^{240}Pu content when compared with fall-out or Sellafield liquid discharges. The authors concluded that "the enhanced plutonium concentration may be due to atmospheric discharge from the early operations on the Sellafield works". These atomic ratios compare favourably with those found in soil close to the works (McCarthy and Nicholls, 1990) and Jones' analysis of plant discharges (Jones et al, 1996). This conclusion was further discussed in an NRPB report on the risk of childhood leukaemia near nuclear installations (Stather et al, 1988).

Jones and Prosser (1997) compared worldwide autopsy data with estimated concentrations using the ICRP Human Respiratory Tract Model (ICRP, 1994) and the ICRP systemic model for plutonium (ICRP, 1993). They concluded that agreement was good, and that the biokinetic behaviour of environmental plutonium fell between that for type M and S compounds.

2.4.3 Summary

Despite the difficulty of analysis, the agreement between median values in Table 2.4.1 is good. All reported median values for liver, lung and bone are within an order of magnitude of each other. Given the large range of individual results, and with the exception of Popplewell's Cumbrian data, there appear to be no significant differences between any of the published mean values. However, the median concentrations in tracheo-bronchial lymph nodes have a larger variation and range. This is probably due to two reasons. First, the mass of tissue and hence total amount of plutonium measured is very small, and second, the selection of tissue to be included in the sample is subjective.

The comparisons of estimated and measured concentrations are good, considering the range of measured values and the assumptions made.

2.5 Uranium and thorium isotopes

2.5.1 Introduction

Uranium has been widely measured in human tissue, probably because it can be readily detected in small samples and in-vivo measurements are not sensitive to typical levels resulting from environmental exposure.

2.5.2 Literature review

Fisenne et al. (1988) published a comprehensive review of uranium in humans. Table 2.5.1 lists the UK and USA data from this review. The mean value for skeleton, 2.4 μg U kg^{-1} wet bone, agrees well with Singh's (1990) estimate of 24.9 μg U in the 10 kg of skeleton in reference man. However these values are between 0.5 and 0.3 of the ICRP estimate (ICRP, 1993).

TABLE 2.5.1 Uranium concentrations in human tissues and blood, μg kg^{-1} wet tissue

Location	Lung	Liver	Kidney	Blood	Bone	Bone type
England		0.25		0.69± 0.45	3.5 ± 0.9	multiple
USA						
New York	0.5 ± 0.4	0.2 ± 0.3	0.4 ± 0.3	1.4 ± 0.09	0.3 ± 0.2	vertebrae
Illinois				0.1	0.3 ± 0.3	unknown
5 regions					0.8 ± 0.6	vertebrae
Pennsylvania	1.17	0.12	0.39		0.39	vertebrae
					0.67	rib
Utah	1.02	0.33	0.90		0.61	vertebrae
					1.7	rib
Colorado	1.02	0.30	1.00		0.92	vertebrae
					1.9	rib
Wisconsin					1.2 ± 0.3	skeleton
World						
Mean ± SD	2.5 ± 2.5	2.3 ± 4.0	2.3 ± 2.3	0.58 ± 0.44	2.4 ± 2.3	
Range	0.5 - 6.8	0.12 - 11	0.34 - 5.5	0.1 - 1.3	0.10 - 7.1	

Taken from Table 1 of Fisenne et al. (1988)

The distribution of uranium between vertebrae, ribs and long bones was investigated by Harley and Fisenne (1990). The authors concluded that there were no significant differences in the concentrations of uranium in the three bone types. The mean concentration of the uranium isotopes measured agrees with those of Fisenne et al (1988) and Singh (1990). The uranium concentrations of tissues from 27 New York City residents were measured in 1985 (Fisenne and Welford, 1986). These results were later summarised in the 1988 review. However, the 1986 paper showed a correlation between uranium concentration and age at death for both lung and vertebrae but not for kidney and liver. While the lung and liver contents were similar to those reported by ICRP (1974) the kidney and skeleton contained significantly less uranium than the ICRP value. These differences between ICRP and measured values also occur in two United States Transuranium and Uranium Registries (USTUR) cases (Kathren, 1997). These were two whole body donations to the Registry; the individuals were known to be exposed to plutonium but not uranium. The tissue distributions for uranium measured for these cases are detailed in Tables 2.5.2 to 2.5.5. The total skeletal contents of 45 and 34 μg U are similar to Singh's estimate of 24.9 μg, confirming that there was little or no occupational exposure.

TABLE 2.5.2 Skeletal distribution of uranium in USTUR Case 0242

Tissue / Organ	Autopsy weight (g)	Uranium content (μg)	Tissue / Organ	Autopsy weight (g)	Uranium content (μg)
Skull and teeth	638	3.130	Sternum	118	NA
Mandible	57	0.293	Humeri	605	2.220
Hyoid	4.2	0.0185	Radii	177	1.520
Cervical spine	152	0.386	Ulnae	209	1.030
Thoracic spine	579	1.180	Carpals *	184	1.840
Lumbar spine	518	1.012	Femora	1810	8.720
Sacrum and coccyx	357	0.669	Tibiae	1090	5.050
Pelvis	1020	3.850	Fibulae	209	4.220
Clavicles	97.6	NA	Patellae	68.6	0.298
Scapulae	285	NA	Tarsals †	553	3.200
Ribs	615	2.280			
Total analysed	8790	40.900	(Total skeleton	9670	≡ 45.0)

NA, not analysed

* Including metacarpals and hand phalanges

† Including metatarsals and foot phalanges.

(Data from Kathren (1997) and personal communication R E Filipy, USTUR)

TABLE 2.5.3 Whole body distribution of uranium in USTUR Case 0242

Tissue/organ	Autopsy weight (g)	U content (µg)	% of systemic
Lungs inc TBLN	1440	1.96	
Pulmonary0 LN	11.4	0.513	
Liver	1620	0.2	0.4
Kidneys	312	0.302	0.5
Spleen	205	0.133	0.2
Testes	23.5	0.0438	0.1
Muscle	43893	8.919	15.9
Skin	20000	0.716	1.3
Other soft tissue	2730	0.866	1.5
Bones and teeth	9670	45	80.1
Total		58.65	
Total Systemic		56.18	

(Data from Kathren (1997) and personal communication R E Filipy, USTUR)

TABLE 2.5.4 Skeletal distribution of uranium in USTUR Case 0213

Tissue / Organ	Autopsy weight (g)	Uranium content (µg)	Tissue / Organ	Autopsy weight (g)	Uranium content (µg)
Skull and teeth	770	3.070	Sternum	131	0.209
Mandible	85.8	0.300	Humeri	514	2.050
Hyoid		NA	Radii	149	0.683
Cervical spine	161	0.738	Ulnae (shaft)	92.2	0.597
Thoracic spine	472	1.670	Carpals *	221	2.180
Lumbar spine	419	1.210	Femora	1560	NA
Sacrum and coccyx	172	0.329	Tibiae	969	2.820
Pelvis	242 ‡	0.939	Fibulae	209	1.100
Clavicles	75	0.420	Patellae	74	0.347
Scapulae	283	0.901	Tarsals †		NA
Ribs	531	2.260			
Total analysed	5570	21.800	(Total skeleton	8740	≡ 34.2)

NA, not analysed
* Including metacarpals and hand phalanges
† Including metatarsals and foot phalanges
‡ Ischium only.
(Data from Kathren (1997) and personal communication R E Filipy, USTUR)

TABLE 2.5.5 Whole body distribution of uranium in USTUR Case 0213

Tissue/organ	Autopsy weight (g)	U content (µg)	% of systemic
Lungs inc TBLN	1690	1.4	
Pulmonary LN	10.8	0.236	
Liver	2600	0.206	0.4
Kidneys	670	0.627	1.1
Spleen	236	0.094	0.2
Testes		NA	
Muscle	43893	15.163	26.4
Skin	12700	6.089	10.6
Other soft tissue	856	0.834	1.5
Bones and teeth	8740	34.2	59.5
Central nervous system	1110	0.246	0.4
Total		59.095	
Total Systemic		57.459	

NA, not analysed
(Data from Kathren (1997) and personal communication R E Filipy, USTUR)

Table 2.5.6 summarises the UK and USA data found for thorium isotopes in non-exposed subjects. Some of the measurements were reported as activity concentration in bone ash or mass of natural thorium in bone ash. For these cases the listed values have been converted to mBq kg^{-1} wet mass using ICRP reference man (ICRP, 1974) values of 10 kg wet bone ≡ 2.8 kg of ash and the specific activity of ^{232}Th (1 ng ≡ 4.10^{-6} Bq).

TABLE 2.5.6 Thorium concentrations in human tissue, mBq kg^{-1}

		Lung	TBLN	Liver	Kidney	Bone	Testes	Spleen	Thyroid
London (Hill, 1962)	^{232}Th	20-100							
UK (Clifton et al, 1971)	^{232}Th					34 ± 14			
USA (Lucas et al, 1970)	^{232}Th					<0.1 - 80*			
Colorado (Singh et al, 1982)	^{228}Th	10	189	3	3	20	1	2	12
	^{230}Th	31	407	6	11	34	2	5	30
	^{232}Th	21	289	1	3	6	2	3	24
Washington (Singh et al, 1982)	^{228}Th	9	96	3	3	22			
	^{230}Th	11	170	6	6	12			
	^{232}Th	12	104	2	1	4			
USA (Harley and Fisenne, 1990)	^{230}Th					17†			
	^{232}Th					11†			
Hanford (Glover et al, 2001)	^{232}Th	69±2	490±20	0.29±0.01	0.76± 0.04	9.86±0.09	<0.9	0.82±0.08	

* Values originally presented as ng g^{-1} bone ash, converted to activity (1 ng ≡ 4.10^{-6} Bq) and then to mBq kg^{-1} as above.

† Values originally presented in kg^{-1} bone ash, converted to wet tissue assuming 1 kg tissue ≡ 0.28 kg ash (ICRP, 1974).

In the UK, Hill (1962) measured ^{232}Th concentrations in the lungs of nine "normal subjects" from London by alpha spectrometry. He reported concentrations ranging from 74 to 370 mBq kg^{-1} of wet tissue. The only other paper identified which reports UK data (Clifton et al, 1971) measured ^{232}Th concentration in rib bones of twelve subjects. The mean value was 34 ± 14 mBq kg^{-1} of wet tissue with a range of 1 to 190 mBq kg^{-1}.

There have been some more extensive studies performed in the USA. Lucas et al (1970) published results for thorium in rib bone from 38 subjects. The reported concentrations ranged from <0.1 to 80 mBq kg^{-1}. The highest value was from an individual known to have obtained water from a supply with a high ^{226}Ra content, and presumably a high thorium content. Excluding this case, the upper bound of the range was 50 mBq kg^{-1}. The authors also noted a correlation of thorium concentration with age at death. The best fit line had the formula $Y = (0.16 \pm 0.02)T$, where Y is the thorium concentration in ng g^{-1} bone ash and T is the age in years. The other (Singh et al, 1982) large study performed in the USA measured the thorium concentration in tissues from 32 people, comparing Colorado with Washington DC residents. In this study, the bone sample was vertebra rather than

rib. The concentration range reported for bone (0–12 mBq kg^{-1}) is lower than that reported by Lucas. Again, a correlation of thorium concentration with age was noted; the difference between the two populations was mainly due to the different average ages. A smaller study on the assay of three skeletons was published by Harley et al (1990). The samples had been collected in the 1960's in order to study strontium and the remaining tissue was analysed for uranium and thorium. This study found no significant difference in the thorium concentrations of ribs, vertebrae and long bones. Similarly a study of a whole body donor to the USTUR (Glover et al, 2001), detailed in Tables 2.5.7 and 2.5.8, found similar thorium concentrations in spine and ribs. The subject was known to be exposed to plutonium and americium during his lifetime but had no known exposure to thorium. The study found about 70% of the total thorium divided equally between the lungs and bones with most of the remainder in muscle and skin. Less than 0.5% of the thorium was present in liver and kidney. The measured concentrations were compared with those predicted by the ICRP (1979; 1995) and the fraction of total thorium found in the liver was even lower than that predicted while that in soft tissues was considerably higher.

TABLE 2.5.7 Skeletal distribution of ^{232}Th

Tissue	Wet weight (g)	Activity (mBq)	% skeletal activity
Skull and teeth	972.6	11	10.36
Mandible	105.5	0.81	0.76
Hyoid	4.3	-	-
Cervical spine	174.4	3.2	3.01
Thoracic spine	616.6	8.79	8.28
Lumbar spine	501.4	4.88	4.60
Sacrum and coccyx	242.7	1.56	1.47
Pelvis	1189	10.3	9.70
Clavicles	109.4	0.93	0.88
Scapulae	315.3	2.53	2.38
Ribs	727.9	12.8	12.06
Sternum	136.1	0.9	0.85
Costal cartilage	237.8	0.6	0.57
Humerii	676.4	5.1	4.80
Radii	194.7	1.27	1.20
Ulnae	218.2	1.77	1.67
Carpals	83.4	1.63	1.54
Metacarpals	127.8	1.49	1.40
Hand phalanges	106.7	0.68	0.64
Femora	1824	15	14.13
Tibiae	1106	6.3	5.93
Fibulae	229.1	2.4	2.26
Patellae	82.6	0.65	0.61
Tarsals	560.8	7.8	7.35
Metatarsals	167.0	2.3	2.17
Foot phalanges	61.3	1.46	1.38
Totals	10771	106.15	100

(Data from Glover et al, 2001)

TABLE 2.5.8 Distribution of ^{232}Th in the tissues from USTUR case 0212

Tissue	Wet weight (g)	Activity (mBq)	% of systemic	Concentration (mBq g^{-1} wet wt)
Respiratory tract	1640	121 ± 3.6		74 ± 2
Lungs	1558	108 ± 3.5		69 ± 9
Trachea	56.0	0.30 ± 0.02		5.4 ± 0.4
TBLN	25.9	12.8 ± 0.6		490 ± 20
Liver	2382	0.68 ± 0.03	0.36	0.29 ± 0.01
Kidneys	461	0.35 ± 0.02	0.19	0.76 ± 0.04
Spleen	243	0.20 ± 0.02	0.11	0.82 ± 0.08
Smooth muscle *	5407	2.39 ± 0.08	1.26	0.44 ± 0.01
Striated muscle	33896	48.0 ± 0.9	25.4	1.42 ± 0.03
Misc. muscle †	626	0.8 ± 0.1	0.42	1.3 ± 0.2
Skin with fat	23537	26.0 ± 0.8	13.7	1.10 ± 0.03
Cent. Nerv. System	936	1.02 ± 0.06	0.54	1.09 ± 0.06
Other soft tissues ‡	1507	3.5 ± 0.1	1.85	2.32 ± 0.07
Testes	60.6	< 0.055	<0.03	<0.91
Bones and teeth	10772	106 ± 1	56.1	9.86 ± 0.06
Total	81455	310 ± 4		
Systemic		189 ± 2		

* Stomach, intestine, urinary bladder, mesentary from intestine.

† Heart (Th content estimated from total muscle concentration) and tongue.

‡ Pancreas, gall bladder, prostate, eyes, salivary glands, pituitary, miscellaneous fluids, adrenals, thyroid, ears, hair, penis, epidura, pelvic fat, larynx, scrotum and fat, oesophagus, nails.

(Data from Glover et al. (2001))

2.5.3 Summary

The reported UK values for uranium concentrations in human tissue are in good agreement with the world mean reported by Fisenne et al (1988). The skeletal and kidney burdens measured in the USA are significantly lower than those reported by the ICRP (1974).

Both uranium and thorium show a correlation between skeletal content and age. They are also likely to be affected by the differing concentrations of these elements in the local environment and hence the diet. For thorium, the range of reported mean concentrations in bone by different authors is within an order of magnitude. Considering the problem of age dependence and the differences in dietary intake in different geographical areas, agreement to within one order of magnitude is good.

2.6 Lead and polonium isotopes

Lead-210 and polonium-210 are in the same decay chain, of which the original parent is uranium-238:

$$^{210}\mathrm{Pb}\left(T_{1/2}=22\,\mathrm{y}\right) \rightarrow {}^{210}\mathrm{Bi}\left(T_{1/2}=5\,\mathrm{d}\right) \rightarrow {}^{210}\mathrm{Po}\left(T_{1/2}=138\,\mathrm{d}\right) \rightarrow {}^{206}\mathrm{Pb}$$

Lead-210 can be measured directly. However, it is more usually quantified by measuring the ^{210}Po to which it decays. The ^{210}Po concentration is then calculated

either by assuming that the decay chain is in equilibrium or by removing all ^{210}Po and making a measurement after a known ingrowth time. There is some confusion in the literature over these measurements, although authors have reported measurements as being for either one or the other radionuclide, in all cases the actual measurement was of ^{210}Po. Some authors confirmed that sufficient time has elapsed since death to allow equilibrium, while others have assumed that the two are in equilibrium at death. In Table 2.6.1 measurements have only been listed for ^{210}Pb where the authors have confirmed that equilibrium between the two elements had occurred before the polonium measurement was made.

TABLE 2.6.1 Lead-210 and polonium-210 concentrations in human tissue, Bq kg^{-1}

Bone	Liver	Kidneys	Lung	Sample country and type	Reference
^{210}Pb					
	0.34±0.04	0.159±0.002	0.18±0.05	USA, adults	Blanchard, 1967
1.3±0.6	0.4±0.2	0.3±0.1		UK, adults	Bradley and Fry, 1989
^{210}Po					
1.5 (0.4-0.5)*	0.4 (0.2-0.7)			USA, adults	Holtzman, 1963
0.63	0.37	0.26	0.11.	UK, adults	Osborne, 1963a
	0.54±0.07	0.42±0.09	0.19±0.05	USA, adults	Blanchard, 1967
1.5±0.1				UK, adults	Henshaw et al, 1988
0.33±0.03	0.11±0.02		0.11±0.03	UK, children	Henshaw et al, 1988

*Values originally presented in g^{-1} bone ash, converted to wet tissue using the bone:ash ratio from the original paper.

2.6.1 Literature review

One of the most extensive studies was performed by Holtzman (1963). He measured the ^{210}Po concentration in bones for 128 subjects, nearly all being adults (Table 2.6.1). In a previous report Holtzman (1960) explored the possibility of a correlation between the measured concentration and age at death, but found no such correlation. He estimated bone concentrations from radium in air and in food and concluded that the measured and estimated concentrations were in good agreement. In the same year measurements on samples from the London area were published (Osborne, 1963a). Although the authors describe the results as preliminary, and only 9 bone and 2-4 soft tissue samples were analysed, the data are in good agreement with Holtzman's more extensive study. In 1967 Blanchard published measurements for both ^{210}Pb and ^{210}Po in soft tissues for persons resident in the USA. The values for polonium were in good agreement with those previously discussed; however equilibrium between ^{210}Pb and ^{210}Po was not evident. The average ^{210}Po:^{210}Pb ratio ranged from 3 for kidney to 0.7 for thyroid. While the ratio in individual samples covered a large range, the differences in Po : Pb ratio are significant, with unsupported ^{210}Po present in kidneys and liver.

Henshaw et al (1988) published data from autoradiographs on UK bone samples from both adults and children. The measured ^{210}Po concentrations were in good agreement with those previously discussed. The measurement technique provided data on the spatial distribution of the radioactivity within the bone and its surrounding tissue. The authors concluded that the polonium produced by decay from the lead and bismuth precursors was released from the bone and was free to migrate into the marrow and surrounding tissue. They speculated that this could give rise to a higher than expected radiation dose to the sensitive red bone marrow. This contrasts with the behaviour of plutonium, which is known to be retained in the bone structure.

The largest UK study measured the ^{210}Pb in tissue samples from 17 subjects (Bradley and Fry, 1989). The authors' noted that there was sufficient time delay from death to analysis to ensure that all the ^{210}Po measured was from the ^{210}Pb present in the sample at death (Table 2.6.1). The measured concentrations were in good agreement with other published values. They included the result of a single measurement on bone marrow which, because of its low value, appears to contradict Henshaw's theory on the mobility of polonium. They compared their measurements with concentrations estimated from average dietary intake and metabolic models (ICRP, 1979; Nuclear Energy Agency, 1988) and achieved an agreement within a factor of 2 for bone and better for soft tissues. From this they concluded that estimates from dietary intake and metabolic models are reasonable.

Cohen et al. (1979) investigated the effects of cigarette smoking on the concentration of ^{210}Po in lungs. They concluded that lungs from smokers contained a significantly higher concentration of ^{210}Po. In 1998 a paper was published investigating the relative importance of inhalation and ingestion as sources of ^{210}Pb (Salmon et al, 1998). The authors suggest that while dietary intake is higher than that from inhalation, the fractional uptake to blood is higher after inhalation. They concluded that, because it is very difficult to estimate accurately the concentration in the air and because of the possible effects of smoking, they could not conclude that inhalation is insignificant. Nevertheless they estimate that as an international average the contributions to ^{210}Pb uptake were 12% from atmospheric ^{210}Pb, 2%, from radon and 86% from dietary ^{210}Pb.

2.6.2 Summary

Overall the measurements both in the UK and the USA are in reasonable agreement with each other. In most cases it is unclear whether the measurements were of supported (in equilibrium with ^{210}Pb) or unsupported ^{210}Po. However Blanchard's work suggests that in most samples the unsupported ^{210}Po is insignificant. The theory of the mobility of the polonium in bone marrow is unproven. Henshaw's work showed the presence of ^{210}Po outside the bone surfaces. However, the measurement technique required long exposure times of thin slices of bone and Bradley's work on bone marrow appears to contradict this theory. Further research would appear to be necessary to resolve this issue.

3 FETAL TISSUE MEASUREMENTS

3.1 Introduction

In common with autopsy work, measurements on fetal tissue samples are time consuming and present administrative and legal difficulties. Moreover, radionuclide concentrations in fetal samples will, in general, be lower than in adult autopsy samples and the sample size will be smaller. This means that measurements are nearly always very close to or below the limit of detection of the techniques used. The technical and legal problems, coupled with the expense of such low-level measurements means that there are very few publications in this field.

Owing to the relative scarcity of UK data, world-wide data have been included in this review. All the values quoted have been converted to mBq kg^{-1}.

3.2 Strontium isotopes

No published work on the measurement of strontium isotopes in fetal tissues has been identified.

3.3 Caesium isotopes

No published work on the measurement of caesium isotopes in fetal tissues has been identified.

3.4 Iodine isotopes

No published work on the measurement of iodine isotopes in fetal tissues has been identified.

3.5 Plutonium isotopes

Apart from mass spectrometric measurements for ^{239}Pu all other references to ^{239}Pu in this section refer to $^{239+240}Pu$. The published measurements for ^{239}Pu in fetal tissues are summarised in Table 3.5.1.

Weiner et al (1985) published results for both fetal and placental samples. They were unable to identify ^{239}Pu (or ^{241}Am) in samples from the first trimester of pregnancy. Samples from both the second and third trimester gave results of a few mBq kg^{-1}.

The NRPB has published several papers on the measurement of ^{239}Pu in fetal tissue from the second trimester (Bradley and Prosser, 1993; Prosser et al, 1994a; 1994b). Plutonium-239 was undetectable using alpha spectrometry; with more sensitive mass spectrometric techniques the ^{239}Pu contents were still not significantly different from the reagent blanks (see Table 3.5.1). The limits of detection in these studies were one or two orders of magnitude lower than those reported by Weiner et al; the NRPB authors noted this difference but could offer no explanation. The work at NRPB continued using an even more sensitive mass spectrometer (Ham, 1999). The estimated limit of detection was 0.5 μBq per sample, but the ^{239}Pu content were still not significantly different from reagent blanks (Table 3.5.1).

TABLE 3.5.1 ^{239}Pu concentrations measured in second trimester fetal samples

Location	^{239}Pu (mBq kg^{-1})	Measurement Technique	Reference
USA	1.8 - 35	Alpha Spectrometry	Weiner et al 1985
Oxfordshire	<0.1	Alpha Spectrometry	Prosser et al 1994a
Cumbria	<0.4	Alpha Spectrometry	Prosser et al 1994a
Oxfordshire	<0.01 – 0.02	Mass Spectrometry	Prosser et al 1994a
Cumbria	<0.04	Mass Spectrometry	Prosser et al 1994a
Cumbria	<0.002 - <0.01	Mass Spectrometry	Ham, 1999

In conclusion, workers in the UK have not detected any ^{239}Pu in fetal tissue. Ham (1999) outlined the rigorous precautions needed to prevent adventitious contamination of the sample. This may explain the much higher concentrations reported by Weiner in 1985.

Placental tissue has also been analysed. This has the advantage of easier availability. However, it suffers from the obvious disadvantage of being a maternal rather than a fetal material. The NRPB has published values for the placental tissues that matched the fetal samples (Prosser et al, 1994a). They were unable to detect any activity by alpha spectrometry but reported ^{239}Pu concentrations between 0.004 – 0.09 mBq kg^{-1} in the individual placental samples, measured by mass spectrometry. In a further study two or three placentas were measured at a time in an attempt to increase sensitivity. Plutonium–239 was not detected in these samples and a limit of detection of a few tens of microbequerels was reported. The only other published work on placental tissue concerned samples from the Nenetz district of Russia near a weapons test site (Lund and Tkatchev, 1996). The reported ^{239}Pu concentrations were between 6 and 70 mBq kg^{-1}. The authors noted that their results were an order of magnitude higher than those measured by Prosser et al (1994a). However the Russian samples cannot be considered as resulting from global fall-out.

No studies measuring any other isotope of plutonium were identified.

3.5.1 Summary

The reported concentrations for ^{239}Pu in both fetal and placental tissue in the UK show considerable variability in the limits of detection. This is not unexpected given the very low concentrations in the tissues, the small sample sizes available for analysis and the different analytical methods used. It should be noted that the potential for contamination of the samples was considerable. Ham (1999) calculated that 5 mg of house dust would contain more ^{239}Pu than the reported limit of detection for these samples. Given the large variability in the measurements, it is therefore not possible to confirm or deny if the placenta acts as a barrier to plutonium.

3.6 Uranium and thorium isotopes

Only two reported studies have been identified on uranium and thorium concentrations in fetal or placental tissue. These results are summarised in Table 3.6.2.

TABLE 3.6.2 ^{238}U and ^{232}Th concentrations measured in second trimester fetal and placental samples

Location	Sample type	^{238}U, mBq kg^{-1}	^{232}Th, mBq kg^{-1}	Reference
USA	Fetal	2 – 5.3		Weiner et al, 1985
Oxfordshire	Fetal	2 – 7	0.1 – 3	Bradley and Prosser, 1993
Cumbria	Fetal	0.1 - 9	0.2 – 4	Bradley and Prosser, 1993
USA	Placental	5 - 9		Weiner et al 1985
Oxfordshire	Placental	2 - 7	<0.8 – 2	Bradley and Ewers, 1995
Cumbria	Placental	<1 - <11	<0.2 - <5	Bradley and Prosser, 1993

Weiner et al (Weiner et al, 1985) published results from fetal samples for uranium. They noted that thorium was also measurable but did not give the concentrations. Bradley has also published measurements for both radioelements in fetal and placental samples (Bradley and Prosser, 1993), (Bradley and Ewers, 1995). In all cases, there was no evidence of a placental barrier to these radioelements. The ranges of concentration were similar for both placenta and fetus. As expected, the measured concentrations for these radioelements are significantly higher than those measured for plutonium because they occur naturally.

3.7 Lead and polonium isotopes

Although lead and polonium are likely to be present at higher concentrations in fetal and placental tissues than the other radioisotopes discussed in this review, there have still been very few data published.

Bradley has reported the results of radiochemical measurements on whole fetal samples (Bradley and Ewers, 1995) which are summarised in Table 3.7.1. In all but one case the polonium concentration in the placenta was larger than that in the associated fetus. The situation was less clear for lead. The ratio of fetal to placental concentration ranged from 0.4 to 4. There seemed to be no correlation between the lead and polonium concentrations in the same sample, but some correlation occurred between both lead and polonium concentration with fetal age.

TABLE 3.7.1 ^{210}Pb and ^{210}Po concentrations measured in second trimester fetal and placental samples

Location	Sample type	^{210}Pb, mBq kg^{-1}	^{210}Po, mBq kg^{-1}	Reference
Oxfordshire	Fetal	<7 - 40	5 – 60	Bradley and Prosser, 1993
Cumbria	Fetal	<2 - 40	4 – 40	Bradley and Prosser, 1993
Bristol	Fetal		20 - 70[1]	Henshaw et al 1995
Pripyat	Fetal		150 – 410*	Henshaw et al 1995
Oxfordshire	Placental	<9 - 60	8 – 140	Bradley and Ewers, 1995
Cumbria	Placental	<4 - 30	30 – 150	Bradley and Prosser, 1993
Pripyat	Placental		260†	Henshaw et al 1995

* Range is the different mean concentration of individual organs (thymus, liver and spleen). All other ranges are for measurements on whole samples.

† Value is the mean of 16 measurements.

Henshaw (Henshaw et al 1995) used an entirely different measurement technique in which the sample was placed in contact with CR-39 plastic detectors and the alpha-particle tracks in the etched detectors counted. This enabled the concentrations of ^{210}Po in individual organs to be measured. The published organ means for UK samples are close to the range of concentrations measured by Bradley in whole fetus. Henshaw also measured samples from Pripyat taken after the Chernobyl accident. He found elevated levels when compared with the UK samples and suggested that these were probably due to dietary differences rather than the accident.

4 IN VIVO MEASUREMENTS

In this section, results for Southern England, Northern England, Scotland and Wales are described separately mainly as a consequence of the regional variations which occurred after the Chernobyl accident.

4.1 Southern England

The earliest measurements were made in 1956 and the most recent in 1997. The area covered includes all of Southern England as far north as a line from Hull

(Humber estuary) in the east to Preston in the west. In fact the furthest north that measurements have been made is Cambridge.

Most of the measurements made are of ^{137}Cs before the Chernobyl accident, ^{134}Cs, ^{137}Cs and ^{131}I after the Chernobyl accident and ^{131}I after the Windscale accident. Some measurements of ^{95}Zr have also been reported.

4.1.1 Strontium isotopes

4.1.1.1 Literature review

No reports of measurements of strontium isotopes in residents of Southern England were found in the literature.

4.1.2 Caesium isotopes

4.1.2.1 Introduction

In general whole body caesium values are reported in the literature in terms of Bq of caesium per g of potassium rather than Bq kg^{-1} body weight. This is because there is a significant correlation between total body radiocaesium and total body potassium, whereas there is not a strong correlation between total body radiocaesium and body weight. In the case of older papers data expressed in Curies has been converted to Becquerels. In some references values are expressed as Bq of caesium per mmol of K or Bq of caesium per Bq of ^{40}K. In this report values are expressed in the units of Bq g^{-1} K. If the original paper does not use these units and there are no details of the subjects' potassium content then values from ICRP have been used. For adult males a value of 133 g K is assumed and for adult females 87g (ICRP, 1975). In cases where males and females are not treated separately an average adult value of 110 g is used. Potassium contents for children have also been taken from ICRP (1975) where necessary.

As far as possible the literature review is in order of measurement date with the oldest measurements being discussed first.

4.1.2.2 Literature review

Rundo has reported the earliest whole body ^{137}Cs measurements made at the Harwell laboratories from 1956-1960 (Rundo, 1960). The subjects were residents of Berkshire or Oxfordshire. The results are reported in Table 4.1.1. The values were compared with groups from the United States at the Argonne National Laboratory, Illinois and the Los Alamos Laboratory, New Mexico. It was concluded from the similarity of the data that the radiocaesium found in Harwell employees is a result of worldwide fallout and not local contamination.

The measurements at Harwell laboratories were continued between 1960 to 1962 (Rundo and Newton, 1962) and again between 1962 and 1964 (Rundo and Newton, 1964) The results for these periods and are also reported in Table 4.1.1.

The large increases in whole body ^{137}Cs in 1962 and 1963 are attributed to the resumption of atmospheric testing of nuclear weapons in September 1961.

Measurements of ^{137}Cs in whole body were also made on staff at the Radiological Protection Service (RPS) at Sutton in Surrey. The staff were mainly residents of the Greater London area and were measured between 1958 and 1967 (Godfrey and Vennart, 1968). The results are very similar to those made at the Harwell laboratory over the same period. The RPS made measurements of a group of women from local antenatal clinics during and immediately following pregnancy (between 1963 and 1965) and also sometimes of their babies (1964 and 1965) (Godfrey and Vennart, 1968). The measurements of pregnant women produced results which were very similar to the staff measurements. The measurements of babies gave mean $^{137}Cs/K$ ratios of 115-198 pCi $g^{-1}K$ (4.4-7.3 Bq $g^{-1}K$).

A group of adult males, generally 10-20 in number was measured between 1964 and 1976 again at Harwell (Newton et. al, 1977). The results for these measurements are also reported in Table 4.1.1. The results show that ^{137}Cs in residents of Central Southern England were smaller during 1974-76 than at any time during the previous 17 years.

TABLE 4.1.1 Mean ^{137}Cs/K ratios between 1956 and 1977 in male Harwell employees not exposed occupationally. (Rundo, 1960; Rundo and Newton, 1962; Rundo and Newton, 1964; Newton et al, 1977)

Date of measurement	Number of subjects	Mean ^{137}Cs/K (Bq g^{-1})
February-June 1956	11	1.2
July-December 1956	3	1.1
January-June 1957	11	1.3
July-December 1957	7	1.5
January-June 1958	6	1.6
July-December 1958	8	2.0
January-June 1959	12	2.1
July 1959	11	2.3
December 1959	7	2.1
March 1960	10	2.1
June 1960	10	1.9
September 1960	13	1.8
December 1960	14	1.5
March 1961	14	1.3
July 1961	12	1.3
October 1961	14	1.1
January 1962	10	1.0
April 1962	15	0.9
July 1962	6	1.3
April 1962	15	0.9
July 1962	12	1.3
October 1962	15	1.9
January 1963	10	2.0
April 1963	12	2.4
July 1963	11	3.3
October 1963	13	4.3
January 1964	11	5.6
April 1964	11	5.6
July 1964	13	6.0
October 1964	14	6.4
January 1965	15	6.3
April 1965	16	6.2
July 1965	16	5.7
October 1965	17	4.8
January 1966	17	4.3
April 1966	17	3.8
July 1966	16	3.2
October 1966	17	2.6
January 1967	19	2.1
April 1967	17	1.9
July 1967	18	1.6
October 1967	18	1.3
January 1968	16	1.1

Date of measurement	Number of subjects	Mean ^{137}Cs/K (Bq g^{-1})
April 1968	16	0.9
July 1968	16	0.9
October 1968	15	0.9
January 1969	16	0.8
April 1969	16	0.7
July 1969	16	0.8
October 1969	13	0.8
January 1970	14	0.7
April 1970	13	0.7
July 1970	14	0.7
October 1970	14	0.7
January 1971	15	0.6
April 1971	15	0.7
July 1971	13	0.7
October 1971	14	0.7
January 1972	15	0.9
April 1972	14	0.8
July 1972	15	0.8
October 1972	12	0.7
January 1973	14	0.6
April 1973	13	0.6
July 1973	13	0.5
October 1973	14	0.4
April 1974	15	0.3
July 1974	15	0.3
October 1974	14	0.3
March 1975	12	0.4
July 1975	12	0.4
September 1975	10	0.4
February 1976	16	0.4
July 1976	16	0.3
October 1976	15	0.3
January 1977	15	0.3

In 1963-64 Rundo made measurements of ^{137}Cs in babies and their mothers (Rundo, 1970; Rundo and Turner, 1992). The measurements were made at the Harwell laboratory and are reported in Table 4.1.2. The data is expressed as Bq kg^{-1} and not Bq g^{-1} K as little information is available on potassium content of infants. The mothers values have been converted to Bq/gK by multiplying by 58 (the average weight of a female in kg (ICRP, 1975)) and divided by 87 (the average K content in g, (ICRP, 1975). The values of Bq/gK are shown in brackets in Table 4.1.2. This gives values which are very similar to the values reported in Table 4.1.1.

TABLE 4.1.2 Caesium-137 content of babies of various ages and their mothers (Rundo, 1970)

		^{137}Cs (Bq kg^{-1})	
Subject	Age (days)	Infant	Mother
A	57	4.4	6.3 (4.2)
A	95	5.2	7.8 (5.2)
A	137	5.9	7.6 (5.1)
A	174	6.4	8.3 (5.5)
B	153	7.0	7.8 (5.2)
C	75	4.6	10 (6.7)
C	92	4.9	10 (6.7)
D	76	7.1	7.4 (4.9)
D	140	16	8.5 (5.7)
D	170	17	-
D	203	14	-
E	56	7.2	7.8 (5.2)
E	105	11	8.9 (5.9)
E	148	10	-
F	57	17	-
F	96	17	-

Measurements have also been made at NRPB of a group of residents of Southern England who are not occupationally exposed. Caesium-137 values have been reported for the males measured in 1977 (NRPB, 1993), 1980 (Fry et al, 1981) and 1981 (Fry et al, 1982) and are reported in Table 4.1.3. Measurements of this group were not made between 1981 and the Chernobyl accident.

TABLE 4.1.3 Mean ^{137}Cs/K ratios for male residents of Southern England (Fry et al, 1981; 1982; NRPB, 2003)

Date	Number of measurements	Mean ^{137}Cs/K ratio (Bq g^{-1})
1977, Quarter 1	20	0.3
1977, Quarter 2	20	0.3
1977, Quarter 3	16	0.3
1977, Quarter 4	16	0.3
1980, Quarter 1	164	0.4
1980, Quarter 2	67	0.4
1980, Quarter 3	86	0.3
1980, Quarter 4	20	0.3
1981, Quarter 1	154	0.31
1981, Quarter 2	27	0.25
1981, Quarter 3	85	0.29
1981, Quarter 4	37	0.36

Kang has reported on measurements of whole body radiocaesium made at the Harwell Laboratory in Oxfordshire in the period 1976-1988 (Kang, 1989). The subjects are 16 adult males with no known occupational exposure who are residents of Oxfordshire of Berkshire (Table 4.1.4). Changes in the membership of the subject group through the years have been minimal.

TABLE 4.1.4 Mean ^{137}Cs/K ratios from 1977 to 1986 in male Harwell employees not exposed occupationally. (Kang, 1989)

Date of measurement	Number of subjects	^{137}Cs/K (Bq g^{-1}) mean ± 1 s.d.
January, 1977	15	0.33 ± 0.15
April, 1977	15	0.29 ± 0.12
July, 1977	14	0.26 ± 0.12
October,1977	16	0.33 ± 0.17
January, 1978	16	0.32 ± 0.14
April, 1978	16	0.39 ± 0.10
July, 1978	16	0.38 ± 0.15
September,1978	15	0.38 ± 0.16
January, 1979	15	0.40 ± 0.14
April, 1979	15	0.43 ± 0.19
July, 1979	16	0.45 ± 0.17
October,1979	16	0.41 ± 0.15
January, 1980	18	0.32 ± 0.13
April, 1980	18	0.28 ± 0.11
July, 1980	17	0.30 ± 0.14
October,1980	17	0.31 ± 0.16
January, 1981	17	0.22 ± 0.15
April, 1981	17	0.23 ± 0.15
July, 1981	18	0.27 ± 0.17
October,1981	17	0.25 ± 0.13
January, 1982	18	0.24 ± 0.07
April, 1982	18	0.24 ± 0.11
July, 1982	16	0.26 ± 0.13
October,1982	16	0.22 ± 0.11
January, 1983	16	0.21 ± 0.09
April, 1983	16	0.20 ± 0.09
July, 1983	14	0.16 ± 0.06
October,1983	16	0.19 ± 0.08
January, 1984	16	0.14 ± 0.07
April, 1984	16	0.20 ± 0.08
July, 1984	15	0.15 ± 0.07
October,1984	17	0.14 ± 0.06
January, 1985	16	0.15 ± 0.06
April, 1985	17	0.14 ± 0.07
July, 1985	18	0.10 ± 0.07
October,1985	19	0.13 ± 0.08
January, 1986	16	0.09 ± 0.08
April, 1986	16	0.07 ± 0.07

Estimates of whole body ^{137}Cs and ^{134} Cs following the Chernobyl accident are shown in Table 4.1.5 for the Harwell group (Kang, 1989; Cambray et al, 1987). Measurements were also made of workers at the Buckinghamshire laboratories of Amersham International. The measured ^{134}Cs values, which are unlikely to be complicated by occupational exposure, were found to be very similar to the Harwell group reaching a peak value in July 1987 and exhibiting comparable values throughout (Cambray et.al., 1987).

By April 1986, prior to the Chernobyl accident, whole body radioactive caesium in residents of central Southern England had declined to the lowest value since monitoring began in 1956. Caesium-137 derived from Chernobyl reached peak values in July 1987 at roughly 25% of the levels found in Harwell subjects during 1964-1965, the years following intensive weapons testing. A model using measured values for ^{137}Cs in rainfall predicts that during the years 1977-1985 a maximum of 27% of the whole body ^{137}Cs may have been due to discharges to sea during fuel reprocessing (Kang, 1989).

TABLE 4.1.5 Mean ^{137}Cs/K and ^{134}Cs/K ratios in Harwell subjects from June 1986 to January 1989 (Kang, 1989; Cambray et al, 1987)

Date of measurement	Number of subjects	^{137}Cs/K (Bq g^{-1}) mean ± 1 s.d.	^{134}Cs/K (Bq g^{-1}) mean ± 1 s.d.
24th June to 8th July 1986	8	0.40 ± 0.76	0.22 ± 0.09
16th July to 28th July 1986	10	0.76 ± 0.26	0.38 ± 0.16
18th August to 29th August 1986	17	0.84 ± 0.41	0.42 ± 0.19
24th September to 8th October 1986	15	1.07 ± 0.48	0.53 ± 0.21
6th November to 20th November 1986	18	1.10 ± 0.33	0.51 ± 0.18
12th January to 22nd January 1987	16	1.15 ± 0.29	0.51 ± 0.16
18th February to 2nd March 1987	16	1.31 ± 0.32	0.56 ± 0.16
18th March to 31st March 1987	14	1.42 ± 0.41	0.60 ± 0.15
13th April to 12th May 1987	15	1.49 ± 0.49	0.58 ± 0.20
1st July to 29th July 1987	17	1.63 ± 0.60	0.62 ± 0.23
4th September to 1st October 1987	16	1.44 ± 0.40	0.52 ± 0.15
18th November to 4th December 1987	17	1.26 ± 0.38	0.42 ± 0.11
12th January to 29th January 1988	17	1.17 ± 0.49	0.38 ± 0.14
10th March to 13 April 1988	15	0.93 ± 0.39	0.30 ± 0.12
13th June to 14th July 1988	13	0.75 ± 0.26	0.20 ± 0.07
19th September to 5th October 1988	15	0.63 ± 0.21	0.16 ± 0.05
20th December 1988 to 6 January 1989	16	0.54 ± 0.21	0.13 ± 0.04

In 1986, the Department of the Environment commissioned research to measure volunteer members of the public using whole body monitoring. Although originally intended to investigate the effects of Sellafield discharges the research began shortly before the Chernobyl accident. This large study of ^{137}Cs and ^{134}Cs whole body contents of people in England, Scotland and Wales data was obtained from a mobile whole body counter which visited Bristol (December, 1987), Canterbury (January 1988), Exeter (December 1987), Ipswich (February

1988) and Reading (May 1988). The date in parenthesis is the month and year that the measurements were carried out (Boddy et al, 1989a) and (McKenzie et al, 1989). In these reports values are reported as Bq in whole body or as the ratio of Bq of radiocaesium to Bq of potassium-40. This has been done as the dose delivered per unit activity of caesium-137 or caesium-134 in soft tissues is similar to that delivered per unit activity of potassium-40.

The measured ratio of $^{137}Cs/^{134}Cs$ was close to 2.0 and the authors conclude from this is that the primary source for the radiocaesium was the Chernobyl accident. The results are reported in Table 4.1.6.

TABLE 4.1.6 Results for volunteers in Southern England for December 1987 to May 1988 (Boddy et al, 1989a)

Location	Age	Sex	Number of volunteers	mean ^{137}Cs, Bq	mean ^{134}Cs, Bq	Average Cs/K
Bristol	≤ 6		3	135 (1.8)	48.8 (0.65)	0.079
Bristol	6-15		13	89.4 (1.2)	32.4 (0.43)	0.053
Bristol	>16	M	53	195 (1.4)	70.6 (0.51)	0.063
Bristol	>16	F	60	138 (1.4)	49.9 (0.50)	0.061
Canterbury	≤ 6		11	21.1 (0.77)	8.3 (0.30)	0.035
Canterbury	6-15		30	66.9 (0.97)	26.2 (0.38)	0.044
Canterbury	>16	M	83	175 (1.2)	68.6 (0.47)	0.055
Canterbury	>16	F	118	109 (1.1)	42.8 (0.45)	0.052
Exeter	≤ 6		1	27.9 (0.89)	10.5 (0.34)	0.040
Exeter	6-15		10	41.9 (0.65)	15.8 (0.24)	0.029
Exeter	>16	M	66	188 (1.3)	70.9 (0.50)	0.059
Exeter	>16	F	36	128 (1.3)	48.1 (0.49)	0.058
Ipswich	≤ 6		8	26.9 (1.1)	10.2 (0.42)	0.050
Ipswich	6-15		7	74.6 (0.98)	28.3 (0.37)	0.044
Ipswich	>16	M	50	160 (1.2)	60.2 (0.44)	0.052
Ipswich	>16	F	55	114 (1.2)	43.3 (0.45)	0.053
Reading	≤ 6		7	40.1 (0.53)	18.3 (0.24)	0.025
Reading	6-15		11	11.2 (0.21)	5.1 (0.10)	0.010
Reading	>16	M	41	119 (0.85)	54.0 (0.38)	0.040
Reading	>16	F	71	66.3 (0.72)	30.2 (0.33)	0.034

Note
Figures in parentheses are in Bq g^{-1} K and have been calculated using the authors' Cs/K ratio and assuming an activity of 30.7 Bq per g K.

The average Cs/K ratios are the activity of ^{134}Cs plus ^{137}Cs divided by the activity of ^{40}K.

As part of the wider survey described in Boddy et al. (1989a) measurements made at Cambridge are described by Dendy et al. (1992) and Hayball and Dendy (1991). The average radiocaesium to potassium-40 ratios for measurements between April 1988 and August 1989 were 0.023, 0.021, 0.015 and 0.019 for adult males, adult females, male children and female children respectively. These values are similar to the values measured at Reading in May 1988 by Boddy et al (1989a) which is

the closest geographically to the areas covered in this survey. An attempt was made to correlate Cs/K activity ratios in Cambridgeshire residents with diet, age and other factors. It was found that children had lower ratios than adults, and male adults had higher ratios than adult females. However, no trend with diet or area of Cambridgeshire was found.

On 30th July 1986 two members of staff at Hammersmith hospital were measured for ^{134}Cs and ^{137}Cs (Bewley and Freemantle, 1986). The results for ^{134}Cs were 42 and 115 Bq (approximately 0.3 and 0.9 Bq g^{-1} K) and for ^{137}Cs the results were 82 and 235 Bq (approximately 0.6 and 1.8 Bq g^{-1} K).

At NRPB a group of approximately 30 people employed at the Chilton site in Oxfordshire were measured regularly from May 1986 to late 1989, a period of some 1200 days after the Chernobyl accident. The results have been extensively reported in the literature (Fry and Britcher, 1987; Despres, 1990; Fry et al, 1989 and Etherington and Dorrian, 1990). Summaries of the whole body ^{137}Cs and ^{134}Cs measurements are shown in Figure 4.1.1 and Figure 4.1.2 respectively against the number of days following the Chernobyl accident.

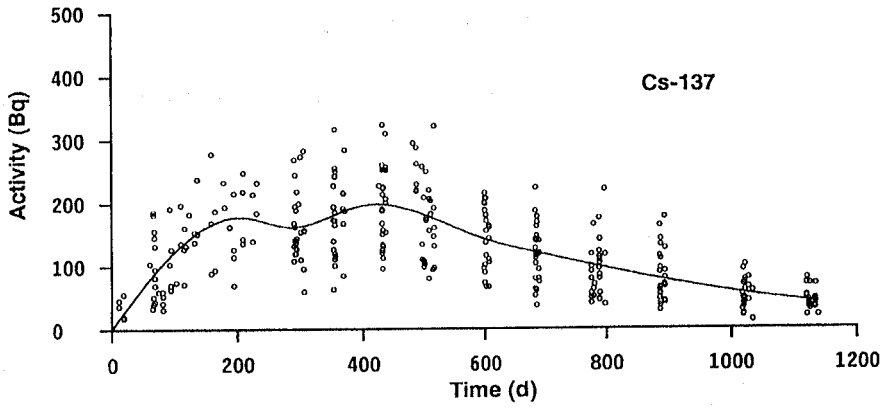

FIGURE 4.1.1 Whole body ^{137}Cs measurements for NRPB employees (Etherington and Dorrian, 1990)

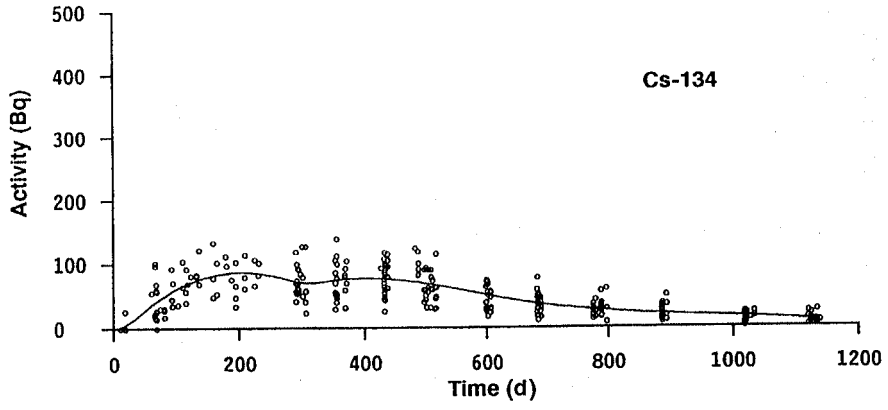

FIGURE 4.1.2 Whole body ^{134} Cs measurements for NRPB employees (Etherington and Dorrian, 1990)

It can be seen that after an initial rapid rise to a mean value for ^{137}Cs of 180Bq the average radiocaesium level began to decrease before increasing again to a second maximum in the summer of 1987. Since then the results show a decreasing level and in June 1989 the average level of ^{137}Cs was approximately 50 Bq. The results for ^{134}Cs show a similar trend. There are large variations between persons depending on the dietary habits and individual variations in metabolism; extreme values differ typically by a factor of four.

Between 1990 and 2001 NRPB have made occasional measurements of between 4 and 20 staff who are resident in Oxfordshire of Berkshire who are not occupationally exposed. The only nuclide detected is ^{137}Cs (apart from 1990) and the results are reported in Table 4.1.7 and Table 4.1.8. The levels were found to have stabilised in 1994 close to the levels measured in this area prior to the Chernobyl accident. This implies that most of the ^{137}Cs now measured originates from past atmospheric weapons tests.

TABLE 4.1.7 ^{137}Cs measurements in whole body for residents of Oxfordshire and Berkshire (NRPB, 2003)

		Results above the detection limit		
Year of measurements	Number of results below the detection limit	Number	Mean, Bq	Range, Bq
1990	7	10	33.4	20-55
1991	26	7	26.1	21-35
1992	23	9	22.8	18.5-29.3
1993	13	1	23.6	-
1994	22	2	22.7	21.8-23.6
1997	2	2	30.0	29.4-30.6
2000	5	1	22.5	-
2001	6	0	-	-

Note: 95% detection limit is approximately 18 Bq

TABLE 4.1.8 ^{134}Cs measurements in whole body for residents of Oxfordshire and Berkshire (NRPB, 2003)

		Results above the detection limit		
Year of measurements	Number of results below the detection limit	Number	Mean, Bq	Range, Bq
1990	12	5	15.4	14-18
1991	32	1	15.4	-
1992	31	1	12.6	-
1993	14	0	-	-
1994	24	0	-	-
1997	4	0	-	-
2000	5	1	13.9	-
2001	6	0	-	-

Note: 95% detection limit is approximately 12 Bq

4.1.2.3 Summary

Measurements of ^{137}Cs made at the Harwell laboratories between 1956 and 1966 and at Sutton are very similar.

Since 1966 measurements made at Harwell have shown a steady decrease to 1977. Later measurements from 1977 to April 1986 at NRPB and Harwell have confirmed the downward trend before the Chernobyl accident.

Measurements of ^{134}Cs and ^{137}Cs made at Harwell after the Chernobyl accident show an increase up to July 1987 where the levels of ^{137}Cs were approximately 20 times the levels immediately prior to the accident. The measured ratio of ^{137}Cs/^{134}Cs confirms the principal source of the radiocaesium to be the Chernobyl accident. The peak ^{137}Cs in these groups was only 25% of the peak levels measured in 1965/1965. Similar results were obtained from the NRPB study.

The results of the nation-wide survey conducted by the Northern Region Health Authority for Bristol, Canterbury, Exeter, Ipswich and Reading gave results which are very similar to the Harwell measurements.

After the Chernobyl accident measurements at NRPB show that ^{137}Cs levels stabilised in 1991 close to the levels measured prior to the Chernobyl accident.

4.1.3 Iodine isotopes

4.1.3.1 Literature review

From October 1957, soon after the Windscale accident, until January 1958, measurements of ^{131}I in thyroid were made of 15 residents of the London area (Maycock and Vennart, 1958). The results of these measurements are shown in Table 4.1.9. The authors conclude that the main source of ^{131}I was ingestion as the atmospheric contamination of ^{131}I was negligible at the time of measurement.

TABLE 4.1.9 Thyroid ^{131}I activity (Bq) of London area residents (Maycock and Vennart, 1958)

	Date of measurement							
Subject	31/10/57	1/11/57	4/11/57	5/11/57	8/11/57	15/11/57	22/11/57	2/1/58
A	96							
B	22							
C	67			56	37	26		
D	74					30		
E	78					7		
F	67							
G	48							
H		233		190	140	96	67	3.3
I		89			55			5.9
J		33						
K		315						
L		37						
M		44						
N		59	37					
O		52				11		
P			44					
Q				56	52	37		
R				22				

Notes
Subjects B and L were residents of mid-Surrey
Subject K was a visitor from Leeds
Subjects F and G were children

The thyroid ^{131}I activity was measured in five residents of Oxfordshire and Berkshire between 7th May 1986 and 30th May 1986 (Cambray et al, 1987). Four of the subjects had maximum ^{131}I activities between 18 and 28 Bq. The fifth subject was measured on 4 occasions and no evidence of ^{131}I was found.

Fry and Milton measured ^{131}I activity in the thyroid of over 100 residents of Oxfordshire immediately after the Chernobyl accident (Despres, 1990). The results are given in Table 4.1.10.

TABLE 4.1.10 Thyroid ^{131}I activity (Bq) in residents of Southern England (Despres, 1990)

Date of measurement	Age	Results below the detection limit		Results above the detection limit		
		Number	Detection limit, Bq	Number	Mean	Range
9-15th May 1986	>20	6	10	5	18	13-28
14th May 1986	14			1	12	
14th May 1986	8			1	12	
13-14th May 1986	1-2	2	20			
16th May 1986	>20	4	30			
16th May 1986	1	10	90			

Measurements of ^{131}I in thyroid were also measured in May 1986 for residents of London suburbs by Hill (Despres, 1990). The results are shown in Table 4.1.11 and show that the maximum activity was reached between 15 and 20 May 1986.

TABLE 4.1.11 Thyroid ^{131}I activity (Bq) in London residents in May 1986 (Despres, 1990)

Date	Age and sex												
May 1986	60M	56M	33M	36M	35 F	35 M	34 M	26F	12M	6F	4M	4M	2.5 F
10		8±4	21±5	17±5		27±6		15±5		6±5			
11	8±4						17±5		8±4				11±6
14								20±4					
15			30±5			30±5							
16	10±4	13±4											
17							23±4						16±5
19		7±4	33±6					13±4		2±2			
20						23±4							
21		6±3			8±3			20±5			8±3	13±4	
22			26±4					15±4					

Note: Uncertainties are one standard deviation

Immediately following the Chernobyl accident ^{131}I measurements were made at Addenbrookes Hospital in Cambridge (Dendy et al, 1986). These measurements were mainly of visitors to countries in Europe but included 28 adults living in the

Cambridge area. The maximum ^{131}I in whole body measurement was 140 Bq on 6th May 1986. A group of 15 adult volunteers all measured on 21st May had an average ^{131}I content of 35 Bq.

4.1.3.2 Summary

Measurements of ^{131}I in thyroid were made in residents of London after the Windscale accident. The measured values are much less than for residents of Leeds and Cumbria made at this time (section 4.2.6).

A small number of ^{131}I in thyroid measurements were made in Southern England after the Chernobyl accident. Measurements between 7th May and 22nd May 1986 gave results ranging from 2 to 30 Bq. All measurements made in Oxfordshire after 16th May were below the detection limit.

4.1.4 Plutonium and americium isotopes

4.1.4.1 Literature review

No reports of measurements of plutonium and americium isotopes in residents of Southern England were found in the literature.

4.1.5 Uranium thorium and polonium isotopes

4.1.5.1 Literature review

No reports of measurements of uranium, thorium or polonium isotopes in residents of Southern England were found in the literature.

4.1.6 Other radionuclides

4.1.6.1 Literature review

Rundo and Newton made measurements of zirconium-95 in a small group of residents of Oxfordshire and Berkshire between January 1962 and October 1963 (Rundo and Newton, 1962; 1965). The results are reported in Table 4.1.12. The range quoted in October 1962 is because of uncertainty on the ratio of ^{95}Zr to ^{95}Nb which both emit gamma rays of about 0.76 MeV. Repeat measurements of four subjects show a minimum value in autumn 1962 and a maximum value in spring/summer of 1963. After 1963 the decreasing lung content of ^{95}Zr and the increase in the adjacent ^{137}Cs peak made estimates of the ^{95}Zr content unreliable.

TABLE 4.1.12 ^{95}Zr content of the lung for residents of Oxfordshire and Berkshire in 1962 and 1963 (Rundo and Newton, 1965).

Date of measurement	No. of subjects	Average ^{95}Zr content of lung, pCi	Average ^{95}Zr content of lung, Bq
January,1962	10	~130	~4.8
April,1962	14	129	4.8
July,1962	13	79	2.9
October,1962	16	68-120	2.5- 4.4
Nov/Dec 1962	7	~120	~4.4
Jan/Feb 1963	9	145	5.4
April 1963	12	161	6.0
July/Aug, 1963	12	149	5.5
October 1963	11	73	2.7

From April 1988 to March 1991, 1220 volunteers were monitored using the whole body monitor at Addenbrookes hospital in Cambridgeshire. Fourteen volunteers were found to have measurable activities of radionuclides in addition to ^{137}Cs and ^{134}Cs. The nuclides detected and the estimated activity are shown in Table 4.1.13. (Dendy et al, 1992). The probable reason for the contamination is also shown in Table 4.1.13. The main contaminants were ^{99m}Tc and ^{131}I because of their widespread use in medicine.

TABLE 4.1.13 Man-made radionuclides other than radiocaesium detected in or on volunteers (Dendy et al, 1992)

Nuclide	Number of cases	Estimated activity, Bq	Reason for contamination
^{99m}Tc	6	< 1000	subjects had been in Nuclear Medicine waiting room prior to monitoring
^{22}Na	1	1000	accidental spillage during a Nuclear Medicine test
^{226}Ra	1	200	luminous watch- after removal
^{86}Rb	2	2000 600	used in studies at medical research laboratory on site
^{232}Th	1	100	a physics teacher, thorium was used at school (may be natural, poor statistics).
^{131}I	3	1000 200 100	all visitors to patients on radionuclide therapy ward

4.1.6.2 Summary

Measurements of ^{95}Zr in the lung of residents of Oxfordshire and Berkshire in 1962 and 1963 reached a maximum of 6.0 Bq in April 1963.

In Cambridgeshire during measurements of radiocaesium, nuclides other than radiocaesium were found in a small number of volunteers. These generally were nuclides which are frequently used in medicine.

4.2 Northern England

This section covers in vivo measurements made on residents of Northern England and the Isle of Man. The earliest measurements reported were made in 1957 and the most recent in 1989. The area covers the area north of a line from Hull (Humber estuary) in the east to Preston in the west.

Most of the measurements made are of ^{137}Cs before the Chernobyl accident and ^{134}Cs and ^{137}Cs after the Chernobyl accident. After the Windscale and Chernobyl accidents measurements of ^{131}I in the thyroid were made. Ruthenium-106 and single measurements of ^{60}Co and ^{144}Ce have also been reported for residents of Cumbria.

4.2.1 Strontium isotopes

No reports of measurements of strontium isotopes in residents of Northern England were found in the literature.

4.2.2 Caesium isotopes

4.2.2.1 Introduction

In general whole body caesium values are reported in the literature in terms of Bq of caesium per g of potassium rather than Bq kg^{-1} body weight. This is because there is a significant correlation between total body radiocaesium and total body potassium, whereas there is not a strong correlation between total body radiocaesium and body weight. In the case of older papers data expressed in Curies has been converted to Becquerels. In some references value are expressed as Bq of caesium per mmol of K or Bq of caesium per Bq of ^{40}K. In this report values are expressed in the units of Bq g^{-1} K. If the original paper does not use these units and there are no details of subjects' potassium content then values from ICRP have been used. For adult males a value of 133 g K is assumed and for adult females 87g (ICRP, 1975). In cases where males and females are not treated separately then an average adult value of 110 g is used. Potassium contents for children have also been taken from ICRP (1975) where necessary.

As far as possible the literature review is in order of measurement date with the oldest measurements being discussed first.

4.2.2.2 Literature review

A few subjects resident near Windscale were measured for ^{137}Cs around the time of the accidental release from Windscale No.1 Pile in October 1957 (Rundo, 1960). The results are reported in Table 4.2.1.

TABLE 4.2.1 ^{137}Cs measurements of residents near Windscale (Rundo, 1960)

		^{137}Cs/ gK	
Subject	Date Measured	Pre-accident	Post-accident
G	29-3-1957	5.1	
	26-11-1958		6.6
C	25-4-1957	4.0	
	13-3-1958		16
	21-7-1958		11
T	27-6-1957	4.0	
	15-1-1958		14
	3-7-1958		8.4
	11-1-1960		4.7
Mrs T	9-9-1957	2.8	
Go	2-4-1958		8.3
R	7-5-1958		6.8
W	18-5-1960		5.0

The average of the pre-accident ^{137}Cs level is 4 Bq/gK. The author states this is 2.5-3 times the average value for residents of Berkshire and Oxfordshire which was attributed partly to the higher rainfall and to the slight environmental contamination from the Windscale works. The post-accident levels increased by roughly a factor of four around January to March 1958 and fell to less than half the maximum post-accident levels towards the end of 1958.

Hesp has reported measurements of ^{137}Cs in a small group of residents of West Cumbria (Hesp, 1965). The subjects were employed at Windscale and Calder Hall on work which did not involve contact with radioactive material. Measurements of 14 subjects were made from 1961 to 1964 and the results for male subjects are reported in Table 4.2.2.

TABLE 4.2.2 ^{137}Cs measurements of residents of West Cumbria (Hesp, 1965)

		^{137}Cs/gK (Bq g^{-1})	
Year	Quarter	Mean	Range
1961	2	2.3	1.2-3.6
1961	4	2.3	1.1-4.1
1962	1	1.6	0.8-2.4
1962	2	1.8	1.0-2.9
1962	3	2.5	1.8-3.3
1962	4	3.4	2.2-5.5
1963	1	4.0	2.0-6.1
1963	2	5.1	3.3-7.8
1963	3	7.3	4.3-12
1963	4	9.2	6.2-16
1964	1	9.4	6.4-16
1964	2	9.5	6.6-17
1964	3	10.1	7.3-16

The author reports that the measured values are approximately double those in adult males in Berkshire and West Oxfordshire (Rundo and Newton, 1964). This is presumed to be as a result of different amounts of fallout in the diet in the respective areas and this is supported by measurements of ^{137}Cs in milk.

In September 1977, at the time of the Windscale enquiry, seventeen volunteers from the Ravenglass and Whitehaven areas of Cumbria were measured for whole body ^{137}Cs (BNFL, 1977). Initial measurements were made and follow-up measurements 2 and 4 weeks later. After the initial measurements the volunteers consumed known amounts of fish caught in the sea adjacent to the Windscale works. The aim of this work was to determine the activity of ^{137}Cs that would be present if the volunteers continued to eat locally caught fish at a known rate indefinitely. The measured activities at the initial measurement and at 2 and 4 weeks are shown in Table 4.2.3. This table also gives the estimated equilibrium values. The original data was reported as nCi which has been converted to Bq/g K assuming an average K content of 110 g.

TABLE 4.2.3 ^{137}Cs content of Cumbrian fish consumers (BNFL, 1977)

Volunteer	Average weekly quantity of fish consumed, g	Whole body ^{137}Cs activity, Bq/g K			
		Initial	2nd week	4th week	Estimated equilibrium value*
1	298	0†	6.4	7.4	41
2	284	3.0	6.1	7.1	27
3	340	2.0	5.0	7.7	34
4	284	3.4	8.4	8.4	32
5	255	0.7	N/A‡	5.4	28
6	340	0.7	9.8	11	58
7	213	8.1	10.1	11	9.8
8	397	3.0	9.4	11	48
9	397	4.7	12	12	49
10	354	1.0	7.1	8.7	45
11	241	4.4	7.7	7.1	20
12	170	12	13.1	13	13
13	170	14	18	17	16
14	936	83	86	76	82
15	191	9.8	13	12	12
16	170	3.4	6.1	5.4	14
17	269	3.0	9.1	9.4	39

* The equilibrium value is the whole body activity which would be reached if the volunteers continued eating the supplied fish indefinitely.

† Reproduced from BNFL table.

‡ N/A indicates the volunteer was unavailable for measurement.

Between 1978 and 1989, BNFL Sellafield made whole body ^{137}Cs measurements on residents of Cumbria who requested a measurement (Peace, 2002). These

people were not occupationally exposed. The results are summarised in Table 4.2.4.

TABLE 4.2.4 Measurements of residents of Cumbria between 1978 and 1989 (Peace, 2002)

Year	Number of measurements	Average ^{137}Cs, Bq	Range of ^{137}Cs, Bq	Average ^{137}Cs/K ratio* (Bq/gK)
1978	2	140	130 – 150	1.3
1979	9	320	37 – 999	2.9
1980	3	210	37 – 481	1.9
1981	11	250	52 – 533	2.3
1982	31	160	3.7- 670	1.5
1983	41	290	11 – 2960	2.6
1984	21	250	3.7 – 2690	2.3
1985	3	470	74 – 1070	4.3
1986	10	470	140 – 1700	4.3
1987	13	180	80 – 350	1.6
1988	2	150	70 –220	1.4
1989	3	130	60 – 230	1.2

* Assuming an average K content of 110 g (ICRP, 1975)

During the same study a few measurements were also made of ^{134}Cs (Peace, 2002) and are shown in Table 4.2.5 with the associated ^{137}Cs measurement.

TABLE 4.2.5 Measurements of residents of Cumbria between 1978 and 1989 (Peace, 2002)

Date of measurement	^{137}Cs ,Bq	^{134}Cs, Bq	Ratio ^{137}Cs/^{134}Cs
19/11/81	533	48.1	11
5/12/83	2430	167	15
14/12/83	781	25.9	30
9/1/84	633	59.2	11
24/2/84	2690	207	13
8/7/86	1700	534	3.2
8/7/86	956	565	1.7
28/7/86	360	520	0.69
28/7/86	140	290	0.48
23/8/86	195	86	2.3
1/10/86	411	294	1.4
21/9/87	90	130	0.69
25/9/87	170	190	0.89

In addition to residents of Cumbria BNFL Sellafield made some measurements of radiocaesium in some residents of the Isle of Man (Peace, 2002). The results are reported in Table 4.2.6.

TABLE 4.2.6 Measurements of residents of the Isle of Man (Peace, 2002)

Date of measurement	Total radiocaesium, Bq	^{137}Cs, Bq	^{134}Cs,Bq
21/4/83		215	
20/11/86	1040		
10/9/87		892	445
10/9/87		452	
26/10/87		445	
28/4/88	533		

In a study of ^{137}Cs levels in residents of Scotland Williams et al (1981). made one incidental measurement of a resident of Liverpool in 1979 of 0.7 Bq ^{137}Cs/gK.

In 1984, 290 residents of Seascale in Cumbria and its environs were measured for ^{137}Cs using a mobile whole body monitor (Fry and Sumerling, 1984). The results are summarised in Table 4.2.7. Measurements were made on all who volunteered; no selection criteria were used. In more than 90% of cases no ^{137}Cs was detected, and for 17 others the result was close to the detection limit and therefore subject to considerable statistical uncertainty. In only four cases was ^{137}Cs readily detectable. The authors conclude that the higher contents for these are either due to consumption of local foodstuffs or employment on the Sellafield site. The median ^{137}Cs value for all those measured was calculated by statistical means to be 0.09 kBq which is approximately twice the value for a southern England population. The difference was attributed to higher levels of fallout, consumption of local foodstuffs, particularly fish and to the inclusion of persons who work on the Sellafield site. The authors reported ^{137}Cs body content in kBq, and these have been converted to Bq g^{-1}K assuming a K content for all individuals of 110 g. The measured values were compared with the levels expected from measurements of activity in foodstuffs and estimates of consumption rates. The measured values were mostly around 20% of the predicted values except for the four highest values where the measured quantity was between 20 and 100% of the predicted value. In three of these four cases the possibility of some intake arising from occupational exposure can not be ruled out.

TABLE 4.2.7 ^{137}Cs measurements of residents of Seascale, Cumbria (Fry and Sumerling, 1984)

		Number in range	
^{137}Cs body content (kBq)	^{137}Cs body content (Bq g^{-1}K)	Total	Number currently employed at BNFL Sellafield at the time of measurement
Activity not detected		269	35
≤0.6	≤5.5	17	3
0.6-1.5	5.5-14	3	3
1.6-3.7	15-34	1	0
> 3.7	> 34	0	0

After the Chernobyl accident, Fry and Britcher (1987) compared monthly mean values of body content of radiocaesium in two groups of people. One group (Section 4.1.2.2) was resident mainly in Oxfordshire, an area which received little deposition of Chernobyl radionuclides. The second group was resident in west Cumbria and included some individuals who may have been exposed to radiocaesium in the course of their work. However, measurements before the Chernobyl accident demonstrated that this source of exposure was insignificant compared to dietary intakes since the accident. The comparison covers the period from May 1986 to April 1987. The Oxfordshire group of 30 individuals show large variations between persons; extreme values differing by a factor of four depending on dietary habits and metabolism. The Cumbrian group is larger and also shows variations between persons of a factor of four over a period of one month. The Cumbrian data show a rise to a peak of 700 Bq (~6.4 Bq/gK) in June 1986 and a decline to 450 Bq (~4.1 Bq/gK) which was maintained between July 1986 and August 1987. In contrast the Oxfordshire group over the same period show a steady increase to 250 Bq (~2.3 Bq/gK) which was maintained from September 1986 to August 1987 (Figure 4.2.1).

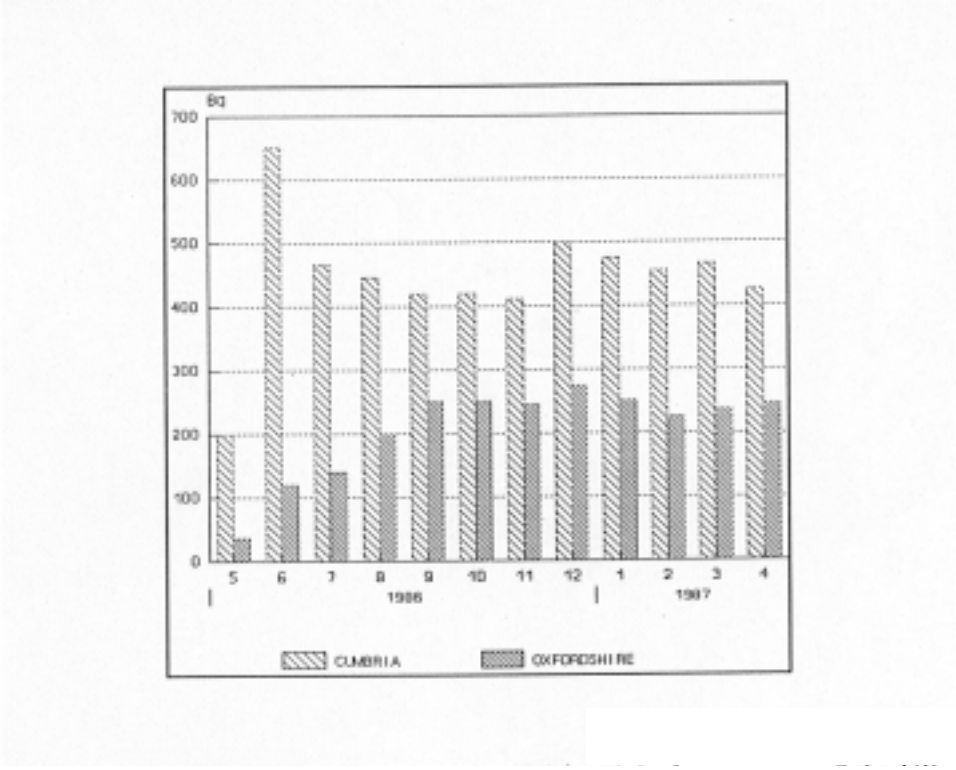

FIGURE 4.2.1 Comparison of ^{137}Cs levels in populations of Cumbria and Oxfordshire (Fry and Britcher, 1987)

During November 1987, measurements of the body content of radioactive caesium (^{134}Cs + ^{137}Cs) were made in Yorkshire on nearly 400 people, of whom about 250 lived in the Ilkley/Skipton area. These measurements were carried out in response to public concern that a 'hotspot' of Chernobyl fallout had been

overlooked (Bailey et al, 1987). In all but three cases no radiocaesium was detected. The detection limit was 370 Bq for an adult and 250 Bq for a child. The three measured values were all close to the limit of detection and were 340 ± 200 (child), 390 ± 250 Bq (adult) and 490 ± 250 Bq (adult). The results indicated that average radiocaesium levels in the measured population are less than those parts of the UK where extensive enhanced deposition of Chernobyl fallout and are similar to the body contents found in Southern and Eastern England.

On 7th May 1986 and again on 19/25th May, 6 volunteers were measured for whole body ^{137}Cs at Leeds (Oxby et al, 1986). The average result on 7th May was 30±21 Bq of ^{137}Cs in whole body (~0.27 Bq/gK) and 39±36 Bq (~0.35 Bq/gK) on 19/25th.

In 1986, the Department of the Environment (DoE) commissioned research to measure volunteer members of the public using whole body monitoring. Although originally intended to investigate the effects of Sellafield discharges the research began shortly before the Chernobyl accident. For this large study of ^{137}Cs and ^{134}Cs whole body contents of people in England, Scotland and Wales data were obtained from a mobile whole body counter and a fixed system at Whitehaven, Cumbria. In May 1988, 155 volunteers living in the Whitehaven area of Cumbria were measured (Boddy et al, 1989b). The results are reported in Table 4.2.8.

The average ratio of caesium-137 to caesium-134 was 3.28 at the time of measurement, corresponding to a ratio of 1.99 at the time of the Chernobyl accident. This is consistent with Chernobyl origin as the primary source for the radiocaesium. There were discernible but not marked trends of increasing body radiocaesium with milk and meat/fish consumption.

As part of the DoE funded study, between June 1987 and December 1988 a mobile whole body counter visited a number of sites throughout Northern England (Boddy et al, 1989a). Results from some installed counters at Medical Physics Departments were included. The mean ^{134}Cs values have been calculated using the measured ratios of ^{137}Cs/^{134}Cs for all age groups and the average measured ^{137}Cs value. This is necessary as in many cases the ^{134}Cs levels are below the limit of detection.

The measured data are summarised in Table 4.2.8.

TABLE 4.2.8 Whole body radiocaesium measured between June 1987 and December 1988 (Boddy et al, 1989a)

Location	Measurement date	Age	Sex	Number of volunteers	Mean ^{137}Cs (Bq)	Mean ^{134}Cs (Bq)	Average Cs/K
Barrow	April, 1988	≤ 5		3	33 (0.86)	11 (0.30)	0.038
		6-15		14	81 (1.2)	28 (0.42)	0.054
		>16	M	31	191 (1.3)	67 (0.45)	0.057
		>16	F	48	133 (1.3)	47 (0.46)	0.058
Blackpool	Sept, 1987	≤ 5		5	38 (1.1)	14 (0.41)	0.05
		6-15		30	107 (1.4)	39 (0.52)	0.063
		>16	M	51	276 (2.0)	102 (0.75)	0.091
		>16	F	57	176 (2.0)	65 (0.73)	0.088
Carlisle	Feb, 1988	≤ 5		4	40 (1.3)	15 (0.33)	0.057
		6-15		9	67 (1.2)	25 (0.43)	0.052
		>16	M	37	179 (1.3)	67 (0.47)	0.056
		>16	F	55	117 (1.2)	44 (0.46)	0.055
Hartlepool	June, 1987	≤ 5		4	60 (1.2)	23 (0.45)	0.053
		6-15		4	83 (1.9)	32 (0.75)	0.088
		>16	M	23	215 (1.5)	83 (0.58)	0.068
		>16	F	30	137 (1.5)	53 (0.56)	0.066
Isle of Man	Dec, 1988	6-15		10	60 (0.75)	16 (0.20)	0.031
		>16	M	86	223 (1.5)	61 (0.43)	0.065
		>16	F	53	134 (1.4)	36 (0.38)	0.058
Penrith	March, 1988	≤ 5		5	26 (0.76)	13 (0.38)	0.033
		6-15		6	33 (0.55)	16 (0.28)	0.027
		>16	M	24	156 (1.3)	78 (0.67)	0.065
		>16	F	39	105 (1.2)	53 (0.61)	0.059
Seascale	August, 1987	≤ 5		5	116 (2.0)	41 (0.69)	0.087
		6-15		48	140 (1.8)	50 (0.64)	0.08
		>16	M	48	456 (3.1)	161 (1.1)	0.138
		>16	F	86	286 (2.9)	101 (1.0)	0.126
Stockton	July, 1987	≤ 5		1	48 (1.2)	20 (0.47)	0.053
		6-15		2	69 (1.3)	28 (0.52)	0.058
		>16	M	29	218 (1.5)	89 (0.62)	0.07
		>16	F	60	160 (1.7)	65 (0.67)	0.076
Workington	May, 1988	≤ 5		1	52 (1.3)	17 (0.43)	0.057
		6-15		12	82 (0.95)	27 (0.31)	0.041
		>16	M	14	236 (1.7)	76 (0.54)	0.072
		>16	F	31	128 (1.3)	41 (0.44)	0.058
Middles-borough	August, 1988	≤ 5		10	17 (0.44)	4 (0.12)	0.018
		6-15		41	49 (0.75)	13 (0.20)	0.031
		>16	M	52	142 (1.0)	37 (0.26)	0.041
		>16	F	51	98 (0.95)	26 (0.25)	0.039
Whitehaven	May, 1988	≤ 5		6	62 (1.1)	19 (0.32)	0.045
		6-15		22	104 (1.1)	32 (0.35)	0.048
		>16	M	51	241 (1.9)	74 (0.57)	0.079
		>16	F	76	167 (1.7)	51 (0.52)	0.072

Note
Figures in parentheses are in Bq g^{-1} K and have been calculated using the authors Cs/K ratio and assuming an activity of 30.7 Bq per g K.

At any given site the radiocaesium levels are greatest for adult males and are progressively less for females, school children and pre-school children. The average radiocaesium/potassium activity ratios are less diverse over the four population subgroups than the measured radiocaesium activities.

Seascale (and Glasgow) have the greatest number of volunteers with a caesium/potassium activity ratio in excess of 25%. Generally central Scotland, North Wales and north-west England had twice the radiocaesium/potassium levels found in the rest of England and Wales (Boddy et al, 1989a).

Mean adult caesium/potassium activity ratios are shown in Figure 4.2.2 and are the ratios at the time of measurement.

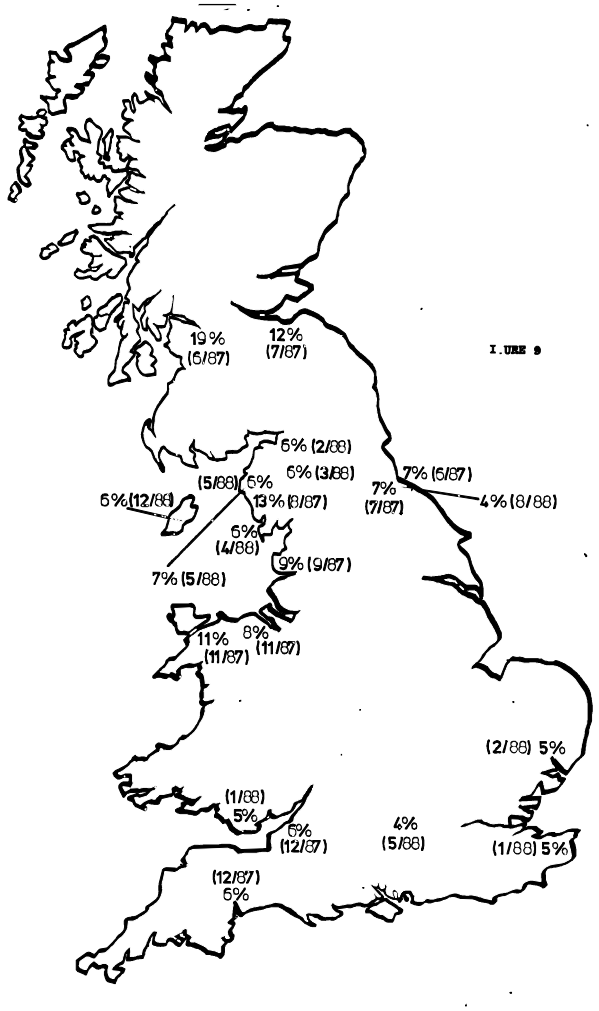

FIGURE 4.2.2 Caesium / Potassium activity ratios and measurement dates (Boddy et al, 1989a)

Extrapolation of the $^{137}Cs/^{134}Cs$ ratio to the date of the Chernobyl accident for all sites gives a value close to two, which is the value found in Chernobyl fallout.

Therefore the measured body caesium in this survey is consistent with an intake attributable primarily to fall-out from the Chernobyl accident. The variation in the range of average caesium/potassium ratios is well below the 20:1 measured variation in deposition, which presumably reflects a non-local component of diet.

Between May 1989 and May 1990 a second nationwide study of body radiocaesium was conducted (Fenwick et al, 1992) which visited many of the sites which were used in the first study between June 1987 and December 1988 (Boddy et al, 1989a). In fact 36% of the volunteers measured in the second survey were measured in the earlier survey. The ratio of ^{137}Cs/gK in the two surveys (first survey/ second survey) is shown in Table 4.2.9.

TABLE 4.2.9 Ratio of measured ^{137}Cs between the same location on the two surveys (Fenwick et al, 1992; Boddy et al, 1989a)

Site	Ratio of ^{137}Cs/K (first : second survey)
Barrow	2.10
Blackpool	5.40
Carlisle	2.74
Penrith	3.10
Seascale	3.15
Stockton	3.71
Workington	1.99

At each site a minimum of 80 subjects were measured. ^{134}Cs was found to be below the detection limit for almost all measurements. The levels of ^{137}Cs had fallen by a factor of between about 2 and 5 and there was at this time little difference in body activities around the UK. Seascale remained above average, but had shown the second largest fall in body activity. A few individuals who had measurable ^{134}Cs levels confirm the Chernobyl accident as the primary source of the radiocaesium.

4.2.2.3 Summary

In 1957 the levels of ^{137}Cs in residents near Windscale were about 3 times the values for residents of Berkshire and Oxfordshire. After the Windscale accident these levels increased by a factor of four.

Levels of ^{137}Cs measured in West Cumbria between 1961 and 1964 were double the corresponding values for residents of Berkshire and Oxfordshire.

The highest measured ^{137}Cs whole body contents were measured in 1977 in a study of local fish eaters.

The measured ^{137}Cs activities measured in Cumbrian residents between 1978 and 1989 were very variable prior to the Chernobyl accident. A peak was reached in 1985/1986 which was followed by a decrease in later years. The peak

in 1986 was due to the Chernobyl accident, and the high value in 1985 was based on only three measurements.

The $^{137}Cs/^{134}Cs$ ratios measured at BNFL Sellafield prior to the Chernobyl accident were in the range 11 to 30. This is consistent with the source being from nuclear fallout or discharges of radioactive waste from UK nuclear establishments (Hunt, 1988). Measurements after the Chernobyl accident are generally consistent with the primary source of radiocaesium being Chernobyl fallout.

In 1984 a NRPB study of residents in and around Seascale gave an average content of 90 Bq of ^{137}Cs which is about a factor of 2 lower than levels for Cumbrian residents measured by BNFL in the same year.

The ^{137}Cs whole body activities measured at Whitehaven in May 1986 in the DoE study were about half those measured in Cumbrian residents at BNFL. The DoE UK-wide study found that the highest levels of radiocaesium at Seascale, north-west England (along with Scotland and north Wales) having twice the radiocaesium/potassium activity levels found in the rest of the England and Wales. In 1989/90 the levels of ^{137}Cs had fallen by a factor of between 2 and 5 and at this time there was little difference in activities around the UK. However, levels at Seascale remained above average.

4.2.3 Iodine isotopes

4.2.3.1 Literature review

Following the announcement of the Windscale reactor accident of October 10th 1957, Burch made measurements of the ^{131}I content of thyroids for residents of Yorkshire and Cumbria (Burch, 1959). The most comprehensive sets of measurements were made of 2 residents of Leeds from October 15th 1957 to January 10th 1958 and this data is reproduced in Figure 4.2.3.

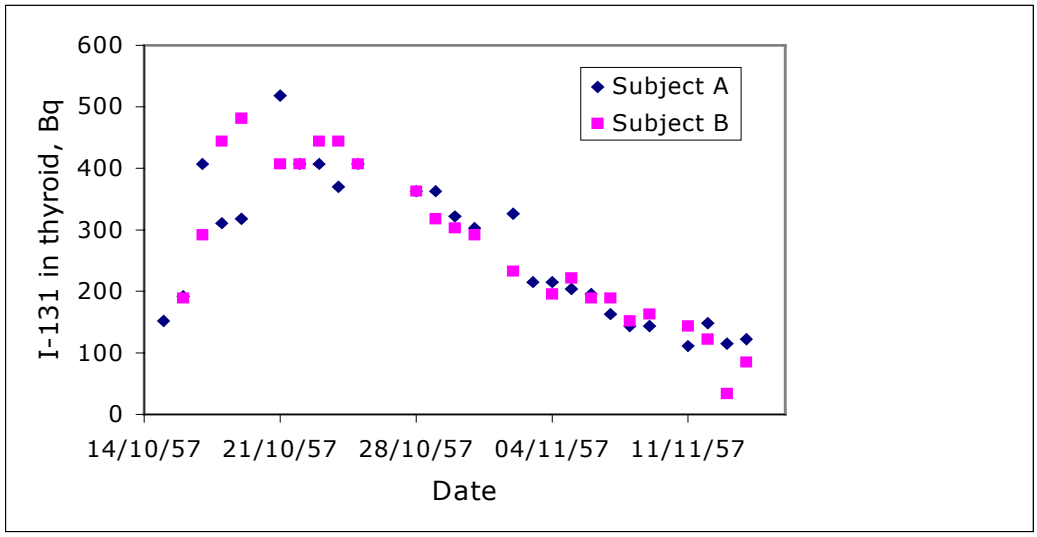

FIGURE 4.2.3 I-131 thyroid measurements of two residents of Leeds (Burch, 1959)

Results for eight other Leeds residents between 16th October and 23rd October gave a minimum value of 120 Bq and a maximum value of 960 Bq although this individual spent some time at Skipton before the measurement. In the same study 2 residents of Keswick were also measured on 18th November and gave results of 100 and 143 Bq (Burch, 1959). Another study of ^{131}I in thyroid carried out soon after the Windscale accident was made by Dunster et al. (1958). This study made was of children and adults living in the area surrounding Windscale and the results are shown in Table 4.2.10. These results were calculated assuming the intake occurred on 11th October and that the effective half-time of ^{131}I in the thyroid is 8 days.

TABLE 4.2.10 Thyroid activity for adults and children in the Windscale district (Dunster et al. 1958)

	Average activity in thyroid on 11th October 1957, Bq	
Location	Adults	Children
Beckermet	12,000 (11)	-
Seascale	3,300 (8)	-
Drigg	4,100 (3)	3,000 (2)
Holmrook	8,100 (6)	-
Ravenglass	12,000 (7)	19,000 (3)
Bootle	8,500 (12)	7,000 (11)
Millom	2,600 (19)	-
Ulverston	3,300 (5)	6,700 (3)
Barrow	5,900 (9)	-

Figures in brackets indicate the number of people examined

Between 1978 and 1989, BNFL Sellafield made a few measurements of ^{131}I in thyroid of residents of Cumbria (Peace, 2002). On 6/5/1986 560 and 930 Bq were measured in two residents of Carlisle and on 12/5/86 67Bq was measured in a resident of Workington.

The thyroid ^{131}I activity was measured in one adult resident of Leeds on 13th May 1986 (Despres, 1990). The result was 10 Bq.

In May 1986, six residents of Leeds were measured for ^{131}I by whole body counting or by thyroid counting. The results are shown in Table 4.2.11.

TABLE 4.2.11 ^{131}I content of Leeds residents (Fenwick et al, 1992)

Date, 1986	Measurement	Mean activity ± s.d (Bq)
6/7 May	whole body	73 ± 39
23 May	thyroid	7.8 ± 6

4.2.3.2 Summary

I-131 measurements made after the Windscale accident in Cumbria were much higher those of Leeds and London, although the London measurements were made later than at the other locations.

The measurements of ^{131}I in thyroid after the Chernobyl accident are similar to those made for residents of southern England, with the exception of the relatively high values measured on 6th May 2002 for two residents of Carlisle.

4.2.4 Plutonium and Americium isotopes

4.2.4.1 Literature review

No reports of measurements of plutonium and americium isotopes in residents of Northern England were found in the literature.

4.2.5 Uranium thorium and polonium isotopes

4.2.5.1 Literature review

No reports of measurements of uranium, thorium and polonium isotopes in residents of Northern England were found in the literature.

4.2.6 Other radionuclides

4.2.6.1 Literature review

Between 1978 and 1989, BNFL Sellafield made whole body measurements of residents of Cumbria who requested a measurement (Peace, 2002). These measurements were mainly of radiocaesium but ^{106}Ru was measured in a few subjects. The results are summarised in Table 4.2.12 with the ^{137}Cs measurement made at the same time. In addition ^{144}Ce and ^{60}Co were measured on one occasion each. On 16/10/84, 74 Bq of ^{144}Ce was measured in a resident of Egremont and on 28/6/84, 67 Bq of ^{60}Co was measured in a resident of Millom.

TABLE 4.2.12 ^{106}Ru measurements of Cumbrian residents (Peace, 2002)

Date of measurement	^{137}Cs, Bq	^{106}Ru, Bq
14/12/83	781	189
14/12/83	152	159
26/6/85	1070	303
26/11/83	31.4	152
9/1/84	633	107
9/12/83	11.1	155
26/11/83	747	185
26/11/83	511	178
5/12/83	2430	721

4.2.6.2 Summary

The average ^{106}Ru content for 9 measurements for Cumbrian residents made between 1983 and 1985 was about 240 Bq.

4.3 Scotland

This section covers measurements made on residents of Scotland, including the Scottish islands.

There are a large number of in vivo measurements for residents of Scotland. The vast majority of these are measurements of radiocaesium, although a small number of iodine-131 measurements were made immediately after the Chernobyl accident. In addition, measurements have been made of americium-241 in the skull from residents of the Caithness area.

The earliest identified in vivo measurements of radionuclides in residents of Scotland were made in 1961 and the most recent measurements were made in residents of the Caithness area in 1990/1991. There was a marked increase in measurements following the Chernobyl accident.

4.3.1 Strontium isotopes

4.3.1.1 Literature review

No reports of measurements of strontium isotopes in residents of Scotland were found in the literature.

4.3.2 Caesium isotopes

4.3.2.1 Introduction

The majority of in vivo measurements which have been made in Scottish residents are of ^{137}Cs in whole body. A small number of measurements were made before the Chernobyl accident and a much larger number in the years following the accident. After the Chernobyl accident a smaller number of ^{134}Cs in whole body were also made.

In general whole body caesium values are reported in the literature in terms of Bq of caesium per g of potassium rather than Bq kg^{-1} body weight. This is because there is a significant correlation between total body radiocaesium and total body potassium, whereas there is not a strong correlation between total body radiocaesium and body weight. In the case of older papers data expressed in Curies have been converted to Becquerels. In some references values are expressed as Bq of caesium per mmol of K or Bq of caesium per Bq of ^{40}K. In this report values are expressed in the units of Bq g^{-1} K. If the original paper does not use these units and there are no details of subjects' potassium content then values from ICRP have been used. For adult males a value of 133 g K is assumed and for adult females 87 g (ICRP, 1975). In cases where males and females are not treated separately then an average adult value of 110 g is used. Potassium contents for children have also been taken from ICRP where necessary (ICRP, 1975).

As far as possible the literature review is in order of measurement date with the oldest measurements being discussed first.

4.3.2.2 Literature review

The first reported measurements of ^{137}Cs in members of the public were made in 1961 and 1963 at Dounreay (Vennart, 1964). In 1961 at the Dounreay reactor establishment the average ^{137}Cs/K ratio was 3.4 Bq/g. In 1963 when there was a large increase in fuel processing the average value was 10.8 Bq/g.

Caesium-137 levels relative to whole body potassium were measured in both healthy and diseased inhabitants of Scotland in June 1978 to March 1979 (Williams et al, 1981). Two groups of adults were studied. The first group was patients with hypertension or with renal disease whose body composition was being investigated. These were residents of mainland Scotland (59 subjects), the majority from the Glasgow area and 5 from the Hebrides Islands. The second group consisted of male laboratory staff from the Glasgow area who were in good health (10 subjects). The average weekly fish consumption for each of the subjects was also recorded. The data for residents of mainland Scotland are reported in Table 4.3.1.

TABLE 4.3.1 Mean Whole body ^{137}Cs / potassium ratio and fish consumption in inhabitants of mainland Scotland, Bq g^{-1}K (Williams et al, 1981)

	Fish consumption			
	Rarely or never	less than once/week	once/week	more than once/week
Healthy subjects	0.57	0.90	1.0	0.72
Patients	0.46	0.75	1.1	1.4

The authors report a significant correlation between the ^{137}Cs/K ratio and the number of times per week that fish is eaten. Five subjects from the Hebrides Islands had higher mean ^{137}Cs values of 5.5 Bq g^{-1}K. These values were compared to an average level measured in central southern England in January 1977 of 0.33 Bq g^{-1}K which is similar to the average for Scottish non-fish eaters. The authors conclude that the most likely explanation is the ^{137}Cs content of fish eaten locally.

Isles et al (1991) made measurements of ^{137}Cs in patients suffering from hypertension who were having electrolyte concentrations measured. Most patients came from the Glasgow area but 60 were from the Western Isles, and 101 from other parts of mainland Scotland. The underlying diseases were mainly essential hypertension, renovascular disease, and primary hyperaldosteronism. The measurements were made between 1979 and 1986 prior to the Chernobyl accident. The measurement results are shown in Table 4.3.2.

TABLE 4.3.2 Median whole body ^{137}Cs/K ratios for residents of various regions of Scotland (Isles et al, 1991)

Place of residence	No. of subjects	Median Cs-137 in body (Bq g^{-1}K)	Interquartile range (Bq g^{-1}K)
Western Isles	60	2.54	1.25-3.73
Glasgow	218	0.47	0.26-0.66
Other parts of Scottish mainland	101	0.42	0.24-0.71

Patients from the Western Isles had five times higher concentrations of ^{137}Cs than did those from other areas. Concentrations higher than 2 Bq g^{-1}K were found in 35 of 60 islanders but in only one of 218 Glaswegians. Apart from patients with primary hyperaldosteronism (22 patients) there was no evidence that ^{137}Cs concentrations differed in patients with different types of hypertension.

Measurements of ^{137}Cs whole body measurements have been made at the UKAEA establishment at Dounreay for subjects employed at UKAEA but not involved with contact with radioactive material (Newton, 2002). These measurements were made between June 1978 and September 1982. The results are summarised in Table 4.3.3.

TABLE 4.3.3 Whole body ^{137}Cs/K ratios for Dounreay employees (Newton, 2002)

Date of measurements	Number of measurements	Mean ^{137}Cs/K (Bq g^{-1}K)
June 1978-March 1979	64	0.8
April 1979-September 1982	145	0.53
June 1978-June 1980	70	0.8
July 1980-September 1982	139	0.52
June 1978-December 1980	84	0.76
January 1981-September 1982	125	0.51

The measurements from Dounreay show a downward trend in nuclear weapon fallout ^{137}Cs levels in the subjects since June 1978.

In 1986, the Department of the Environment commissioned research to measure volunteer members of the public using whole body monitoring. Although originally intended to investigate the effects of Sellafield discharges, the research began shortly before the Chernobyl accident. This large study of ^{137}Cs and ^{134}Cs whole body contents of people in England, Scotland and Wales data was obtained from installed whole body counters at Glasgow and Edinburgh (Boddy et al, 1989a; McKenzie et al, 1989). The measured ratio of ^{137}Cs/^{134}Cs was close to 2.0 and the authors conclude from this is that the primary source for the radiocaesium was the Chernobyl accident. In this report values are reported as Bq in whole body or as the ratio of Bq of radiocaesium to Bq of potassium-40. This has been done as the dose delivered per unit activity of ceasium-137 or

caesium-134 in soft tissues is similar to that delivered per unit activity of potassium-40. The results are reported in Table 4.3.4 and Table 4.3.5.

TABLE 4.3.4 Results for Edinburgh volunteers for July 1987 from (Boddy et al, 1989a)

Age	Sex	Number of volunteers	Mean ^{137}Cs (Bq)	Mean ^{134}Cs (Bq)	Total Cs	Range of total Cs (Bq)	Average Cs/K
6-15		1	283.5 (3.0)*	111.9 (1.2)	395.4	372-372	0.138
>16	M	18	398.2 (2.8)	157.17 (1.1)	555.37	292-835	0.129
>16	F	11	254.55 (2.5)	100.47 (1.0)	355.02	214-846	0.115

* Figures in parenthesise are in Bq g^{-1}K and have been calculated using the authors Cs/K ratio and assuming an activity of 30.7Bq per g K.

TABLE 4.3.5 Results for Glasgow volunteers for June 1987 from (Boddy et al, 1989a)

Age	Sex	Number of volunteers	Mean Cs-137 (Bq)	Mean Cs-134 (Bq)	Total Cs	Range of total Cs (Bq)	Average Cs/K
>16	M	20	606.85 (4.4)*	247.89 (1.8)	854.74	246-2198	0.200
>16	F	20	382.55 (4.2)	156.27 (1.7)	538.82	202-860	0.188

* Figures in parenthesise are in Bq g^{-1}K and have been calculated using the authors Cs/K ratio and assuming an activity of 30.7Bq per g K.

The author's note that in all of the areas monitored in England, Scotland and Wales, Glasgow (and Seascale) have the greatest number of volunteers with caesium/potassium activity ratios exceeding 25%. The measured activity ratio of ^{137}Cs/^{134}Cs for both sites are within 15% of the expected Chernobyl ratio.

Residents of the Glasgow area, 13 men and 5 women, underwent ^{137}Cs in whole body measurements four times in the year following the Chernobyl accident. (Watson, 1986; 1987). A larger group of 40 adults was measured at one-year post-Chernobyl. The average measured values for the 40 adults reached a plateau 3-4 months after the Chernobyl accident. The average plateau value for ^{137}Cs was approximately 0.17 Bq/mmol K (4.3 Bq g^{-1}K) and 0.06 Bq/mmol K (1.5 Bq g^{-1}K) for ^{134}Cs. Three males from the same area measured in 1983 (before the accident) had a ratio of 0.037 Bq/mmol (0.9 Bq g^{-1} K) of ^{137}Cs. Watson (1990) has shown that these measurements of whole body radiocaesium could have been predicted by the radiocaesium content of milk.

East and Robertson (1988; 1990a; 1990b) measured ^{137}Cs and ^{134}Cs in members of the Scottish population between May 1986 and February 1988. Some 250 men, women and children were examined, in many cases sequentially. The mean maximum body content ^{137}Cs level was 3.8 Bq g^{-1}K with a upper boundary value of 6.2 and a lower boundary value of 1.6 Bq g^{-1}K. These levels were observed 8 months after the accident. The corresponding values for ^{134}Cs were 2.8, 1.7 and 0.7 Bq g^{-1}K respectively. For 80% of those measured the ^{137}Cs level varied by a factor of four. The results are summarised in Table 4.3.6. There was no evidence that age and sex influenced the radiocaesium content. However levels were higher in the West and Southwest (Dumfries, Galloway and West Strathclyde)

and in the northern and north-eastern regions (Highland, Grampian, Tayside and Islands) of the country where deposition was known to be higher.

East et al (1992) continued making whole body ^{137}Cs measurements on residents of Scotland between March 1988 and March 1990. For this study 216 measurements were made of radiocaesium in members of the Scottish population. The influence of sex, age, diet and geographical location on ^{137}Cs uptake was re-examined and it was found that: there was no difference between males and females; only game and venison eaters showed distinctly higher body radiocaesium compared with the rest of the population; and only marginal differences due to geographical location were observable. The results for each geographic region are summarised in Table 4.3.6.

TABLE 4.3.6 Mean body levels of Cs-137 (Bq g^{-1}K) in relation to area of Scotland (age>18 years) (East and Robertson, 1988; East et al, 1992)

Period	West and S West	Central	North and N East	East and S East
Mar/Apr 1987	4.19	3.30	3.25	2.78
May/Jun 1987	-	3.36	5.02	2.67
Sep/Oct 1987	2.95	2.39	2.68	1.79
Nov/Dec 1987	1.97	2.06	2.56	-
March/ April 1988	1.93	0.99	1.59	-
July 1989	0.66	-	0.16	-
August 1989	-	-	0.5	-
September 1989	-	0.61	0.64	-
Oct/Nov 1989	0.61	0.44	0.74	0.56
November 1989	0.72	0.37	0.60	0.58
March 1990	0.62	0.51	0.65	0.71

Between October 1990 and December 1991 Watson and Sumner (1996) measured ^{134}Cs and ^{137}Cs in residents of two areas of Scotland, the Caithness area (66 subjects) adjacent to the Dounreay Nuclear Establishment and the central western area (56 subjects) which is remote from any nuclear establishment. The ^{134}Cs whole body activity was essentially undetectable in all of the subjects. The ^{137}Cs whole body activity ranged from below the limit of detection to 315 Bq (~2.9 Bq g^{-1} K). To compare the groups the radiocaesium values were normalised to total body potassium to help eliminate group differences associated with different body habitus. When this was done no significant inter-group differences were identified for ^{137}Cs.

4.3.2.3 Summary

Inhabitants of mainland Scotland were first measured for ^{137}Cs in 1961. Non-fish eaters had values close to measurements for southern England while regular fish eaters had 2 or 3 times the levels for southern England. Residents of the Hebrides Islands had higher ^{137}Cs levels than residents of the Scottish mainland.

Measurements of residents of the Western Isles between 1979 and 1986 showed ^{137}Cs contents which were 5 times those for the Scottish mainland. The Scottish mainland residents had ^{137}Cs levels which were approximately double that of residents of southern England at this time.

Subjects employed at Dounreay who were measured between 1978 and 1982 also had ^{137}Cs levels a factor of 2 higher than residents of southern England.

Post-Chernobyl levels of radiocaesium in Edinburgh and Glasgow were among the highest found in the UK. The peak levels of ^{137}Cs for Glasgow were about double the corresponding values for southern England. A study of post Chernobyl ^{137}Cs levels in people across Scotland showed higher levels where ground deposition levels were known to be higher.

Measurements of residents near the Dounreay nuclear establishment in 1990/1991 gave a maximum measured ^{137}Cs content which was a factor of 10 higher than a mean value for southern England at the same time.

4.3.3 Iodine isotopes

4.3.3.1 Introduction

Two papers report values for ^{131}I in the thyroid for residents of Scotland, both of which are for measurements carried out immediately after the Chernobyl accident. No measurements have been found for any other iodine isotope.

4.3.3.2 Literature review

Following the Chernobyl accident, ^{131}I uptake was measured in the thyroid of 11 volunteers from the Glasgow area (East and Robertson, 1990b). Measurements were made from 10 to 50 days after the accident. The average measured value was 16 Bq in the Glasgow area with a maximum observed value of 30 Bq. One volunteer who used a mountain water supply had a clearly higher value of 57.5 Bq. A series of measurements on one individual show a peak of activity about 20 days after the Chernobyl accident.

Fry and Milton measured ^{131}I activity in the thyroid of over 100 residents of Southern Scotland immediately after the Chernobyl accident (Despres, 1990). The results are given in Table 4.3.7.

TABLE 4.3.7. ^{131}I in the thyroid of residents of Southern Scotland 1990. (Despres, 1990)

			Results below the detection limit		Results above the detection limit		
Place of residence	Date of measurement	Age	Number	Detection limit	Number	Mean	Range
Glasgow	14/5/1986	>20			1	15	
Glasgow	16/5/1986	6	13	100	3	130	100-190
Castle Douglas	28/5/1986	15-17	15	60	4	89	79-98
Kirkcudbright	29/5/1986	>20	5	100			
Kirkcudbright	29/5/1986	15-17	15	100	6	120	100-140
Duns	30/5/1986	>20	7	100			
Duns	30/5/1986	13-14	18	100	1	110	

The maximum amount of ^{131}I measured in the thyroid of a Scottish resident after the Chernobyl accident was 190 Bq, 20 days after the Chernobyl accident on 26th April 1986. The majority of ^{131}I measurements made after the accident were less than 100 Bq (Despres, 1990).

4.3.3.3 Conclusion

The maximum ^{131}I thyroid content for measurements carried out in Scotland after the Chernobyl accident was 190 Bq. This is much higher than values for southern England where the maximum measured value was 28 Bq.

4.3.4 Plutonium and Americium isotopes

4.3.4.1 Introduction

Only one paper describing measurements of americium-241 in residents of Scotland was found in the literature. There were no references describing measurements of plutonium isotopes in members of the public. This probably is because of the need for specialist measurement equipment and the relatively high detection limits compared with, for example, caesium-137.

4.3.4.2 Literature review

Between October 1990 and December 1991 Watson and Sumner (1996; 1997) carried out measurements of ^{241}Am in the skulls of 66 residents of the Caithness area of Scotland living close to the Dounreay Nuclear Establishment. All of the subjects measured had no detectable ^{241}Am. The MDA (minimum detectable activity) for these measurements was 3.8 Bq (95% confidence interval) which corresponds to less than 24 Bq in the total skeleton. The 66 subjects consisted of 4 children with leukaemia, 7 case siblings, 16 case parents, 13 case control children and 26 case control parents. Measurements were also made of 56 residents from West Central Scotland, an area remote from nuclear reprocessing

plants. Results for these subjects were not distinguishable from those for the Caithness residents.

4.3.4.3 Summary

Measurements were unable to detect ^{241}Am in 66 residents of the Caithness area of Scotland.

4.3.5 Uranium thorium and polonium isotopes

4.3.5.1 Literature review

No reports of measurements of uranium, thorium and polonium isotopes in residents of Scotland were found in the literature.

4.3.6 Other isotopes

4.3.6.1 Literature review

No reports of measurements of any other isotopes in residents of Scotland were found in the literature.

4.4 Wales

This section covers measurements made on residents of Wales.

There are a small number of in vivo measurements for residents of Wales, and most of these are measurements of radiocaesium made after the Chernobyl accident. A small number of iodine-131 measurements were also made on residents of Wales after the Chernobyl accident. All measurements found in the literature were carried out between May 1986 and May 1990.

4.4.1 Caesium isotopes

4.4.1.1 Literature review

Between May 6th and 2nd July 1986 after the Chernobyl accident, 6 volunteers were measured on five occasions at University Hospital of Wales in Cardiff (Burch and Owen, 1986). The volunteers were all members of staff. The mean values are shown in Table 4.4.1.

TABLE 4.4.1 Mean whole body ^{137}Cs content of six volunteers (Burch and Owen, 1986)

Date of measurement	Mean ^{137}Cs content, Bq	Mean ^{137}Cs/K (Bq/g)*
6 May 1986	43	0.39
12 May 1986	36	0.33
2 June 1986	55	0.50
18 June 1986	76	0.69
2 July 1986	105	0.95

* Assuming an average K content of 110 g (ICRP, 1975)

A number of local people resident in the Swansea area (members of the Medical Physics Department, Singleton Hospital, Swansea) were measured two weeks and eight weeks after the Chernobyl accident (Ali et al, 1986). After two weeks, there was no increase in the body activities, although at eight weeks increases in the body content of ^{137}Cs and ^{134}Cs were found (Table 4.4.2). Figures in paranthesis are in Bq/gK. The increases in ^{137}Cs ranged from 45 Bq for one person who did not drink milk to 520 Bq for another who relied mainly on lamb as his source of meat.

TABLE 4.4.2 Body radioactivity measurements in different subjects measured at Singleton Hospital, Cardiff eight weeks after the Chernobyl accident (Ali et al, 1986)

Subject	^{137}Cs, Bq	^{134}Cs, Bq
1	201 (1.8) *	92 (0.84) *
2	94 (0.85)	70 (0.64)
3	76 (0.69)	38 (0.35)
4	45 (0.41)	-
5	520 (4.7)	286 (2.6)

* Cs/K ratios in Bq g^{-1} are given in parentheses assuming an average K content of 110 g (ICRP, 1975)

In 1986, the Department of the Environment (DoE) commissioned research to measure volunteer members of the public using whole body monitoring. Although originally intended to investigate the effects of Sellafield discharges the research began shortly before the Chernobyl accident. This large study of ^{137}Cs and ^{134}Cs whole body contents of people in England, Scotland and Wales data was carried out using installed whole body counters at Cardiff and a mobile system which visited Rhyl and Bangor (Boddy et al, 1989a; McKenzie et al, 1989).

Preliminary results for Bangor and Rhyl were also published earlier (Boddy et al, 1988; Despres, 1990).

The measured data are summarised in Table 4.4.3.

TABLE 4.4.3 whole body radiocaesium measured between November 1987 and January 1988 (Boddy et al, 1989a)

Location	Measurement date	Age	Sex	Number of volunteers	Mean ^{137}Cs (Bq)	Mean ^{134}Cs (Bq)	Average Cs/K activity ratio
Bangor	Nov, 1987	≤ 5		10	76 (1.9)*	26 (0.66)	0.085
		6-15		20	92 (1.5)	31 (0.51)	0.066
		>16	M	65	359 (2.6)	123 (0.88)	0.112
		>16	F	70	221 (2.3)	76 (0.78)	0.100
Rhyl	Nov, 1987	≤ 5		3	96 (1.8)	34 (0.65)	0.081
		6-15		9	108 (1.5)	38 (0.51)	0.063
		>16	M	37	238 (1.8)	83 (0.61)	0.077
		>16	F	55	162 (1.8)	57 (0.64)	0.080
Cardiff	Jan, 1988	6-15		18	78 (0.89)	23 (0.27)	0.038
		>16	M	20	153 (1.0)	46 (0.31)	0.044
		>16	F	20	112 (1.1)	34 (0.34)	0.048

* Cs/K (Bq/g) ratios are given in parentheses.

At any given site the radiocaesium levels were greatest for adult males and became progressively less for females, school children and pre-school children. The average radiocaesium/potassium activity ratios are much more similar over the four population subgroups.

In this UK-wide survey Seascale and Glasgow had the greatest number of measured caesium/potassium ratios, in excess of 25%. However, Bangor had the highest recorded individual level in the survey (91%).

Between May 1989 and May 1990 a second nationwide study of body radiocaesium was conducted (Fenwick et al, 1992) which visited many of the sites which were used in the first study between June 1987 and December 1988 (Boddy et al, 1989a). In fact, 36% of the volunteers measured in the second survey were measured in the earlier survey. The ratio of ^{137}Cs/gK in the two surveys (first survey/ second survey) is shown in Table 4.4.4.

TABLE 4.4.4 Ratio of ^{137}Cs to potassium for two nation-wide surveys (Fenwick et al, 1992)

Site	Ratio of ^{137}Cs/K on the first and second surveys
Bangor	4.42
Rhyl	4.00

At each site a minimum of 80 subjects were measured. ^{134}Cs was found to be below the detection limit for almost all measurements. The levels of ^{137}Cs in Wales had fallen by a factor of about 4. This was similar to the decreases found throughout the UK of between about 2 and 5. At the time of the second survey there was little difference in body activities around the UK.

4.4.1.2 Summary

Average ^{137}Cs whole body contents for Cardiff residents in June 1986 were similar to those measured in southern England. However by July 1986 the levels of radiocaesium in people were approximately double those for southern England.

The DoE nation-wide survey showed levels of radiocaesium in north Wales that were double those in south Wales. Measurements made in the south were similar to sites across southern England and north Wales had radiocaesium levels in people which were typical of northern England.

4.4.2 Iodine isotopes

4.4.2.1 Introduction

A few measurements of ^{131}I in thyroid have been reported following the Chernobyl accident.

4.4.2.2 Literature review

Between May 6th and 2nd July 6, 1986 volunteers were measured on five occasions at University Hospital of Wales in Cardiff for ^{131}I in whole body (Burch and Owen, 1986). The volunteers were all members of staff. The mean values are shown in Table 4.4.5.

TABLE 4.4.5 Mean whole body ^{131}I content of six volunteers (Burch and Owen, 1986)

Date of measurement	Mean ^{131}I content, Bq
6 May 1986	11
12 May 1986	28
2 June 1986	17
18 June 1986	13
2 July 1986	11

4.4.2.3 Summary

The mean ^{131}I in whole body reached a peak in Cardiff on 12th May 1986 of 28 Bq. This is similar to measurements of ^{131}I in thyroid made in southern England at this time.

5 TEETH MEASUREMENTS

5.1 Introduction

In recent years there has been much concern about the possible harmful effects of radioactive releases to the environment. Several studies have reported

increased incidences of cancer in the vicinity of nuclear installations (Independent Advisory Group, 1984; Committee on Medical Aspects of Radiation in the Environment, COMARE, 1988; 1989). Among recommendations of these reports was that studies should be carried out to monitor the general public for exposure to long-lived radionuclides.

For legal and social reasons the collection and analysis of autopsy samples for assessing their radionuclide content is extremely difficult. Since teeth are an extension of the skeleton and accumulate stable and radioactive bone seeking elements that enter the body, the analysis of teeth provides a viable alternative to the analysis of bone samples but with some limitations, as described below.

The advantages of analysing teeth are that they are easily collected from orthodontists and can be used as a basis for investigating regional uptakes of the actinides and other bone seeking radionuclides. These assays are also important for establishing whether the concentrations increase appreciably in the vicinity of nuclear power and reprocessing plants. Recent developments of analytical techniques, such as low level alpha spectrometry and mass spectrometry, have increased the potential for this type of analysis.

Studies with radium-226 (Yamamoto et al, 1994), lead (Lovaas and Hursh, 1968; Stack, 1999) and other elements, have indicated that their levels in teeth are related to their skeletal content. Post mortem measurements of former workers undertaken by the USTUR have been carried out to determine if either the concentration or total quantity of an actinide element in the teeth could be correlated with the total skeletal content of that actinide (Kathren, 1995). In general the concentration ratios of the actinide in each tooth compared with the mean skeletal concentration was reasonably consistent for an individual but varied appreciably between individuals. No correlation appeared to exist between total actinide in the skeleton and the activity in the teeth or between the average skeletal concentration and the concentration in the teeth (Kathren, 1995). Hence it would appear that the teeth do not provide a reliable means of determining the total skeletal content or average concentration of plutonium and americium and thus cannot be used as a surrogate for the skeletal burden of the actinides. Despite this, teeth may still provide an alternative to analysing bone samples for the study of geographical variations in bone-seeking radionuclide contamination.

5.2 Strontium isotopes

A number of studies have investigated the concentrations of ^{90}Sr in children's teeth collected throughout the United Kingdom and Ireland.

Starkey and co-workers reported a survey of the accumulation of ^{90}Sr in human teeth for the Royal Navy. It covered the years 1959 – 71 in which fallout levels rose to a peak and declined again. Teeth were collected from all parts of the UK and teeth collected from areas with more than 40 inches mean annual rainfall were analysed separately.

Starkey and Fletcher (1969) reported that the results for ^{90}Sr accumulation in permanent teeth in persons aged 9-21 showed that the uptake was related to levels prevailing in the diet as a result of atmospheric pollution. The rate of fallout was negligible before 1953 but between 1954 and 1964, the world-wide deposition of ^{90}Sr increased from 3.7×10^{15} to 4.28×10^{17} Bq. This was reflected by the ratio of strontium-90 to calcium rising in the teeth of individuals over time. This trend is clearly shown in Table 5.2.1.

TABLE 5.2.1 Mean values of ^{90}Sr (mBq g^{-1} Ca) in premolars of individuals. Cohorts born between 1947 and 1956 (Starkey and Fletcher, 1969)

Extracted (born)	1960	1961	1962	1963	1964	1965
1947	7.77					
1948	9.99	11.1				
1949	12.2	13.7	14.4			
1950	13.7	16.3	17.0	20.0		
1951	15.2	20.7	21.1	21.5	29.2	
1952		28.9	25.5	24.8	36.3	37.7
1953			32.9	31.5	38.9	42.6
1954				39.2	44.4	51.8
1955					62.9	59.2
1956						68.5

Between 1960 and 1965, the ^{90}Sr concentration increased in all age groups. At any given time, the concentrations were higher in the younger age groups. This was because the teeth were in an active state of mineralisation and dietary levels were increasing.

TABLE 5.2.2 Strontium in human premolars (mBq g^{-1} Ca) 1959-1965 (Starkey and Fletcher 1969)

Extracted (age, years)	1960	1961	1962	1963	1964	1965
Whole tooth						
9	15.2	28.0	32.9	39.2	62.9	68.5
10	13.7	20.7	25.5	31.5	44.4	59.2
11	12.2	16.3	21.1	24.8	38.9	51.8
12	9.99	13.7	17.0	21.5	36.3	42.6
13	7.77	11.1	14.4	19.9	29.2	37.7
Roots						
9	40.7	51.8	51.8	51.8	114.7	122.1
10	37.0	41.4	39.9	44.4	66.6	96.2
11	33.7	35.9	38.9	36.3	62.9	88.8
12	28.5	35.2	34.0	39.2	53.7	70.3
13	24.4	28.1	31.1	38.5	51.8	64.8
Crowns						
9	10.7	23.7	28.5	34.8	51.8	53.7
10	10.4	14.8	21.5	27.0	38.5	46.3
11	7.03	9.99	15.5	20.7	30.7	39.9
12	4.81	6.66	10.7	14.8	29.9	32.2
13	3.33	4.07	7.03	12.6	20.4	27.0

Table 5.2.2 shows that the concentrations (mBq g^{-1} Ca) were higher in the roots than in the crowns in any given age group because the roots were affected by recent uptake but the crowns had mineralised when the dietary intakes were lower. Starkey and Brooks (1974) reported that between 1966 and 1971, while environmental levels of Sr-90 were falling, crown values rose towards the 1966 peak and root values began to fall away from the peak. The authors suggested that interesting correlations emerged between the uptake of ^{90}Sr in developing teeth and developing bone. Bone seeking elements are fixed in teeth by irreversible deposition, and therefore tooth ratios (mBq g^{-1} Ca) reflect original ratios that prevailed in the skeleton at the time the teeth were being laid down. Table 5.2.3 shows the relationship between premolar root values for 10- and 11-year-old groups and UKAEA posthumous bone returns for persons aged 5-19. The bone results show quicker responses to changing nuclear yields but the authors suggest an unmistakable underlying relationship between bone and teeth.

TABLE 5.2.3 Comparison of ^{90}Sr (mBq g^{-1} Ca) in tooth and bone (Starkey and Brooks 1974)

	Posthumous bone	Premolar roots	
	Age 5-19	Age 10	Age 11
1958	27.8	-	-
1959	33.3	-	-
1960	40.7	37	33.7
1961	38.9	41.4	35.9
1962	43.3	40.0	38.9
1963	56.6	44.4	36.3
1964	81.4	66.6	62.9
1965	99.9	96.2	88.8
1966	96.2	103.6	96.2
1967	77.7	103.6	98.1
1968	74.0	85.1	90.7
1969	66.6	70.3	77.7
1970	59.2	64.8	59.2
1971	-	57.4	57.4

The concentrations of ^{90}Sr in the teeth of Czechoslovakian children of different age groups (3-6 year-old group, 107.3 mBq g^{-1} Ca, 6-9 y old group, 92.5 mBq g^{-1} Ca and adults 51.8 mBq g^{-1} Ca) extracted in 1964, are in broad agreement with the values quoted by Starkey (Santholzer and Knaifl, 1966).

Premolar teeth from children were collected around the UK by Area Health Authorities and analysed for ^{90}Sr content (Priest et al, 1992). The concentrations of ^{90}Sr, expressed as mBq g^{-1} Ca, from over 70 batches, consisting of 8-13 teeth per batch, ranged from 8 to 120. There was a large variability within counties (Priest, 1992). However averages over larger regions were remarkably constant, as summarised in Table 5.2.4. The data did not correlate with rainfall statistics for the regions and increased deposition of ^{90}Sr in weapons test fallout did not

lead to elevated levels in teeth. The authors concluded that there was no evidence of increased levels in the vicinity of the nuclear fuel reprocessing plant at Sellafield.

TABLE 5.2.4 Summary of Strontium-90 results in Great Britain (Priest et al 1992)

	^{90}Sr (mBq g^{-1} teeth)		^{90}Sr/Ca (mBq g^{-1} Ca)	
Region	Mean	1 σ	Mean	1 σ
Scotland	9	4	34	16
N E England	9	6	38	28
N W England	9	4	35	19
Wales	7	5	26	18
Midlands	7	3	26	12
S E England	7	4	27	16
S W England	10	6	37	24
Average UK	9	5	34	20

The assay of ^{90}Sr in the teeth of children resident in the UK and Ireland have also been reported by O'Donnell et al. (1997). The average concentrations of ^{90}Sr were 8 Bq kg^{-1} (ashed weight). Analysis of the data showed that the strontium concentrations were log-normally distributed with each distribution biased towards lower strontium concentrations. It follows that the median concentrations were lower (6 Bq kg^{-1} ± 2 (SD)) for most individuals.

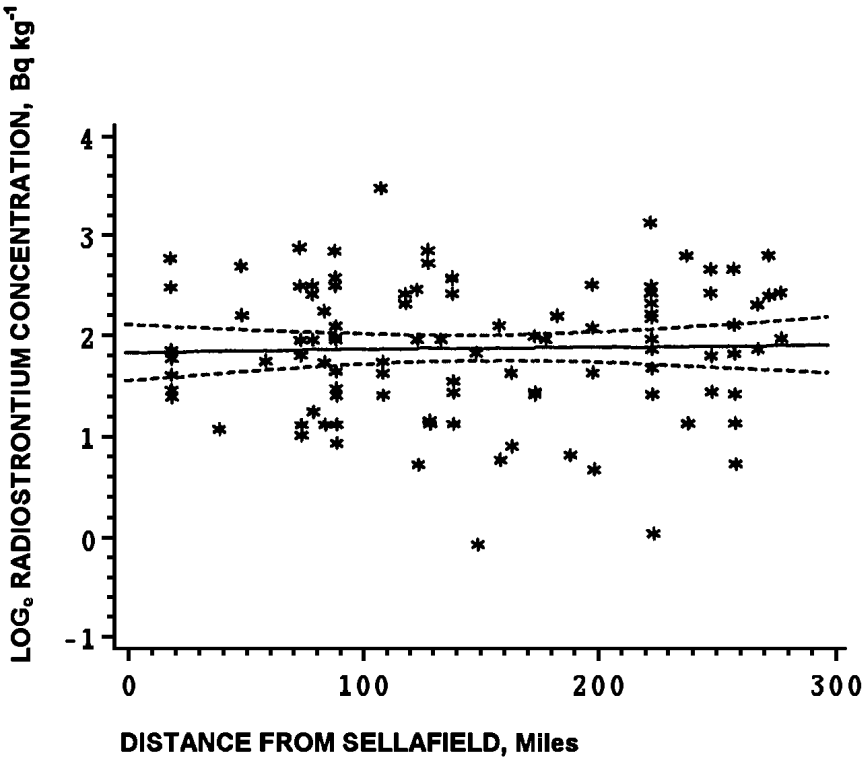

FIGURE 5.2.1 ^{90}Sr content of children's teeth vs. distance from Sellafield. Regression equation: $\log_e [^{90}Sr] = 1.8338 + 0.00003x$ Distance from Sellafield (miles). Statistical significance: P<0.9741. (Reproduced from O'Donnell et al 1997).

The result of the regression analysis (Figure 5.2.1) showed that no correlation existed between the levels of ^{90}Sr in teeth and the distance from Sellafield. The authors suggest that this is because the major route of uptake of this radionuclide is through milk, most of which is redistributed throughout the UK and is mixed before consumption by the public. The ^{90}Sr intakes arising from the waste stream of the Sellafield plant were considered to be of minor importance in comparison.

5.3 Caesium isotopes

No references found.

5.4 Iodine isotopes

No references found.

5.5 Plutonium and americium isotopes

The presence of plutonium in the environment may be attributed largely to fall-out from the atmospheric testing of nuclear weapons during the 1950's and 1960's. Other sources include routine and accidental discharges from various nuclear installations.

Several studies have attempted to determine the comparative exposures to $^{239+240}$Pu of different geographically located child populations of the UK. In this context concentrations of these Pu and other actinides have been measured in teeth removed for orthodontic purposes.

Long et al. (1991) attempted to correlate the concentrations of $^{239+240}$Pu in children's teeth with mean annual rainfall and linear distance from the BNFL reprocessing plant at Sellafield. Table 5.5.1 shows the concentrations of $^{239+240}$Pu found in teeth in different regions of the UK. Table 5.5.2 compares teeth from West Cumbria with those from the rest of the UK

TABLE 5.5.1 $^{239+240}$Pu concentrations in teeth (Long et al 1991)

Location	Concentration $^{239+240}$Pu (mBq kg^{-1}, ash)	Location	Concentration $^{239+240}$Pu (mBq kg^{-1}, ash)
Durham	3.9 ± 0.5*	Glasgow	1.0 ± 0.3
Bristol and Weston	3.5 ± 0.5	Lothian	0.7 ± 0.2
Gateshead	4.9 ± 0.9	South Glamorgan	1.1 ± 0.3
Leicestershire	12 ± 2	South Cumbria	<5
Dewsbury	21 ± 3	Tyne and Wear	<3
Croydon	3.6 ± 0.8	Norwich	0.8 ± 0.1
Bromley	17 ± 3	Merseyside	<1
West Glamorgan	3.7 ± 0.8	Bristol and Avon	0.2 ± 0.04
North and South Tees	3.7 ± 0.9	Bristol and Avon	0.2 ± 0.06
Forth Valley	15 ± 2	Wolverhampton	1.0 ± 0.5
Northampton	4.3 ± 0.8	Wolverhampton	0.7 ± 0.4
Exeter	3.6 ± 0.4	Clwyd	1.7 ± 0.7
Sunderland	8.7 ± 1.2	Winchester	0.6 ± 0.3
Sheffield	5.8 ± 0.6	West Cumbria A	8.7 ± 1.6
Blackburn	0.5 ± 0.3	West Cumbria B	15 ± 3
Cheltenham	1.3 ± 0.1	West Cumbria C	7.8 ± 1.6
Barnet	1.2 ± 0.4	Wexford	0.2 ± 0.07
Blackpool and Fylde	1.0 ± 0.3		

*Reported uncertainties are ± 1 standard deviation.

TABLE 5.5.2 $^{239+240}$Pu concentrations in teeth, mean, median, standard deviation and range (mBq kg^{-1} ash) (Long et al 1991).

Mean	Standard deviation	Median	Range
4.8[1]	5.6	3.6	0.2-21
4.2[2]	5.4	1.7	0.2-21
10.5[3]	3.9	8.7	7.8-15

[1]All samples.

[2]All samples excluding West Cumbrian Samples.

[3]West Cumbrian samples alone.

The mean of the samples from the West Cumbria region was 10.5 ±3.9 mBq kg^{-1} ash. This is not significantly different from the overall mean when the standard deviations are taken into account. Note that none of the West Cumbrian samples exceeded the levels found in samples from Bromley and Dewsbury.

The authors concluded that there was no correlation found with mean annual rainfall, nor did there appear to be any correlation with linear distance from Sellafield. The origin of the detected plutonium could not be established because of the ultra low levels involved.

In teeth sampled from 50 different areas in the UK, the average concentrations of $^{239+240}$Pu varied from 0.5 mBq kg^{-1} to 20 mBq kg^{-1} (ash weight) with a log normal distribution. Teeth collected from West Cumbria (average 14 mBq kg^{-1}) were in the range of values typical for rest of the UK and for the Irish Republic. No correlation was found with rainfall statistics (Priest et al, 1995)

A summary of UK and Irish data has also been reported by O'Donnell et al. (1997), reproduced in Table 5.5.3. The data shows that average plutonium concentrations in teeth collected from within the United Kingdom and Republic of Ireland (5 mBq kg^{-1} ash weight) are more than three orders of magnitude lower than the levels of alpha activity.

TABLE 5.5.3 $^{239+240}$Pu and total α-content of teeth collected within Great Britain and Ireland (O'Donnell et al 1997)

Radionuclide	*n*	Mean content (±SD) (Bq kg^{-1} ash weight)*	Median Content (±SD) (Bq kg^{-1} ash weight)*
$^{239+240}$Pu	67	0.005±0.004	0.002±0.003
Total α emitters	53	7±2	7±2

*All values are given to one significant figure. *n*= number of teeth

It should be noted that only batched samples of teeth were analysed and that some of the batches were measured at the detection limit for plutonium measurement by low-level α-spectrometry.

Results of a regression analysis for $^{239+240}$Pu concentrations plotted against distance from Sellafield showed a strong correlation (P=0.0081) Figure 5.5.2, but no relationship was shown for total α-emitter concentrations (P=0.91) Figure 5.5.3.

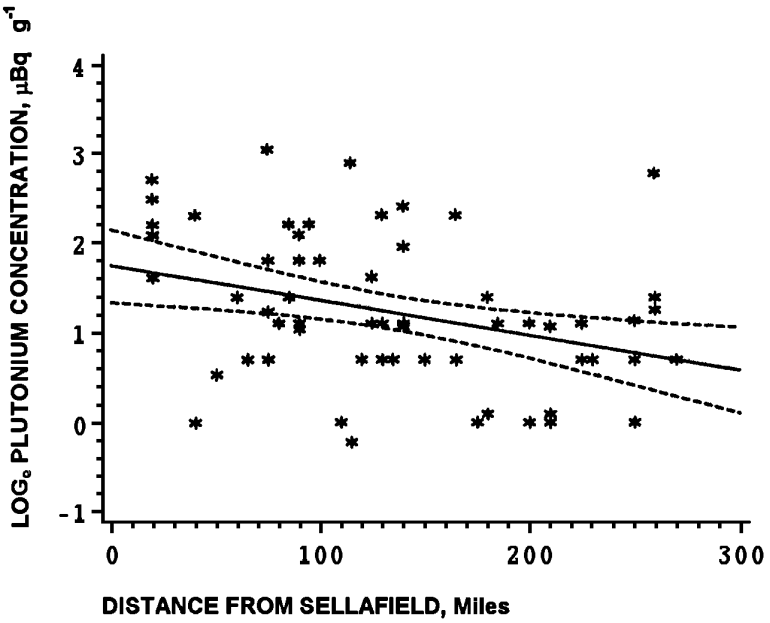

FIGURE 5.5.2 $^{239+240}$Pu content of children's teeth vs. distance from Sellafield. Regression equation: $\log_e[^{239+240}Pu] = 1.5314 - 0.0046$ x Distance from Sellafield (miles). Statistical significance: P<0.0081.

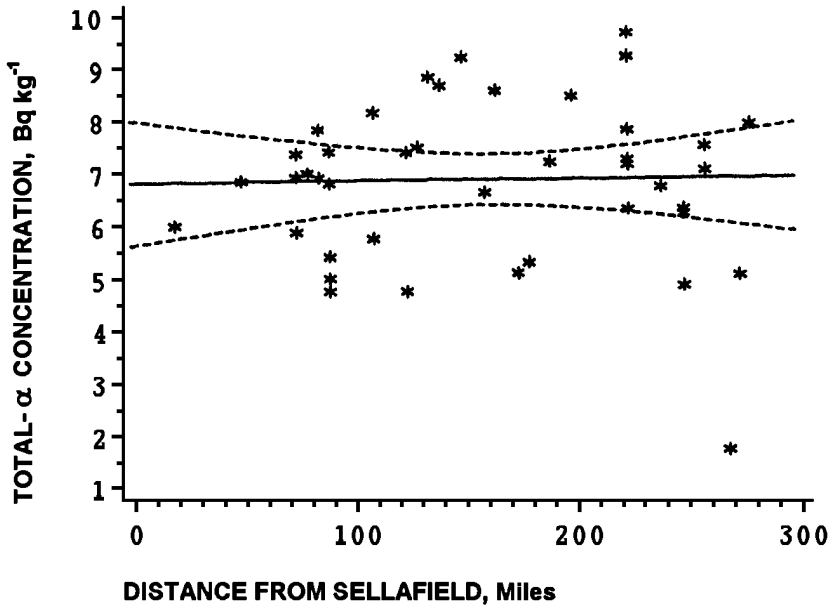

FIGURE 5.5.3 Total α-activity of children's teeth vs. distance from Sellafield. Regression equation: [total α-emitters] = 6.8071 + 0.0004 x Distance from Sellafield (miles). Statistical significance: P<0.9064.

Analyses shown in Figure 5.5.2 demonstrated that the plutonium content of teeth (C) was related to distance in miles from Sellafield (D) according to the following expression:

Log C= 1.5314+0.0046D mBq kg^{-1} ash weight

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 weight (95% confidence limits 8.6 and 3.9 mBq); then fall to approximately 1.8 mBq kg^{-1} ash weight (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 from different areas within defined distances from Sellafield (Table 5.5.4.).

TABLE 5.5.4. $^{239+240}$Pu concentration in teeth as a function of distance from Sellafield. (O'Donnell et al 1997)

Number of Batches	Distance (miles)	Activity (mBq kg^{-1} ash weight)
9	0-50	7.1±4.1
34	50-150	5.0±4.8
24	>150	3.0±2.7

Table 5.5.5 and Table 5.5.3 shows that levels in teeth were at the lower end of the range of equivalent recorded bone values. O'Donnell et al. suggest that the lower age of the childrens' teeth (therefore a shorter exposure period) than the adults donation of bones at death, give credence to the suggestion that plutonium levels in teeth are similar to those in bone. Teeth samples may be analysed to provide an index of variations in plutonium skeletal and hence body content.

The difference between plutonium levels in teeth collected from West Cumbria compared with sites remote from this location (see Table 5.5.4) is similar to that noted by Popplewell et al. (1985) for Pu levels in bone collected from persons who lived in West Cumbria compared with those that lived elsewhere in the UK.

The concentrations of ^{239}Pu in teeth reported by O'Donnell et al. (1997) have been compared with those previously reported for bone by several authors. In broad terms, the concentrations in teeth are reasonably similar to those in bone.

TABLE 5.5.5 Comparison of the measured levels of plutonium in the teeth with some levels previously reported in bone (O'Donnell et al, 1997)

Reference	Sample	Activity (mBq kg^{-1} ash weight)	Origin of samples
Long et al, 1991	Teeth	4.8	Great Britain
O'Donnell, 1997	Teeth	5	Great Britain and Ireland
(Bunzl and Kracke, 1983)	Bone	3-4	Germany
(O'Donnell, 1993)	Bone	3	Greenland
	Bone	14*	Ireland
(Burkinshaw et al, 1987)	Bone	5	Great Britain
Popplewell et al, 1985	Bone	3.5-6	Great Britain

*Hip bones from elderly patients

O'Donnell et al concluded that the BNFL Sellafield nuclear fuel reprocessing plant does contribute to plutonium body contents in the British population but that the levels of this element are so low that any increase in risk of induced skeletal tumours (including leukaemia),would be very small compared with those arising from the intake of natural radionuclides.

5.6 Uranium, thorium and polonium isotopes

Children's teeth obtained from Area Health Authorities in the UK by Priest et al. (1992) were analysed for ^{238}U (18 batches ranging 0.07 – 5.4 mBq g^{-1}) and ^{232}Th (15 batches ranging 0.02 – 0.44 mBq g^{-1}). The authors do not comment on these data.

Polonium-210 has been considered as a pollutant in exhaust fumes. Henshaw et al. (1995) reported raised levels of ^{210}Po in children's teeth who were living near motorways (Table 5.6.1). Linear regression analysis showed that for distances up to 10 km away, significant higher activity concentrations were found near the major motorways. For distances greater than 10 km the association breaks down. However, the authors note that the results have potentially confounding factors such as the closeness of major urban areas with already raised levels of ^{210}Po found in teeth, that a limitation was that there is no information on the time the children had lived in these areas.

TABLE 5.6.1 ^{210}Po in children's teeth (Henshaw et al, 1995)

	Number of teeth	Mean activity (SE) (Bq kg^{-1})	Slope with respect to motorway (Bq kg^{-1} km^{-1})	p-value*
All data†	220	8.93 (0.26)	0.40	<0.001
Tooth type (motorway)				
All				
M5/M6	58	9.32(0.55)	0.86	<0.05
M1/A1	105	7.80(0.41)	0.33	<0.05
M25	15	12.9(1.5)	…	NS
Deciduous				
M5/M6	26	10.1(0.9)	1.2	0.05
M1/A1	37	8.74(0.40)	1.1	<0.001
Permanent				
M5/M6	31	8.51(0.73)	…	NS
M1/A1	65	6.98(0.57)	…	NS
M1/M5/M6/A1‡	13	7.53(1.57)	8.6	<0.002
M5/M6‡	8	8.63(2.32)	12	<0.001

* Probability that slope is zero.
† Maximum distance=10 km, apart from ‡ where maximum distance =2 km. M=motorway, A=main duel carriageway.

Measurements were also made on ^{210}Po present from vehicle exhausts, on the basis of which it was concluded that this radionuclide should be added to the list of potential carcinogens from motorway vehicles.

5.7 Other isotopes

Lovaas and Hursh (1968) suggested that the ^{226}Ra and ^{210}Pb in teeth could be used to estimate the total quantity of these elements in the skeleton as a whole. Bones and teeth from 13 autopsies were analysed for ^{226}Ra and ^{210}Pb; the data are given in Tables 5.7.1 and 5.7.2. For ^{226}Ra the mean concentration in teeth, when compared with various bones (mandible, calvarium, tibia shaft, femur head and rib) lies within 50% of the average bone concentration at the 95% confidence level. For ^{210}Pb, variability was greater and dependent upon the level in the specific bone to which the tooth was compared.

TABLE 5.7.1 Mean concentration (mBq g^{-1}) of material analysed ± SD for 0.90 confidence interval (Lovaas and Hursh 1968)

Material	Teeth	Jaw	Tibia shaft	Calvarium	Femur head	Rib
^{226}Ra/g wet tissue	0.43±0.11	0.33±0.14	0.28±0.09	0.33±0.10	0.16±0.07	0.27±0.12
^{226}Ra/g calcium	1.79±0.45	1.82±0.77	1.60±0.48	1.67±0.50	1.80±0.65	2.24±0.81
^{210}Pb/g wet tissue	1.95±0.31	1.18±0.27	1.12±0.24	1.43±0.26	1.00±0.23	1.24±0.35
^{210}Pb/g calcium	8.18±1.44	6.59±1.52	6.29±1.26	7.34±1.37	12.95±3.22	10.92±3.07

TABLE 5.7.2 Comparison with some published values (mBq/g ash) (Lovaas and Hursh 1968)

	^{226}Ra			^{210}Pb		
Source	Tooth	Bone		Tooth	Bone	
Central US (DiFerrante et al, 1964)	0.592					
Brazil (Penna et al, 1965)	0.962					
NE US (Hunt et al, 1963)	0.592	0.518	Iliac crest	2.566	5.254	
Lovaas & Hursh	0.703	0.629 0.703 0.703 1.036	Compact Mixed Femur head Rib	3.07	2.627 3.478 4.995 4.218	Compact Mixed Femur head Rib
NY Whole body ash (Hursh, 1960)		-			1.11-1.48	
Central US (Lucas, 1964)		1.036	Mixed		2.96	Mixed
Germany (Groos et al, 1963)					4.07	
Central US (Holtzman, 1963)		1.369	Mixed		5.402	Mixed
Germany (Muth et al, 1960)		0.444 0.407	Tibia shaft Femur			Tibia shaft Femur
Worldwide sampling programme (Walton et al, 1959)		0.37 (range, 0.111-1.11)	mixed			

Henshaw et al. (1994) studied the spatial distribution of naturally occurring ^{210}Po and ^{226}Ra in children's teeth, obtained throughout the UK, using α-sensitive plastic track detectors. The distributions of these radionuclides are non-uniform, with ^{210}Po being associated with the outer enamel and the ^{226}Ra with the pulp. Activity concentration was measured in 900 teeth. Total concentration on the outer enamel surface of deciduous teeth, permanent teeth from children less than 10 years old and permanent teeth from children more than 10 years old give mean values of 8.63±0.26, 5.76±0.48 and 7.00±0.15 Bq kg^{-1} respectively. The ^{226}Ra concentrations in pulp, dentine and annular enamel give respective mean values of 0.715±0.055, 0.418±0.083 and 0.514±0.029 Bq kg^{-1}. The mean concentration in 32 foetal teeth was 2.05±0.31 Bq kg^{-1}.

5.8 Summary

Strontium-90 has been measured in permanent teeth of people, and the results have reflected the changes of the isotope in the environment due to fallout from nuclear testing. It has been shown that no statistically significant correlation existed between ^{90}Sr levels in teeth and the distance from Sellafield.

There are higher concentration levels of $^{239+240}$Pu in teeth of people living near the BNFL nuclear plant. However, the levels of these elements are so low that any increase in risk of induced skeletal tumours (including leukaemia) would be very small compared with those arising from the intake of natural radionuclides.

6 URINE MEASUREMENTS

6.1 Introduction

Realising that there were uncertainties in risk and dose calculations the Committee on Medical Aspects of Radiation in the Environment (COMARE) recommended that radioactivity should be measured in the general public living near major nuclear installations and in control areas (COMARE, 1988). However, few studies have been undertaken in such cases, or indeed in the general population. Published studies are reviewed below.

6.2 Strontium isotopes

Following COMARE's recommendations in 1988, ^{90}Sr was measured in the urine of 31 people (including 4 Caithness childhood cancer cases, 16 case parents, 4 child case controls and 9 case control parents) living in the Dounreay area (Watson and Sumner, 1996). They compared levels of radioactivity in subjects with estimated levels required to account for the observed incidence of childhood leukaemia. It was estimated that the body contents of ^{90}Sr required (in 1990 – 91) to explain the incidence of excess leukaemias, would be at least 0.19 MBq. No ^{90}Sr levels in urine were found above the detection limit of 0.05 Bq per 100 ml sample, for any individual. The minimum detection level corresponds to a body content (in 1990-91) of 7500 Bq (Boecker, 1991). The authors deduced that it was unlikely that the population was heavily contaminated with ^{90}Sr and that it was unlikely that the observed increase in childhood leukaemia in the vicinity of Dounreay Nuclear Establishment can be due to the single factor of internal radioactive contamination from the plant.

Warren (1967) measured levels of ^{90}Sr in diet and urine for two age groups of children. The results are summarised in Tables 6.2.1 and 6.2.2.

TABLE 6.2.1 Strontium-90/calcium ratios in the diet and urine of children on a mixed diet (Warren, 1967)

			^{90}Sr (Bq g^{-1} Ca)		
Subject	Age	Date of sampling	Diet	Urine	Urine/ diet
EM	12y	10/63	1.36	0.58	0.43
BM	11y	10/63	1.66	1.15	0.70
BC	9y	11/63	1.19	0.58	0.49
JM	13y	12/63	1.37	0.48	0.35
KM	6.5y	1/64	1.41	0.79	0.56
CM	10y	3/64	1.48	0.56	0.38
LC	9.5y	3/64	1.21	0.54	0.45
AD	10.5y	4/64	1.23	0.51	0.41
SC	4.75y	4/64	1.63	0.81	0.50
PG	4.5y	7/64	2.19	2.82	1.29*
BMc	9y	8/64	1.09	0.95	0.86
MG	2.5y	2/65	1.22	2.97	2.43*
		Mean	1.37	0.69	0.51
					(sd 0.15)

*Excluded from averages, PG was acutely ill with leukeamia. MG was suffering from rickets, resistant to vitamin D.

TABLE 6.2.2 Strontium-90/calcium ratios in the diet and urine of infants with an all-milk diet (Warren, 1967)

			^{90}Sr (Bq g^{-1} Ca)		
Subject	Age	Date of sampling	Diet	Urine	Urine/ diet
SM	3w	2/64	1.28	1.22	0.95
RP	5w	2/64	2.09	2.14	1.02
AG	7w	3/64	1.50	1.57	1.04
JG	3m	1/65	1.33	1.89	1.42
DM	5w	1/65	1.28	1.63	1.27
ST	7w	3/65	1.34	1.97	1.47
WA	3m	3/64	1.45	1.37	0.94
		Mean	1.47	1.68	1.16
					(sd 0.21)

6.3 Caesium isotopes

The uptake of radiocaesium by members of the Scottish population has been measured by whole body counting following the Chernobyl reactor accident (East et al, 1992). The relationship between body radiocaesium levels and urinary output was examined to validate the practicality of using urinary radiocaesium analysis for screening the population. The volunteers were between the ages of 20 and 60 years. Table 6.3.1 shows the results of the comparison of measured urinary and body radiocaesium.

TABLE 6.3.1 Urinary ^{137}Cs and Body ^{137}Cs (East et al. 1992)

Subject no: (F=female)	24 h Urinary ^{137}Cs (Bq)		Body ^{137}Cs (Bq)	*Ratio Body ^{137}Cs/ Urinary ^{137}Cs
	Creatinine corrected	Time-corrected		
1	0.623	0.872	122	196
2	1.02	1.603	183	179
3	1.38	0.409	77	56
4	0.373	0.336	36	97
5	0.662	0.392	96	145
6	0.508	0.428	115	226
7	0.468	0.520	95	203
8	0.824	0.829	157	191
9	0.455	0512	101	222
10	0.490	0.577	87	178
11	0.441	0.553	79	179
12	0.679	0.731	161	237
13	0.619	0.463	48	78
14	0.981	1.085	121	123
15	0.347	0.325	88	253
16	0.410	0.437	<26	<63
17	0.347	0.313	47	135
18	0.941	0.559	161	171
19	1.82	2.383	382	210
20	1.84	1.171	275	150
21	5.56	4.135	1771	318
22F	0.285	0.327	59	207
23	0.140	<0.083	112	800
24F	0.167	0.225	58	347
25F	0.644	0.740	58	90
26F	0.366	0.382	64	175
27F	0.438	0.523	74	169
28	0.430	0.494	95	221
29	5.742	4.151	776	135
30	19.62	29.795	4552	232
31F	6.25	12.013	1041	166
32F	1.76	1.616	331	189
33	0.374	0.451	98	262
34F	0.289	0.350	61	211

* Based on creatinine corrected data

Urinary ^{137}Cs was corrected to standard 24-h output using measured creatinine compared with the reference values for men and women. Urinary ^{137}Cs was also normalised to a standard 24-h output using a time correction factor based on duration of urine collection. The variability in the ratio is due probably to differences in the retention half time of ^{137}Cs in the body. In this same study, ^{134}Cs was measured in urine but often the body measurement was below the limit of detection. However, where both radionuclides could be measured the ratio of ^{137}Cs:^{134}Cs was consistently 6.8:1 in both the body and the urine.

The relationship between stable caesium (which consists of 100% ^{133}Cs) and radioactive Cs has also been investigated. Table 6.3.2 gives the results for the UK measurements of ^{133}Cs in 24-h urine samples expressed as the total weight of caesium excreted in μg d^{-1}. The mean rate of excretion was 9.13 ± 5.18 μg d^{-1} with values ranging from 1.9 μg d^{-1} to 20.3 μg d^{-1} (East et al, 1992).

For comparison, measurements in Italian subjects (Clemente et al, 1971) showed a mean excretion rate for ^{133}Cs of 16 μg d^{-1}; the ICRP value for reference man is 9 μg d^{-1} (ICRP, 1975).

TABLE 6.3.2 Urinary ^{133}Cs, ^{137}Cs and derived body ^{137}Cs (East et al, 1992)

Subject No (F=female)	24h urinary ^{133}Cs (μg)	Urinary ^{137}Cs/ ^{133}Cs (Bq μg^{-1})	Derived body ^{137}Cs (Bq)	Ratio to body ^{137}Cs
2	20.4	0.050	75	2.44
4	1.90	0.196	294	0.122
5	2.76	0.240	360	0.267
6	9.31	0.055	83	1.39
7	13.5	0.035	52	1.82
8	15.3	0.054	81	1.94
10	15.6	0.031	47	1.85
12	10.1	0.067	101	1.59
13	5.59	0.111	167	3.48
14	19.4	0.051	76	1.59
15	5.64	0.062	92	0.96
16	12.3	0.033	50	
17	1.33	0.003	4	11.8
18	5.83	0.161	242	0.67
19	9.18	0.198	297	1.29
20	2.86	0.642	963	0.29
21	6.38	0.872	1310	1.35
22F	6.19	0.046	69	0.86
23	6.56	0.021	32	
24F	6.19	0.030	46	1.26
25F	17.8	0.036	54	1.07
26F	5.84	0.063	94	0.68
27F	5.02	0.087	131	0.56
28	8.49	0.051	76	1.25
30	19.7	0.994	1490	3.06
31F	8.86	0.705	1060	0.98
32F	5.82	0.302	453	0.73
33	5.98	0.063	94	1.04
34F	6.42	0.045	68	0.90

The table also shows the ratio of urinary ^{137}Cs:^{133}Cs (^{137}Cs creatinine corrected values, Table 6.3.1.). Assuming equilibrium, and therefore that this ratio was the same in the urine and the body, and taking a value of 1.5 mg as the total-body content of stable caesium (Ibid, p328), the body activity of ^{137}Cs was calculated. In order to compare this result with that obtained by whole-body monitoring (Table 6.3.1), the ratio of the two methods is calculated in the final column.

The mean rate of urinary excretion of stable caesium was 9 μg d^{-1} but the comparison of body ^{137}Cs calculated from the urinary $^{137}Cs/^{133}Cs$ ratio with that obtained by whole-body monitoring did not give consistent results. For men only, the relationships obtained indicated that the urinary method severely underestimated the body ^{137}Cs. However, for women the relationship was almost perfect. Taken overall, the authors suggest that the method for urine monitoring is adequate for emergency planning situations.

6.4 Iodine isotopes

No references found.

6.5 Plutonium and americium isotopes

Following COMARE's recommendations in their 2nd Report, measurements have been made of the urinary content of ^{239}Pu in 66 subjects associated with the Dounreay area and 42 subjects living remote from reprocessing plants (Watson and Sumner, 1996) (see 6.1 Strontium Isotopes). The highest urinary output of ^{239}Pu measured was 37 x 10^{-6} Bq d^{-1}; it was calculated that this would correspond to a whole body content of about 1.9 Bq if the exposure took place 20 y ago. The assessed effective dose was about 4 mSv. The authors suggest that this is about two orders of magnitude less than the plutonium body content that would be required to explain the excess leukaemias in the vicinity of the Dounreay Nuclear Establishment. This is consistent with the theoretical findings of the NRPB (Stather et al, 1988) and the conclusions of COMARE (1988).

Recently, Accelerator Mass Spectrometry (AMS) has been used for the measurement of ^{239}Pu in urine (Priest et al, 1999). Levels of plutonium excretion were determined for members of the general population which included teenage children resident in North London, a small number of adults, and an adult male volunteer who ingested plutonium-contaminated sediment from the Irish Sea. Table 6.5.1 shows results for the analysis of plutonium in urine donated by young adults in North London.

TABLE 6.5.1 Results of analysis of 24-h urine samples provided by young adults resident in North London (Priest et al, 1999)

Samples (*n*)	Mean age	Mean $^{242}Pu^{*}$ count rate (min^{-1})	Mean ^{239}Pu count rate (min^{-1})	Mean mass ratio ^{242}Pu:^{239}Pu	Mean mass ^{239}Pu (fg)	Mean activity ^{239}Pu (μBq)
Females (11)	17 years 3 months	428	0.39	928:1	1.6	3.3
Males (6)	17 years 5 months	370	0.51	1722:1	1.7	3.7
All (17)	17 years 4 months	408	0.46	1229:1	1.6	3.5

*Internal yield tracer.

It can be seen that the levels of ^{239}Pu excreted were similar between males and females of this age. Measured activities in individual samples ranged from <0.3 to 10 μBq d^{-1}. The data were distributed log-normally with a mean value of 3.5 μBq d^{-1} and a mode of ~2 μBq d^{-1}. There was no background subtraction and thus the values should be regarded as upper limits.

An adult male, with minimal occupational exposure to ^{239}Pu provided four samples measuring 2.8, 1.8, 1.3 and 1.9 μBq d^{-1}. In contrast, the measured daily excretion of plutonium by a radiochemist (formerly employed at Aldermaston) was 260 μBq d^{-1}. Another radiochemist, who had never worked close to substantial amounts of plutonium, excreted less; 58 μBq. A single female excreted 1.8 μBq d^{-1}. This value is very close to both the average activity of ^{239}Pu excreted by the first 'minimally exposed' male (1.9 μBq d^{-1}) and the average excreted by young adults described above (3.5 μBq d^{-1}). Priest et al propose that if the retention half-time of plutonium in adults is assumed to be about 50 years, then these results ranging from about 50 mBq to 10 Bq. This range represents a very small fraction of the ICRP annual limits on intake for this isotope (ICRP, 1979).

A neutron induced fission track analysis (FTA) technique for measuring ultra-low levels of ^{239}Pu in urine samples from the public has been developed (Wrenn et al, 1994). The authors showed that for Western US residents the mean urinary excretion of ^{239}Pu was 0.6 μBq l^{-1} from persons born in 1958 or earlier, and 0.3 μBq l^{-1} for persons born after 1961. The authors suggest that this FTA technique was sufficiently sensitive enough to detect an effective dose of 0.02 mSv.

6.6 Uranium, thorium and polonium isotopes

6.6.1 Uranium

In one UK study, uranium excretion was measured in urine from five male and 1 female 'unexposed' volunteers from a research institute in the vicinity of the BNFL nuclear processing plant at Sellafield (Riddell, 1995). Uranium was determined in 24-hour urine samples by inductively coupled plasma-mass spectrometry (ICP-MS). The results are given in Table 6.6.1.

TABLE 6.6.1 Uranium urinary excretion (Riddell, 1995)

Volume (ml)	Creatinine (g)	U content (ng)
1940	1.22±0.04	146±152
1425*	0.70±0.01*	95±103*
1475	1.31±0.03	127±134
1575	1.35±0.03	189±92
2175	1.21±0.02	66±29
1850	1.22±0.04	182±204

*female

Due to the paucity of UK data, urinary excretion rates have been considered for members of the public living in other countries. The daily excretion rates of uranium in urine measured in an unexposed population in the US ranged from 1.4 to 35 ng l^{-1} (Ting et al, 1999). However, in India daily excretion rates in excess of 200 ng d^{-1} have been reported (Dang et al, 1992).

Research on urinary excretion of uranium by non-exposed persons was carried out on over 200 healthy volunteers of both sexes aged between 7 and 84, from various parts of Germany (Roth, et al, 2001). They reported that there is no difference between men and women in daily renal excretion of uranium and that the levels increase with subjects' age. Excretion levels for children are generally low and much higher values were observed for adults. The authors observed that there were also older persons with very low values and the spread of excretion levels thus increased to about two orders of magnitude. This is not specific to uranium but is also observed for other long-lived radionuclides such as ^{226}Ra and ^{232}Th or for non-radioactive heavy metals.

The authors suggested that data from the healthy volunteers allowed a tentative derivation of an age-dependent mean and an upper empirical value for uranium excretion in the urine by persons with no known additional exposure. (In the course of this work, however, a number of values were found to lie outside this typical reference zone.) The mean value was thus 15 ng d^{-1} for 20-year-olds and about 30 ng d^{-1} for 50-year-olds, while the upper boundary of the reference zone lies at 30 ng d^{-1} and 70 ng d^{-1} respectively. This boundary must not be misinterpreted as a health risk threshold. It merely describes the area encompassing most (not all) of the values found for healthy persons. In the current state of knowledge of uranium toxicity, it appears that a level of intake constituting a danger to health would lead to excretion levels which were greater than those observed by more than one order of magnitude.

The above variations in urinary excretion which could occur in principle in the UK, highlight the importance of establishing adequate controls. Environmental and dietary considerations should clearly be considered before concluding that enhanced intakes of uranium may result solely from civil or military operations involving uranium.

6.6.2 Thorium

References relating to thorium excretion by members of the public in the UK have not been found. However, Table 6.6.2 shows some measurements of thorium excretion rates for members of the public elsewhere. In a comprehensive human study in Germany (Roth et al, 1997), the average daily excretion was investigated in 30 males and 25 females. The analyses were carried out using ICP-MS. The results varied considerably, but the authors concluded that the difference between males and females was not statistically significant and moreover there was no clear dependency on age. The excretion rates for one of the subjects, followed for 15 consecutive days, demonstrated that intra-subject variation could also occur. The average daily excretion of another seven subjects reported by Roth et al (1997) were in reasonable agreement with those above. Other studies however, (Dang et al, 1989; Hewson and Fardy, 1993; Minoia et al, 1990) using a neutron activation technique, compound the uncertainties associated with assigning reference values to Th excretion. It is noteworthy that the values obtained by Hewson for some subjects are more than an order of magnitude less than those found by Roth et al (1997), whilst some of those reported by Minoia et al (1990) are more than an order of magnitude greater.

TABLE 6.6.2 Thorium excretion rates for members of the public

Study	No. Persons	Average daily excretion ($\mu Bq\ d^{-1}$)	Range
		Mean±SD	
ICRP 1975	Reference man	405	
Roth 1997 (Germany)	30 males	55±29	17-121
	25 females	41±21	9-99
	55 total	47±26	
	1 for 15 d	96	26-167
Roth 1997 (Germany)	7	29	
Dang 1989 (India)	11	16±6[a]	
Hewson 1993 (Australia)	2	<4	
	3	29	
Minoia 1990 (Europe)	25	490±220*	56-4060

* Values expressed originally as ng l^{-1} and converted to $\mu Bq\ d^{-1}$ on the basis that the daily urinary excretion is 1.4 l d^{-1} and the specific activity of ^{232}Th is 4050 Bq g^{-1}.

Like uranium, assessments of enhanced intake should be treated with caution, unless adequate control data are available.

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