A Survey of the Specific Activities of ^{210}Pb and ^{210}Po in Terrestrial Foodstuffs from England and Wales: A Potential for the Technological Enhancement of Natural Radioactivity

by

Keith McKay CChem, MRSC

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Scottish Universities Research and Reactor Centre

Abstract

A survey of the specific activities of ^{210}Pb and ^{210}Po in terrestrial foodstuffs from areas in England and Wales close to sources of the possible technological enhancement of natural radioactivity (TENR) has been performed. Sampling of a variety of foodstuffs important in the UK diet from areas close to coal fired power stations, metal smelting plants, a phosphate ore processing plant, a nuclear fuel reprocessing plant and an area of high natural radioactivity was undertaken. The specific activities found were compared to those from control areas to aid in the determination of the level of enhancement of ^{210}Pb and ^{210}Po.

The levels of ^{210}Pb and ^{210}Po in the foodstuffs sampled varied according to type. Root crops and milk had low, leafy green vegetables and fruit had intermediate, and offal and cereals had high specific activities of ^{210}Pb and ^{210}Po. The levels between sites varied but control sites were not always the lowest. This made the decision as to whether enhancement had occurred a difficult one. A series of statistical tests were used to decide on enhancement, and based on these tests, two sites showed enhancement of ^{210}Pb and ^{210}Po in barley, and one site in cabbage. One site showed enhancement of ^{210}Po in bovine liver.

A survey of world-wide data on the levels of ^{210}Pb and ^{210}Po in terrestrial foodstuffs was performed from the literature. Amongst these data were results from areas of known enhancement. In comparing the world-wide data to those found in this study, those enhanced sites in England and Wales would be classed as relatively minor in enhancement or not at all.

The committed effective dose (CED) to the populations at each of the sites was calculated using the specific activities of ^{210}Pb and ^{210}Po, and national statistics on food consumption. Comparisons were difficult to make as not all foodstuffs were available at all sites but based on the CED from the two largest 97.5th percentile consumption rate foodstuffs some comparisons could be made. In general the 6 - 12 month age group received the highest dose with the 16 - 64 year age group the lowest. The highest doses were received from an area close to a metal smelter with the lowest at a low rainfall control site. All doses were within the UK limit of 1 mSv yr^{-1}.

The proportions to the dose of ^{210}Pb and ^{210}Po were important, with that for ^{210}Po more so in the 6 -12 month age group. The proportions of dose depend on the activity ratio of ^{210}Po:^{210}Pb in the foodstuffs and on the dose coefficient for these radionuclides.

Annual intakes of ^{210}Pb and ^{210}Po from this study were compared to world-wide data from the literature. Using these data the CED was also calculated and compared to the results of this study. Both the annual intakes and the CED from these intakes from this study were found to be in the range of the world-wide results.

I dedicate this thesis to my parents, Evelyn Janet McKay, and Thomas McDivitt McKay, and to my wife Carol.

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I have come late in life to thesis writing after a long and varied career. I therefore take this opportunity to thank those colleagues with whom I have worked over the years.

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Chapter 1 Introduction

1.a Initial discussion

The Technological Enhancement of Natural Radiation (TENR), has been defined as, the exposure to sources of a truly natural radiation environment (NRE), which would not occur without, or is increased by, some technological activity not designed to produce radiation (Steinhäusler, 1990). This definition is all encompassing, and depends heavily on the nature of what is truly natural. Thus, the radiation dose received by an astronaut orbiting the earth, as well as the dose received from the consumption of foodstuffs grown on uranium mill tailings, would be classed as legitimate consequences of TENR even though in the case of the astronaut the source remains unchanged.

Most interest in TENR has come from the effects of industrial activities on the human environment (Baxter,1993), in particular, mining, ore processing, and non-nuclear energy production (Steinhäusler, 1991). The pathways through which a radiation dose can be received from these activities are by inhalation, ingestion and external irradiation. For the ingestion pathway, the major route is by the consumption of foodstuffs grown in the locality of the source of TENR, and for the inhalation pathway the breathing of dusts or vapors into the lungs. With external irradiation, significant increases in the natural radioactivity background dose rate can occur from the effects of TENR. As an example, spoil heaps and the waste from radium paint factories can give rise to enhanced doses above the natural background, as can travelling by aeroplane at high altitude. It is important to study those communities close to sources of TENR, to ensure that the doses they receive are within agreed limits.

This thesis concentrates on one particular aspect of TENR, the potential for enhancement of ^{210}Pb and ^{210}Po in terrestrial foodstuffs by technological and natural sources, and the resulting radiological consequences for the consumption of the these foodstuffs.

1.b Sources of ^{210}Pb and ^{210}Po

1.b.i Natural Sources

1.b.i.i Natural Series Decay and Secular Equilibrium

Since Henri Antoine Becquerel's (1896) observation that uranium salts spontaneously blackened a sealed photographic plate, (later given the name of radioactivity by Marie Curie (1898)), the scientific community has been fascinated with the phenomenon. Marie Curie's studies, in the former dissecting room of the Faculty of Medicine, under abominable conditions, led to the discovery of more elements with this property (Curie, 1938). In conjunction with her husband Pierre she discovered first, polonium, (named after the country of her birth, Poland) and then radium, which they isolated in a mammoth separation from the residue of pitchblende ore taken from St. Joachimstal mines in Bohemia. And so, the isolation of the first radioactive element was from the products of the technological enhancement of natural radioactivity.

Within 15 years of Becquerel's discovery of radioactivity, approximately 40 different radioactive species had been identified through their chemical nature, radiation properties, and characteristic half lives. These chemical species or isotopes as they were named by Soddy (1914) were further seen to be part of three distinct disintegration or decay series, which started with a long-lived isotope of uranium or thorium, and ended with a stable isotope of lead (see figure 1 below).

Each nuclide in the decay series decays according to the radioactivity decay law:

$$-\frac{dN}{dt} = \lambda t$$

where λ = the decay constant of the nuclide

N = the number of atoms of the nuclide

t = time

On integrating this equation we get the general equation for radioactive decay

$N_t = N_0 e^{-\lambda t}$ Where N_t = the number of atoms at time t

N_0 = the number of atoms at time 0

Since the decay rate (*A*) is proportional to the number of atoms, the equation can be rewritten as

$$A_t = A_0 e^{-\lambda t}$$

Now, if the daughter of the radioactive species is also radioactive, then, the net rate of formation of the daughter atoms is the difference between the rate of formation of the daughter and its decay rate as follows

$$\frac{dN_2}{dt} = N_1\lambda_1 - N_2\lambda_2$$

where N_1 = number of parent atoms

N_2 = number of daughter atoms

λ_1 = decay constant of the parent

λ_2 = decay constant of the daughter

Again on integrating, the solution of this equation is

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^0 e^{-\lambda_2 t}$$

where N_1^0 and N_2^0 are the number of atoms of parent and daughter at time t=0

If we have pure parent at t=0 then, the number of daughter atoms will be 0, so the equation further simplifies to

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) \quad \text{Equation 1}$$

In a situation where the half-life of the daughter is very much smaller than that of its parent i.e. $\lambda_1 << \lambda_2$, we can further simplify the above equation to become

$$N_2 = \frac{\lambda_1}{\lambda_2} N_1^0 \left(1 - e^{-\lambda_2 t}\right)$$

It can be seen from the above equation that the daughter atoms grow in with its own half-life and that when t is infinity

$$N_2\lambda_2 = N_1\lambda_1$$

or, since $A = N\lambda$,

$$A_2 = A_1$$

Thus at this time the parent and daughter activities are equal and the daughter decays with the half-life of the parent. This situation is known as radioactive or, more commonly, secular equilibrium.

Two other forms of radioactive equilibrium can also occur, transient equilibrium and no equilibrium.

In transient equilibrium, the daughter has a half-life which is an appreciable fraction of the parent's, and as such, during the observation of the daughter's growth, decay of the parent will also be observed . Thus the activity of the daughter will increase to a maximum and then decay with the half-life of the parent. This can be illustrated by manipulation of equation 1.

When t becomes very large, $e^{-\lambda_2 t}$ is small compared to $e^{-\lambda_1 t}$, and equation 1 becomes

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 e^{-\lambda_1 t}$$

or,

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1 \quad \text{since} \quad N_1 = N_1^0 e^{-\lambda_1 t}$$

Thus at equilibrium the daughter activity will be greater than the parent activity by a factor of $\frac{\lambda_2}{\lambda_2 - \lambda_1}$.

In the case of the daughter nuclide having a half-life longer than its parent, the daughter activity will grow into some maximum value and then decay with its own half-life and thus no equilibrium is attained (Friedlander *et al.*1964, Choppin *et al.* 1980).

For the natural decay series a further complication is introduced in the fact that they are chains which have many successive decays, and thus the mathematics becomes more complicated. In 1910, Bateman solved the decay equation for a chain of *n* members, with the special assumption, that at $t=0$, only parent is present. The solution then becomes

$$N_n = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \ldots C_n e^{-\lambda_n t}\text{, where}$$

$$C_1 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)\ldots(\lambda_n - \lambda_1)} N_1^0\text{, and,}$$

$$C_2 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)\ldots(\lambda_n - \lambda_2)} N_1^0\text{, and so on}$$

Equipped with Bateman's solution, the natural decay series have been utilized by many workers for the dating of processes over long and short timescales, such as in the dating of fossil corals (up to 1 million years) to determining the rates of scavenging processes in the oceans (days) (Ivanovich and Harmon, 1992). The natural decay series will always have an important role in the study of natural processes.

Figure 1a The ^{232}Th Natural Decay Series

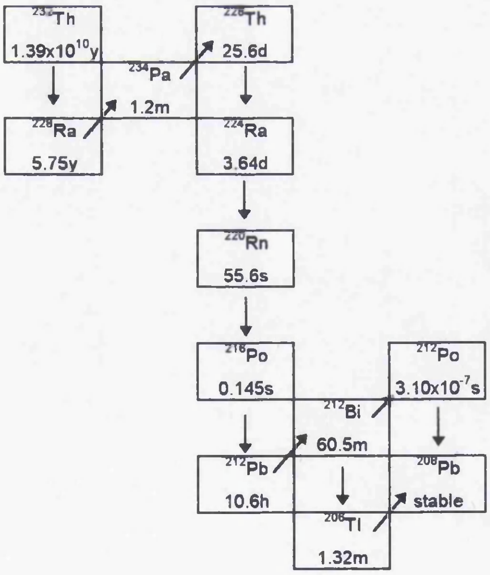


Figure 1b The ^{235}U Natural Decay Series

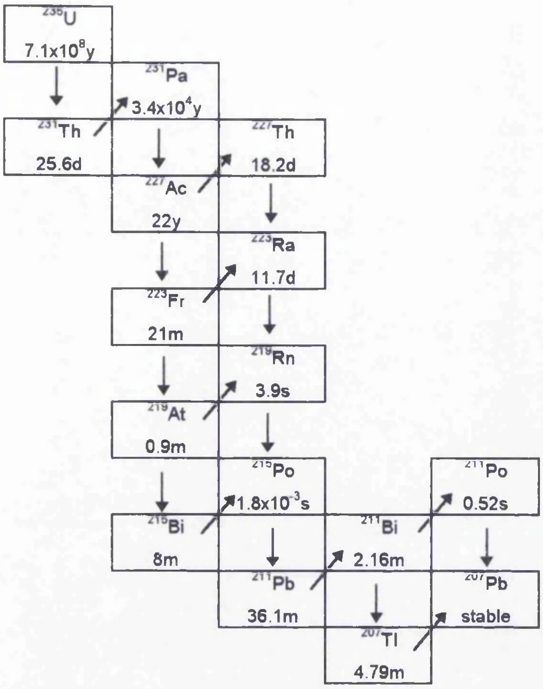
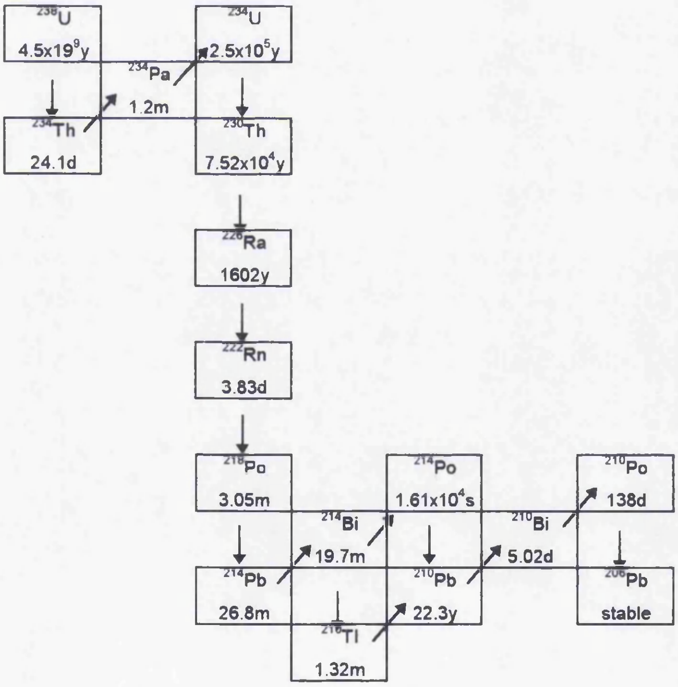


Figure 1c The ^{238}U Natural Decay Series



The very different chemical and physical properties of the radionuclides within the natural decay series make for some interesting observations. Each of the three series has an isotope of the noble gas radon within the decay chain, ^{222}Rn (radon, $t_{½}$=3.825d, ^{238}U-series), ^{220}Rn (thoron, $t_{½}$=54.5s, ^{232}Th-series) and,

^{219}Rn (actinon, $t_{½}$=3.92s, ^{235}U-series). In a system that has been closed for a time which is long relative to the daughters half life, all of the daughters within the decay series will be in equilibrium with their parents, but, in an open system, there is the possibility of disequilibrium because of the loss of radon isotopes within the decay series. This is particularly important in the ^{238}U decay series, since the ^{222}Rn half-life is long enough for it to travel some distance before decaying to its daughter products, two of which are ^{210}Pb and ^{210}Po.

The geochemistry of the individual constituents of the decay series gives ample opportunity for fractionation, to disrupt the secular equilibrium which would otherwise occur. Depending on the oxidation states of the radionuclide this can effect its ability or otherwise to form complexes which could be transported in the hydrogeological cycle. This is amply illustrated with two examples.

Uranium, in the 6+ oxidation state exists in nature as the uranyl ion $(UO_2)^{2+}$, which readily complexes with carbonate, sulphate, or phosphate ions to produce oxyanions which are readily soluble in aqueous systems and are therefore mobile. Whilst in the reduced 4+ state, uranium is insoluble and readily precipitates from solution (Gascoyne,1992). On the other hand radium has one stable oxidation state, 2+, and is chemically similar to barium and to a lesser extent calcium. In sulphate solution, radium will co-precipitate along with barium thus rendering it immobile, whereas in chloride solution, radium forms a complex with chloride ions, which is mobile (Gascoyne,1992). There are many other processes which can be invoked which cause disequilibrium, and indeed disequilibrium would in many cases be the norm, although the earth as a whole would be in a state of secular equilibrium (Cowart and Burnett., 1994).

1.b.i.ii Mineralisation Processes

Uranium and thorium are ubiquitous within the earth and are present in rocks and soils at varying concentrations. Higher levels of uranium and thorium are found in continental igneous rocks than in oceanic igneous rocks. Granites have uranium concentrations in the range 2.2 - 6.1 ppm. This is considerably higher than those in basalts which lie in the range 0.1 - 1 ppm (Gascoyne, 1992). Primary uranium ore deposits are found within granitic bodies where fluids have crystallised as the granitic magma has cooled, forming pegmatites or hydrothermal veins. Similarly, metamorphic changes can dehydrate rocks, expelling hot aqueous fluids which take with them uranium, since it does not readily fit into the new silicate structure of the metamorphic rocks formed, to be later crystallised as pegmatite. In these rare rock types, uranium is found as uraninite (pitchblende) with a composition which varies between UO_2 and U_3O_8.

Rocks on the surface of the earth are continually exposed to weathering as part of the natural geological cycle. This process can redistribute primary uranium minerals to form secondary uranium deposits. Many kinds of secondary deposits are found, the simplest being those formed by the deposition of grains of uranium mineral in new sediments. These kinds of deposits are quite rare because the uraninite mineral in the presence of an oxygen rich atmosphere forms U^{6+} which is soluble in aqueous systems as the rocks weather. However under reducing conditions, U^{6+} will be readily converted to U^{4+} and precipitate from solution.

Typical reducing environments for the deposition of secondary uranium are found in coal swamps, where it is precipitated in uranium-rich organic compounds and as uraninite. Uranium concentrations in coal can be in the range 200 - 6000 ppm and thus because of the age of the coal deposits they are likely to have enhanced levels of daughter nuclides from their decay series (see later).

Marine phosphate deposits are also rich sources of uranium, and again reducing conditions play an important part in uranium deposition within these ores. The principal phosphate mineral is apatite $Ca_5(PO_4)_3(OH,F)$ and because Ca^{2+} and U^{4+} are of a similar size (0.99Å and 0.97Å respectively), U^{4+} can substitute for Ca^{2+} within the crystal lattice of apatite even though the charge difference of 2+ is quite large. This can be accommodated by adjusting the proportions of OH^- and F^- which are loosely held within the crystal structure.

Typical concentrations for uranium in phosphate rocks are, 10 - 200 ppm in phosphate ores from Florida, USA (Lindeken,1980), whilst on a world-wide basis values are in the range 3 - 399 ppm.

Sedimentary "black shales" are also formed under reducing conditions, and, being organic rich can have high concentrations of uranium, typically in the range 3 - 1200 ppm (Gascoyne, 1992).

By far the most important secondary uranium deposits are those which have been formed by deposition from groundwater, since they are proving to be the richest source of uranium because of their immense volume, even though they may be of low uranium specific activity. The classic situation occurs when uranium rich percolating groundwater, on passing through dipping sandstone, will deposit uranium at an oxidation - reduction boundary (redox front) . As erosion occurs this front will move with the groundwater flow. These deposits are known as roll-type deposits.

As can be seen from the previous discussion, the possibility for uranium series disequilibrium is high, and, indeed a comparison of the relative activities of ^{238}U to ^{226}Ra can give a useful indication of the processes affecting or which have affected the deposits (Levinson *et al.*, 1992). ^{210}Pb and ^{210}Po have been used in uranium exploration but considerable care must be taken in the interpretation of the results since the data are ultimately a measure of the excess ^{222}Rn integration with respect to ^{222}Rn emanating power, and the ^{226}Ra specific activity at the site (Levinson *et al.*, 1982).

With the timescales involved in ore body generation, ^{210}Pb and ^{210}Po will be in equilibrium with each other in rocks and soils, but not necessarily with ^{238}U, as this will depend on the geochemical processes affecting the ore deposit, and the mobility of the decay series radionuclides.

1.b.ii Technological sources

1.b.ii.i Uranium Mining and Ore Processing

By far the largest source of technological enhancement of ^{210}Pb and ^{210}Po within the mining industry is in the extraction and processing of uranium ore. In 1979 more than 50 uranium mills processed over $65X10^6$ tons of ore (UNSCEAR, 1982), and in 1994 the production of uranium in the western world was $32.2X10^3$ t of which 55.3% came from Canada, Niger, Namibia, Australia and, South Africa.

The mining operations require the removal from underground, or, from excavated pits, large quantities of material containing uranium and its daughters, the concentrations of which can be more than three orders of magnitude higher than the average found in the terrestrial environment. The ore is crushed, and concentrated through flotation. Further processing occurs depending on the mineralogical composition of the ore. In general the ore is dissolved in sulphuric acid and the uranium is selectively removed from aqueous solution by ion exchange resins or by solvent extraction. The uranium is stripped from the extractant and the final product is normally ammonium diuranate, more commonly known as yellow cake. This result of this extraction process means that the yellow cake is virtually free of radioactive uranium daughters and has a uranium concentration between 65 - 70% (Choppin *et al.*, 1980). The yellow cake is then taken to fuel fabrication plants where it is further refined and the ^{235}U isotope may be enriched before being made into fuel rods.

The waste from the mining operations (tailings) is pumped from the mills as a slurry to the impoundment basin where the particles settle out. The tailings are radioactive and consist of about 85% of the total radioactivity originally present in the ore. At Elliot Lake in Canada, $8X10^6$ t of uranium ore was mined from estimated reserves of $1000X10^6$ t over a twenty year period. The tailings from this operation covered an area of $4X10^6$ m^2 and consist of coarse material (sand) deposited close to the discharge point and fine materials (slimes) closer to the decant point of the tailings dam (Moffett and Tellier, 1977).

Analysis of the tailings showed considerable variation in the levels of ^{226}Ra, ^{210}Pb and ^{210}Po but they were still much higher than average soil values. Mean values of ^{210}Pb and ^{210}Po within the coarse tailings were 1.3 Bq g^{-1} ^{210}Pb and 0.7 Bq g^{-1} for ^{210}Po, whilst for the fine tailings ^{210}Pb activities were 4.8 Bq g^{-1} and for ^{210}Po were 2.7 Bq g^{-1}. ^{226}Ra specific activities were higher being 11.6 Bq g^{-1} in the slimes and 6.1 Bq g^{-1} in the sands. In the early days of uranium mining at this area the liquid effluent from the tailings dam was discharged directly into the local water courses but this practice has now ceased and liquid effluent is treated to remove contamination prior to discharge (Moffett and Tellier, 1977).

Similarly at Ambrosia Lake, New Mexico, USA, a large uranium mining facility was opened in 1957 and was licensed to process 6X10^{6} kg d^{-1} of ore. Two large tailing piles are present covering an area of 1.41 km^{2}. Soil samples taken from two extensive areas close to the mines and tailings piles showed elevated levels of natural radionuclides compared to a control site (Lapham, *et al.*, 1989).

Atmospheric emissions of natural radionuclides also occur during the mining and processing of uranium ore, with the emission rates varying according to the plant type. Reported ranges of emissions from a typical mill processing 2000 t of ore per day are 1 - 4 GBq y^{-1} for ^{238}U, 0.2 - 2 GBq y^{-1} for ^{230}Th, ^{226}Ra and ^{210}Pb and 1 - 7 TBq y^{-1} for ^{222}Rn. Atmospheric emissions from dry tailings are of course reduced in ^{238}U and ^{234}U but are considerably higher for other natural radionuclides. Emissions are in the range 7 - 500 MBq y^{-1} for ^{238}U and ^{234}U, 0.1 - 8 GBq y^{-1} for ^{230}Th, ^{226}Ra and ^{210}Pb, and 500 - 300 TBq y^{-1} for ^{222}Rn. Tailings impoundment areas covered by water will have very low levels of radionuclide emission (UNSCEAR, 1993).

Once mining and milling operations cease, the tailings remain a source of contamination for some time and pose a considerable remediation problem. The principal source of contamination is ^{230}Th, which, because of its long half-life ($t_{½}$ = 8X10^{4} y) continues to produce ^{226}Ra and corresponding radon releases. A proportion of this will be lost to the atmosphere depending on the emanation rate from these areas so it is unlikely that the radon daughters will grow into equilibrium, although the transport of radon in the atmosphere can lead to possible enhancement away from the tailings areas but still within the locality. Considerable effort is being put into the stabilisation of spent uranium mine tailings to minimise erosion, leaching of radionuclides, and radon emissions, either by capping with clays or synthetic covers, or, by the use of sealents such as asphalts, so that these sites can be brought back into useful agricultural production (UNSCEAR, 1982).

Non-uranium mining can also be a source of enhancement of natural series radionuclides. Underground mines require ventilation not only as part of the requirements for a bearable working environment for the workers but also to remove toxic or inflammable gases from the air within the shafts. In some mines ^{222}Rn can build up and so the ventilation system may have to be augmented.

The levels of ^{222}Rn within the mines will vary according to the surrounding rock types, thus, in general, mines extracting ores within sedimentary deposits tend to have lower levels of radon gas compared with mines within igneous deposits because of the higher levels of uranium within these rock types. Also it is important to consider the geological and hydrological characteristics of the surrounding area when considering the likely levels of radon in mines (UNSCEAR, 1988).

Few data exist of the discharges of natural radionuclides from non-uranium mining facilities. Measurements of ^{222}Rn concentrations vary widely with mine type and are in the range 20 - 2900 Bq m^{-3} (UNSCEAR, 1988). Reported emissions for ^{222}Rn are in the range 0.24 to 8.5X10^{6} MBq y^{-1}. For ^{210}Pb and ^{210}Po emissions at a zinc mine were 0.01 and 0.006 MBq y^{-1} respectively. These emissions are lower than those of uranium mining facilities, as would be expected, but the issue of elevated amounts of ^{222}Rn from mine shafts and mine tailings, has the potential for enhancing the natural levels of its daughters ^{210}Pb and ^{210}Po, locally to the area. Elevated levels of ^{210}Pb in vegetation have been found up to 50 meters from uranium mine ventilation shafts (Bunzl *et al.*, 1994).

1.b.ii.ii Phosphate Ore Processing

Although phosphate ores can be either sedimentary, igneous or metamorphic in origin, about 85% of the known phosphorite reserves are sedimentary phosphates. The most easily mined deposits are those from the great sedimentary basins formed about 70 million years ago. Most sedimentary phosphates have high uranium concentrations but low thorium concentrations, whereas the reverse can be said of igneous deposits. Because of the age of the deposits, ^{238}U is in equilibrium with its daughters and, Table 1 shows some typical concentrations of natural radionuclides in some phosphorites.

The starting material for all phosphate containing products is phosphate rock, and it is the main source of phosphorous for fertilisers. Prior to its use in the production of phosphate products, phosphate rocks will usually undergo a benefication process to concentrate the phosphorous content of the ore. It has been reported that benefication can also concentrate natural series radionuclides to between 100 - 300% of that in the original ore (Habashi, 1980) and that the specific activities of uranium and ^{230}Th can increase with decreasing particle size (Metzger *et al.*, 1980).

The process of benefication involves washing, screening, and flotation of the ore, and, if the organic matter content is high, calcination is carried out to reduce it to 1%. The calcination process involves crushing and heating to 910°C, a process which can lead to the release of volatile ^{210}Pb and ^{210}Po to the atmosphere (Rutherford *et al.*, 1994). Normally the tailings from these processes are used as backfill in the original mines (UNSCEAR, 1988).

The ore concentrate is then used as the chemical feedstock in the production of phosphoric acid by either of two processes. These are the thermal process and the wet process. In the thermal process, elemental phosphorous is produced primarily for the production of high grade phosphoric acid to be further used in the production of phosphate fertilisers. By-products of the manufacture are slag and ferrophosphorous.

In the wet process the ore is reacted with sulphuric acid to produce gypsum (calcium sulphate), phosphoric acid, and hydrogen fluoride,

$$Ca_{10}(PO_4)_6F_2 + 10H_2SO_4 + 20H_2O \rightarrow 10CaSO_4 \cdot 2H_2O + 6H_3PO_4 + 2HF$$

(Apatite) (Gypsum)

Once the gypsum has crystallised it is separated from the acid, washed, and disposed as a slurry either to the sea or to a holding area where it may be further processed for use in agricultural or building products. About 90% of all phosphoric acid is produced by this method. It has been estimated that by the year 2000 the annual world-wide production of phosphogypsum will be in the region of 220 - 280 Mt (Ferguson, 1988).

Table 1 Reported natural radionuclide concentrations in phosphate rock (UNSCEAR, 1982)

Production of marketable rock in 1977		Specific Activity ($Bq\ kg^{-1}$)			
Country	Percentage of world production	^{238}U	^{226}Ra	^{232}Th	^{40}K
China	3.3	150	150	25	
Christmas Island	1.0	330	300	7	
Israel	1.0	1500-1700			
Jordan	1.4	1300-1850			
Morocco	14.0	1500-1700	1500-1700	20-30	10-200
Nauru	0.9	810	850	7	
Senegal	1.4	1300	1400	67	
Togo	2.3	1300	1200	110	<100
Tunisia	2.9	590	520	92	
USSR*	19.3				
Apatite		44-90	78-92	30-70	44-170
Phosphorite			25	390	230
United States	37.6	150-4800	150-4800	10-78	48

*Samples taken from the Kola Peninsula now part of the Russian Federation

The major source of radioactivity in phosphogypsum produced by the wet processes is ^{226}Ra. This is to be expected, as radium forms an insoluble sulphate and will therefore precipitate from the sulphuric acid solution used in the wet process. Because of its high insolubility, it is likely that all of the radium sulphate will have precipitated before the crystallisation of gypsum has begun. Some radium may also originate from phosphate rock particles which have survived the acid attack (Rutherford *et al.*, 1994) and become incorporated into the phosphogypsum waste stream. ^{210}Pb would tend to partition itself into the phosphoric acid stream whereas ^{210}Po , along with ^{226}Ra would partition into phosphogypsum. It has been estimated that 99% of ^{210}Po and 60% of the ^{226}Ra partition into the phosphogypsum fraction (Hurst and Arnold, 1982). Table 2 summarises some natural radionuclide concentrations in phosphogypsum produced from phosphate ores from different locations.

Table 2 Radionuclide activities in phosphogypsum (Rutherford *et al.*, 1994)

Location		**Radionuclide activities (Bq kg^{-1})**				
Phospho-gypsum	Phosphate rock	^{238}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Alberta	Idaho		890			
Australia	numerous	510	451-500			
Florida	Florida	93-190	100-2000	1270-1430	984-1480	3.7
Hungary	Syria		1093			
Louisiana	not given		700-1700			
Mississippi	not given		781		8.1	
Sweden	Kola		15			62
Yugoslavia	not given		390			

The distribution of some natural radionuclides with size fraction of phosphogypsum has also been studied. Rutherford *et al.,* (1996) found that the specific activities of ^{226}Ra and ^{210}Pb, and, the concentrations of uranium and thorium were enriched in the fine fraction (<20µm) relative to the bulk and the other size fractions of phosphogypsum produced from different ores. Enrichment factors for the fine fraction to the bulk sample were, 5.7±1.4 for ^{226}Ra and, 6.1±0.5 for ^{210}Pb, averaged from three samples. The specific activities in the size fractions are reproduced in Table 3.

Phosphogypsum itself has some industrial and agricultural uses. It is used as a dilutent within the fertiliser superphosphate, and as a soil amendment in agricultural soils, and, it is extensively used as a building material (Hussein, 1994). Because of the high levels of ^{226}Ra which can be found in phosphogypsum, some concern has been raised as to the possible radiological hazard from ^{222}Rn emanation from building materials particularly, within well insulated buildings (Arman and Seals, 1990).

Table 3 Mean concentrations and specific activities for some natural radionuclides in different size fractions of phosphogypsum (Rutherford *et al.*,1996)

Phosphogypsum origin	**Mean concentration or specific activity (U and Th μg g^{-1}, ^{226}Ra and ^{210}Pb Bq kg^{-1})**			
	^{238}U	^{232}Th	^{226}Ra	^{210}Pb
Idaho Rock				
Bulk	6.6	0.5	1160	901
Coarse	3.9	0.3	640	720
Medium	4.7	0.4	910	620
Fine	17.8	7.4	5120	5270
Togo Rock				
Bulk	1.5	3.5	610	560
Coarse	1.0	3.0	530	500
Medium	0.8	2.9	590	500
Fine	8.6	53.5	4270	3520
Florida Rock				
Bulk	2.0	1.1	690	620
Coarse	1.1	1.1	550	600
Medium	1.0	0.9	710	640
Fine	12.0	7.7	3460	3440

Aerial discharges from phosphate ore processing operations can be of two forms, either particulate emissions or as volatiles. It has been estimated by the US Environmental Protection Agency (USEPA) that particulate emission can be in the region of 0.1 kg t^{-1} of ore processed with most of this occurring during the transfer of rock at the plant (UNSCEAR, 1988). Most volatile emission of natural radionuclides occurs during the calcination process and in the production of elemental phosphorous by the thermal process, which are dominated by emissions of ^{222}Rn, ^{210}Pb, and ^{210}Po. Table 4 summarises the atmospheric discharges from phosphate industrial plants (UNSCEAR, 1988).

Table 4 Estimated atmospheric discharges of natural series radionuclides from phosphate industrial plants

Radionuclide discharges (GBq y^{-1})

Location	**Annual input of rock (10^{6} t)**	**^{238}U**	**^{230}Th**	**^{226}Ra**	**^{222}Rn**	**^{210}Pb**	**^{210}Po**
Ore drying and grinding. United States reference plant							
USA	2.7	0.6	0.6		4000	0.6	0.6
Elemental phosphorous plant							
Netherlands	0.75					150	350
USA	4	0.15	0.15	0.15	1500	4	330
USA	4	0.2	0.2	0.2	1100	210	780
USA	4	0.02	0.02	0.02	2000	4	26
USA	4	0.007	0.007	0.007	40	2	4
USA	4	0.07	0.07	0.07	300	15	22
USA	4	0.007	0.007	0.007	40	2	4
Wet process fertiliser plant. USA reference plant							
USA	1	0.25	0.25	0.25		0.13	0.13

^{222}Rn activities were estimated by UNSCEAR

Data on the environmental concentrations of ^{210}Pb and ^{210}Po in the vicinity of phosphate ore processing plants are rare, but, at an elemental phosphorous plant in the Netherlands, air concentrations of ^{210}Pb and ^{210}Po at a distance of 2km from the plant were, 0.1 and 0.4 mBq m^{-3} above background respectively. Similarly, at a site in Idaho, USA, which operated both wet and thermal processes the annual gross alpha activity was about 0.8 mBq m^{-3} at 800 m downwind from the plants. Most of this activity was attributed to ^{210}Po (UNSCEAR, 1982).

1.b.ii.iii Coal Fired Electricity Generating Power Stations

On a world-wide basis the principal method for generating electricity is by the combustion of coal in Thermal Power Stations. Although in recent years the proportion of electricity generated by fossil fuels has declined, it still accounts for 66.3% of global electricity generated. Within the United Kingdom the total declared net generating capacity for electricity is 102333 MW(e) (see table 5) of which 34840 MW(e) i.e. 34%, are produced by coal fired power stations (Electricity Supply Handbook, 1994).

Various estimates have been made as to the amount of coal consumed in the generation of electricity by thermal power stations. Camplin (1980) has estimated that a typical 2000 MW(e) power station operated by the then Central Electricity Generating Board would consume about $4.7X10^6$ t of coal, whilst Johnes (1980) estimated that a 1000 MW(e) power station would require about $3X10^6$ t. There is likely to be some variability in the amount of coal consumed and this will depend on a variety of factors such as, quality of coal, efficiency of electricity generation and operating practices.

The natural radioactivity content of coal has been studied by many workers, (Hamilton, (1974), Barnes and Ware, (1982), Salmon *et al.*, (1994), Rajan, *et al.*, (1995), Roeck *et al.*, (1987), Man-yin and Leung (1995), Styron, (1981)). Table 6 shows some typical values of the natural radionuclide content of coal, with particular emphasis on the levels found in coal mined in the U. K.. Most workers have found that the ^{238}U and the ^{232}Th decay series nuclides are essentially in secular equilibrium (Styron, (1981), Roeck *et al.*, (1987), Salmon et. al., (1984)). This is an important finding, particularly for the ^{238}U series, because of the possibility for disequilibrium within the series due to loss of radon by emanation from the coal deposit. In many deposits this does not seem to be occurring since radon is trapped within the coal deposit and is decaying further to its daughter products.

The levels of natural radionuclides found in coal can vary with coal type and location. The UNSCEAR report for 1982 ascribed global and average values to coal (see Table 6). More recent data by Rajan *et al.,* (1995) showed also that Indian Lignite has a larger range of concentrations in comparison to normal Indian coals. UK coals have values which lie within the global range but the average values are lower than the global average except for ^{40}K.

Table 5 Coal fired power stations within the UK in 1993

Operator	Name	Declared Net Capability (MW)	Operator Total	UK Total
National Power	Willington A	98		
	Blyth A	448		
	Staythorpe	336		
	Aberthaw A	188		
	Skelton Grange	224		
	Rugeley A	224		
	Uskmouth	224		
	Willington B	376		
	Blyth B	620		
	Thorp Marsh	1098		
	West Burton	1988		
	Eggborough	1954		
	Tilbury	1412		
	Ironbridge	984		
	Aberthaw B	1401		
	Rugeley B	1016		
	Didcot	2060		
	Drax	3890	18541	
Powergen	Castle Donington	564		
	High Marnham	930		
	Drakelow C	910		
	Ferrybridge C	1966		
	Ratcliffe	1974		
	Cottam	1970		
	Kingsnorth	1954		
	Fiddlers Ferry	1914	12182	
Scottish Power	Kincardine	375		
	Methil	57		
	Cockenzie	1152		
	Longannet	2304	3888	
Northern Ireland	Belfast West	229	229	34840

Table 6 Concentrations of some natural radionuclides in coal samples from around the world

	^{40}K (Bq kg^{-1})	^{238}U (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{210}Pb (Bq kg^{-1})	^{232}Th (Bq kg^{-1})
UNSCEAR (1982) World Average World Range	50 37-440	20 15-250	(20) 1.5-100	(20) 10-50	20 <7-110
Hamilton (1974) UK average (120 samples)	120	17			17
Camplin (1980) UK range		11-29	7.4-9.4		2.4-9.4
Barnes and Ware (1982) Yorkshire average (6 samples)	260	20	20	20	20
Salmon *et al* (1984) UK range UK average	55-314 150±58[♣]	7.8-30.0 14.1±5.1	7.8-25.5 14.5±4.6		7.0-19.2 11.75±3.30
Rajan *et al* (1995) India range coal India range lignite		5.7-56.8 0.7-99.5	6.7-54.4 BDL[♦]-109.9	4.9-48.6 1.1-85.3	2.6-50.1 BDL-87.7
Roeck *et al* (1987) USA range USA average (5 samples)		100-355 223	100-355 223	100-355 223	100-355 223
Man-yin and Leung Hong Kong average[♥]	24±12		17±7		20±9[*]

[♣] All errors are reported as 1σ

[♦] BDL means below detectable limit

[*] Reported as Ac-228 but in equilibrium with Th-232

[♥] Coals used in Hong Kong are of South African or Chinese origin

Combustion of coal in thermal power stations gives rise to gaseous and particulate pollution. Natural radionuclides released in the combustion process can find their way into the environment by two routes: by disposal of the solid waste ash, or by release to the atmosphere along with the flue gases through the stack. The atmospheric discharge can be divided into two phases, a gaseous phase consisting of radon gas and other flue gases, and a particulate phase which has natural radionuclides associated with it.

Modern coal burning power stations discharge very little particulate matter into the atmosphere. The introduction of electrostatic separators has greatly decreased the amounts of particulate fly ash released with the flue gases, and it has been estimated that about 99.5% of the fly ash is removed before reaching the environment through the atmospheric route. In the UK, Her Majesty's Inspectorate of Pollution, (now part of the Environment Agency) sets strict limits on the maximum concentration of particulate fly ash within the flue gases emitted from coal burning power stations. For a typical 2000 MW(e) power station this limit is set as 0.115 g m^{-3}, which is equivalent to a mass discharge rate of 220 g s^{-1} or $6.94X10^3$ t y^{-1}. If we further assume a typical load factor of 56%, then, in one year, $3.9X10^3$ t of particulate would be discharged through the flue. This is approximately 0.8 % of the total amount of coal burnt in a typical UK 2000 MW(e) coal burning power station.

Studies on the chemical composition of fly ash collected from electrostatic precipitators have found that some natural radionuclides are enriched in the material. In Roek *et al.,* (1987) extensive study of the partitioning of natural radionuclides in the waste streams of coal fired utilities, they found that both ^{210}Pb and ^{210}Po had the highest average enrichment factors of 11.0 and 11.2 respectively, with ^{238}U, ^{226}Ra, ^{230}Th, and ^{232}Th in the range 1.3 to 2.3. Rajan *et al.,* (1995) also found that fly ash had higher levels than the coal from which it was produced, although he did not calculate enrichment factors. Interestingly, lignite had the highest concentrations of natural radionuclides in its fly ash as compared to normal coal fly ash (Rajan *et al.*, 1995). In the coals used in Hong Kong power stations, Man-yin and Leung (1995) also found enrichment in the fly ash produced. Enrichment factors of 7.8 for ^{228}Ac, 8.2 for ^{226}Ra, and 7.4 for ^{40}K were calculated, although these factors came closer to unity when the radionuclide concentrations were normalised to ^{40}K content.

It is possible to estimate the amounts of radionuclides released in the fly ash from all UK coal fired power stations. Table 5 lists the coal fired power stations in the UK and their declared net capacity for 1993. As mentioned earlier, for a total net capacity of 102333 MW(e) for electricity generation, 34840MW(e) are generated by coal fired power stations. Taking the value of $3.9X10^3$ t of fly ash discharged into the atmosphere in a year from a 2000MW(e) station, then, for all UK coal fired stations, $67.9X10^3$ t of fly ash will be discharged into atmosphere. Using Roek *et al.,* (1987) average enrichment factors for ^{210}Pb and ^{210}Po of 11.0 and 11.2 respectively and taking a specific activity of 14.5 Bg kg^{-1} in coal for ^{210}Pb and ^{210}Po we can calculate the specific activity of these two nuclides in fly ash as being 159.5Bq kg^{-1} and 163.8Bq kg^{-1}. This gives us an estimated discharge of ^{210}Pb and ^{210}Po into the atmosphere of 1.08X10^{10} and 1.11X10^{10} Bq y^{-1} respectively. This can be compared to the amounts of these radionuclides in the coal burnt in power generation over a year, which is 1.15X10^{12} Bq for each radionuclide. Thus more than 100 times as much ^{210}Pb and ^{210}Po ends up in solid waste as is discharged to the atmosphere.

1.b.ii.iv Metal Smelting Processes

It is inevitable that volatile natural radionuclides are discharged into the atmosphere at the high temperatures found in metal smelting processes. The amounts discharged will of course depend on the initial concentrations of the natural radionuclides in the ore and the way they partition within the process streams prior to smelting.

Data on atmospheric emissions are scarce but, of that which is published, ^{222}Rn dominates with most being emitted at the mines themselves. Various estimates have been made as to the amounts of natural series radionuclides discharged into the atmosphere by metal smelting processes. At a zinc smelter in the USA, the annual releases of ^{210}Pb and ^{210}Po were 20 and 2 MBq respectively, whilst at an aluminium reduction plant they were 1200 and 1000 MBq. The highest values were at a copper smelter where the ^{210}Pb and ^{210}Po levels were 7000 MBq each. At a lead smelting plant in the USA, processing 0.22 Mt of ore, the estimated ^{210}Pb and ^{210}Po emissions were 1000 MBq for each nuclide (UNSCEAR, 1988).

In the UK the Capper Pass metal smelter at North Ferriby, Humberside was the subject of considerable investigation into its atmospheric emissions of ^{210}Po because of a putative link between the ^{210}Po content of the wastes and the incidence of leukaemia in the surrounding area (Baxter *et al.*, 1990, Kelly *et al.*, 1993). Over the period 1985 - 87 a total of 8329 MBq (average 2776 MBq y^{-1}) of ^{210}Po was discharged into the atmosphere. This corresponded to 1.67% of the ^{210}Po present in the raw material used in that period (Baxter *et al.*, 1990)

1.c Other Sources

In recent times the largest contribution of stable lead to the atmosphere in the UK is through the use of tetra-alkyl lead additives in petrol. Over the period 1984 to 1990, total emissions of lead in the UK have decreased from 8.2 kT to 3.2 kT, with that due to lead from vehicle emissions being 7.2 kT to 2.2 kT over the same period (Department of the Environment, 1993). The lead which is used in the manufacture of tetra-alkyl lead additives invariably contains ^{210}Pb which has been produced by decay of ^{238}U series radionuclides and has subsequently been concentrated into the ore by the ore making process. Jaworowski (1969) reported that the ^{210}Pb specific activity in the refined ore is about 800 Bq kg^{-1}. With the mean urban air concentration of stable lead in 1989/90 of 0.19 µg m^{-3}, Kelly *et al* (1993) have calculated the ^{210}Pb specific activity as being 0.15 µBq m^{-3}. Similarly, atmospheric deposition rates for stable lead have ranged between 0.046 - 11.5 mg m^{-2} d^{-1} (Harrison and Laxen, 1981) which is equivalent to 0.037 - 9 mBq m^{-2} d^{-1} with the largest being from the vicinity of a lead smelter (Kelly *et al.*, 1993).

The addition of phosphate containing fertilizer to land could result in enhancing the natural levels of radionuclides in soils. ^{226}Ra concentrations are elevated in phosphate fertilisers in comparison to natural soils but its effect on the soil concentrations of ^{210}Pb and ^{210}Po is considered negligible because of the partition of ^{226}Ra between the soil and the interstitial water and, the loss of ^{222}Rn by emanation and other fractionation processes (Pfister and Pauly, 1980).

At one time ^{210}Po was widely used in anti-static devices to remove dust from phonograph records, photographs and, other optical devices. At the time of manufacture each static eliminator contained 20 MBq of ^{210}Po. These devices are rarely used now and have largely been withdrawn from the public domain.

On the eighth of October 1957, during a routine operation to release Wigner energy from the Windscale Number One plutonium production reactor, the core temperature rose to such an extent that it caught fire. This event was not discovered until about two days later when instruments detected radioactivity reaching the air filters at the top of the air cooling discharge stacks. The following day the fire was successfully extinguished but in the mean time $7X10^{14}$ Bq of radioactivity was released into the atmosphere (Clarke, 1987). The principal radionuclide released was the short-lived ^{131}I, but 8.8 TBq of ^{210}Po was also released which was deposited in the surrounding countryside and found its way into the human food chain. The ^{210}Po was produced by the bombardment of Bi-209 for the neutron trigger then used in atomic weapons (Morgan, 1987).

In the savanna area of the Earth, 'forest fires' affect about 40% of the biomass. During the fire season in these areas, the specific activity of ^{210}Po in the aerosols can be increased to 1 mBq m^{-3} compared to normal background levels of about 0.02 mBq m^{-3}. The estimated flux of ^{210}Po from the African savanna by this process has been estimated as 0.14 PBq y^{-1} and for the Global savanna as 0.21 PBq y^{-1} (le Cloarec, *et al.*, 1995). Within the UK, the practice of straw burning is likely to enhance atmospheric levels of ^{210}Pb and ^{210}Po but no data are available to confirm this hypothesis. This practice will also cause an increase in the radiation dose received through the inhalation pathway, although any effect is likely to be more pronounced within the local area of the fires.

A major source of both ^{210}Pb and ^{210}Po in the environment is by the emission of these nuclides during volcanic activity. It has been estimated that the production rate is 44 TBq a^{-1} and 2 PBq a^{-1} for ^{210}Pb and ^{210}Po respectively. This is a contribution of about 50 - 60% of the total ^{210}Po in the atmosphere. Lee *et al.*, (1985) have estimated that the eruption of Mount St. Helens in 1980 released 1.2 PBq into the atmosphere.

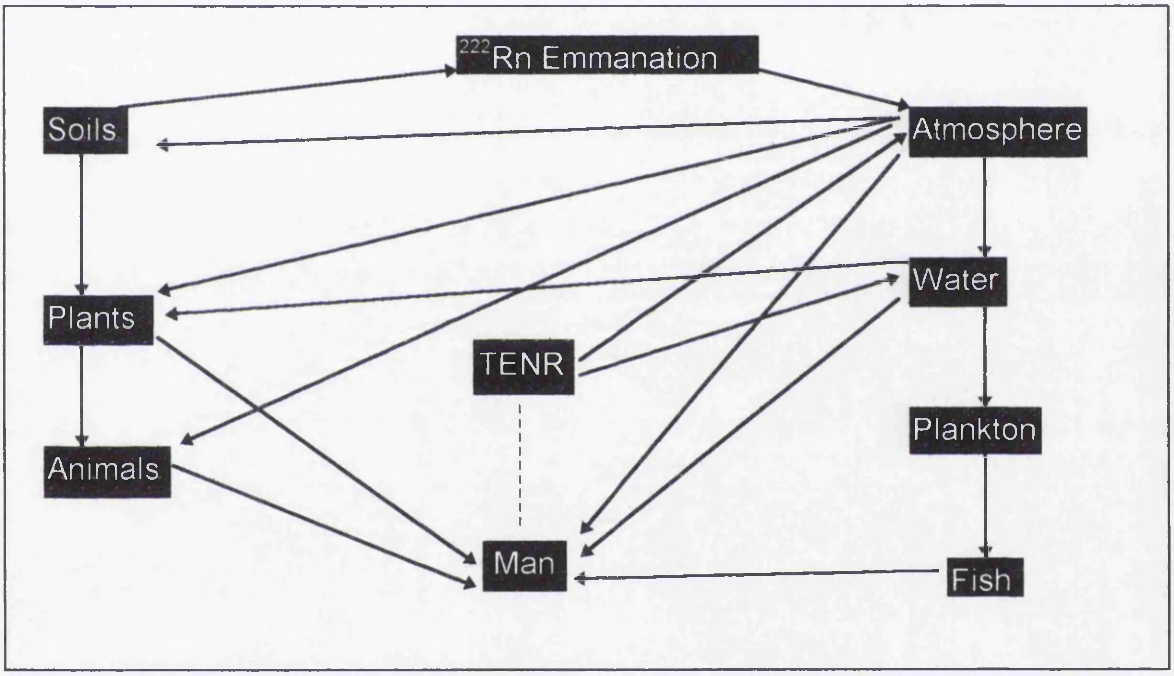
1d ^{210}Pb and ^{210}Po in the Terrestrial Foodchain and Dose Implications.

UNSCEAR (1982) has stated that:

> 'Consumption of food is usually the most important route by which ^{210}Pb and ^{210}Po enter the human organism'

and therefore, it is important that the environmental pathways and distribution of ^{210}Pb and ^{210}Po within the terrestrial foodchain are well understood. The main routes of uptake of ^{210}Pb and ^{210}Po by man are complex and are illustrated in Figure 2. Of prime importance for the terrestrial foodchain is the uptake by vegetation. This can occur by two routes. Either through fallout from the atmosphere onto the leaves of the plant, or through absorption by the plant roots from the soil. These processes depend on a number of factors which include the speciation of the radionuclide, the plant species, site specific environment dependent factors, and, soil to plant transfer factors.

Figure 2 The routes of uptake of ^{210}Pb and ^{210}Po by man



Although it has been well established that lead (and by inference ^{210}Pb) is absorbed through plant roots, up until recently the uptake of ^{210}Po in plants has been little studied. Berger *et al.*, (1965) concluded that ^{210}Po was not absorbed by plant roots because it occured in unavailable forms in soil, whereas Tso *et al.*, (1966) gave evidence for root uptake by the tobacco plant. In 1970, Hanson and Walters grew 9 species of vegetables in a greenhouse on soils to which was applied ^{210}Po. All tissues in the mature plants contained detectable levels of ^{210}Po except corn grain. They also noted that ^{210}Po specific activities in plants grown on non-contaminated soil were higher than could be accounted for by natural ^{210}Pb or ^{210}Po in the soil and they attributed this to fallout from the decay products of gaseous ^{222}Rn.

The influence of chelating agents on plant uptake of ^{51}Cr, ^{210}Pb and ^{210}Po from nutrient solutions and from a Ultisol (laterite) and a Vertisol (medium black) (Athalye, *et al.*, 1995) has been studied. Multidentate chelating agents ethylene diamine tetra-acetic acid (EDTA), ethylene diamine di-o-hydroxyphenylacetic acid (EDDHA) and diethylene triamine penta acetic acid (DTPA) were used, since these agents are extensively used to supply micro-nutrients to plants in agriculture. Both ^{210}Pb and ^{210}Po were also added in the nitrate form. In the nutrient culture study there was no significant increase in the uptake of complexed ^{210}Pb over the nitrate form whereas for ^{210}Po the opposite was observed. This was in contrast to the soil culture studies which showed that in the Ultisol the DTPA complexed forms of ^{210}Pb and ^{210}Po did not significantly contribute to the plant uptake in comparison to the inorganic forms but DTPA complexed ^{210}Po had significantly enhanced uptake in Vertisol compared to inorganic ^{210}Po.

Pietrzak-Flis and Skowronska-Smolak (1995) have studied the transfer of ^{210}Pb and ^{210}Po to plants via the root system and by above ground interception. They conducted experiments with potatoes, vegetables, cereals, and, fodder grown on two soil types in an open field and in a field sheltered by a polythene tent. The experiment showed that above ground interception of ^{210}Pb and ^{210}Po was an important contribution to the uptake of these radionuclides for grass, alfalfa, barley, spinach, lettuce and kale. The percentage contribution from aerial sources for these species of vegetation was in the range 65-95%. Root crops, potatoes, carrot, and turnip had a low contribution from aerial sources with ^{210}Po being the highest at 42%. Interestingly, wheat grain had a low contribution from aerial sources of 5% in contrast to the other grains studied.

Various methods have been used to express the accumulation of radionuclides in plants but all are based on some form of distribution co-efficient (Yamamotto, 1988). The simplest model is the Concentration Factor (CF), which uses the assumption that an equilibrium exists between the soil and the vegetation. This model is expressed by;

$$C_{v,i} = (CF)_{v,i} \times C_{s,i}$$

Where $C_{v,i}$ = the concentration of nuclide i in the vegetation v

$(CF)_{v,i}$ = the concentration factor of nuclide i ie. the concentration ratio of nuclide i in vegetation v to that in soil s

$C_{s,i}$ = the concentration of radionuclide i in soil s

Shepperd and Evenden (1988) have compiled a large number of concentration ratios for U, Th, and Pb which show a substantial variation in the values reported of 1000 to 30000 fold. Variations were also evident amongst plant species and soil types. The overall geometric mean of the concentration ratios of U, Th, and Pb were 0.0045, 0.0036, and 0.052 respectively. In a further experiment Shepperd *et al.* (1989) applied U, Th, and, ^{210}Pb to a variety of soils with different textures and organic matter content. They found the overall geometric means of the concentration ratios for U, Th, and ^{210}Pb to be 0.013, 0.0022, and 0.005 respectively, further reflecting the differences that can be found in the determination of concentration ratios.

As can be seen from the equation above, the form of the model assumes a linear relationship between the vegetation and the soil concentrations with the CF being the gradient of the line. This is a simplistic assumption which is not borne out by empirical data from many plant types and elements.

This lack of linearity has been well documented for a large number of elements used in plant nutrition and, in uptake studies of heavy metals and radionuclides (Simon and Ibrahim, (1987) and references therein). Most uptake studies have shown that the uptake response curve can take the form of a simple saturation function, and it follows therefore that the concentration ratio will be an exponential function, which decreases with increasing concentration of the substrate.

Mathematically this is described as:

$$\text{Plant Concentration} = f(\text{substrate concentration}) = f(s)$$

$f(s) = f_1\left(1 - e^{-f_2 s}\right)$ where f_1 and f_2 are numerical constants

and

$$\text{Concentration Ratio} = \frac{f(s)}{s} = \frac{f_1}{s}\left(1 - e^{-f_2 s}\right)$$

This can be further extended to the concept of multi-phasic saturation-type curves which can be described by successive saturation curves .

There are three basic ways in which plant uptake responds to substrate concentrations. They can be described as 'accumulator', 'indicator', or, 'excluder' responses.

In 'accumulator' species, plant uptake would in general follow a single or multi-phasic type response whereas in 'indicator' species the uptake response would be in the form of a linear function to the substrate concentration. Care must be taken in the interpretation of ' indicator' type response curves since they may be better described as 'accumulator' species depending on the magnitude of the saturation response. Finally in 'excluder' species, the plant response stays low until some threshold value is exceeded, after which the exclusion mechanism breaks down and the response may become unrestricted, or best be described by a saturation-type response.

In a study (Ibrahim and Wicker, 1986) on the transport of U series radionuclides in the environment of a U mine in Wyoming U.S.A., data on the uptake of ^{210}Pb and ^{210}Po in several plant species in areas of varying soil concentrations were established. To minimise the effect of aerial deposition, the plants used were subjected to ultra-sonic washing to remove soil particles adhering to the plant surface. The uptake response for ^{210}Pb indicated a saturation type behaviour and a $\log_e$ transform was fitted to the uptake response. The mathematical fit was:

$$\text{Plant Concentration (pCi g}^{-1}\text{ dry)} = 0.74\left(1 - e^{-1.4s}\right) + 0.16s$$

where s = substrate concentration

i.e. the uptake response has been modelled by the sum of a saturation function and a linear function. For the concentration ratio the fit was:

$$\text{Concentration Ratio} = \frac{0.74}{s}\left(1 - e^{-1.4s}\right) + 0.16$$

where s = substrate concentration

It was inferred from the form of the uptake response, that for ^{210}Pb, the plant behaved as an 'accumulator'. On the other hand the uptake response curve for ^{210}Po appeared to exhibit 'excluder' type behaviour. Uptake was low and constant up to approximately 40 pCi g^{-1} (1.48 Bq g^{-1}) although the concentrations were variable and difficult to model. A fit using a Gompertz function plus a constant was made of the data and was of the form;

$$\text{Plant Concentration} = ab^{-cb^{-ds}} + e$$

where $a, b, c, d,$ and e are numerical constants and,

s = substrate concentration

and

$$\text{Plant Concentration (pCi g}^{-1}\text{ dry weight)} = 70.0 \times 1.08^{-350 \times 1.08^{-0.45s}} + 1.2$$

Further evidence of 'excluder' type behaviour for ^{210}Po was provided from samples taken adjacent to a U mine tailings holdings pond. The soil in this area was saturated with pond derived water which was very acidic (pH 1.8)) and contained elevated concentrations of natural radionuclides. Again a Gompertz function was fitted and the resulting equation was:

$$\text{Plant Concentration (pCi g}^{-1}\text{ dry weight)} = 64.3 \times 1.14^{-430 \times 1.14^{-1.4s}} + 5.8$$

In this case the threshold value for ^{210}Po was approximately 15 pCi g^{-1} (0.55 Bq g^{-1}) and was attributed to the greater availability of ^{210}Po due to soil binding capacity caused by soil saturation at this site.

The authors could not ascribe any simple biological or physical interpretation to the use of the Gompertz function but clearly the uptake behaviour of the plants studied was quite different for both ^{210}Pb and ^{210}Po.

The derivation of CRs of radionuclides is important because of the crucial role they play in the decision making processes concerning radiological protection (Paschos and Amaral, 1990). They are widely used as input parameters along with other environmental transfer factors in a variety of dosimetric models. It is therefor important that CRs for as many plant species, sites, and soil types are determined so that reasonably accurate predictions of the specific activity of radionuclides in foodstuffs can be made. This is particularly true for artificial radionuclides which can be discharged into the environment under normal and emergency situations and much work has been done to this end (Ng, 1982,; International Union of Radioecologists, 1989).

With the exception of ^{226}Ra the database of CRs for naturally occurring radionuclides is not so extensive as that of artificial radionuclides and, as can be seen from the previous discussion on plant uptake much work still needs to be done on ^{210}Pb and ^{210}Po CR's.

Another source to humans of ^{210}Pb and ^{210}Po from the terrestrial foodchain is in the consumption of animal derived food products, in particular meat, milk and other dairy products. Animals in turn incorporate ^{210}Pb and ^{210}Po through consumption of feeds and forage and, through drinking water. Inhalation of ^{210}Pb and ^{210}Po could also be an important route, particularly for animals living close to atmospheric sources of these radionuclides, although little is known about the likely intake from this pathway.

Another potential source to animals is from the inadvertent consumption of soil associated with grass by grazing animals. This is estimated to be 4% and 20% of the dry matter intake for sheep and cattle respectively (Brown and Simmonds, 1995). Animals grazing on land with enhanced levels of ^{210}Pb and ^{210}Po could therefor ingest considerable amounts of these radionuclides but their availability from the soil particles may be much less than that biologically incorporated into grass. Still, the radiological consequences of consumption of foodstuffs derived from these animals by the human population are likely to be increased, compared to those from consuming animals grazed on background areas.

Similarly with the CR concept in plants an empirically derived ratio can be used to describe the transfer of radionuclides from feed to edible animal products. Many such transfer products have been formulated but the two most useful are the milk transfer coefficient f_l and the meat transfer coefficient f_m. These coefficients represent the fraction of the total daily intake of a radionuclide that is transferred to the animals milk (f_l) or incorporated into the animals muscle (f_m) at equilibrium. The units of f_l are d l^{-1} and those of f_m d kg^{-1}.

Again the data for these coefficients are limited for ^{210}Pb and ^{210}Po. For cows milk values for f_l of 2.6×10^{-4} and 3.4×10^{-4} d l^{-1} for ^{210}Pb and ^{210}Po respectively have been reported. Values of 10×10^{-4} d kg^{-1} for ^{210}Pb in beef and 40×10^{-4} d kg^{-1} for ^{210}Po in reindeer have been reported (Linsalata, 1994). Poultry has been recorded at 24000×10^{-4} d kg^{-1} for ^{210}Po although the error on this value was ± 16000×10^{-4} d kg^{-1} (Izak-Biran *et al.*, 1989). Smith-Briggs (1984), has reported f_m for cattle livers in the range 1.2×10^{-3} to 9.6×10^{-3} d kg^{-1} for ^{210}Pb and 1.1×10^{-2} to 4.9×10^{-2} d kg^{-1} for ^{210}Po. Even with the paucity of data above, it is obvious that meat transfer coefficients for both ^{210}Pb and ^{210}Po are very much species and organ dependent.

More data are obviously needed on transfer factors and CRs for ^{210}Pb and ^{210}Po considering the importance of their role in environmental modelling.

1.e Behaviour of ^{210}Pb and ^{210}Po in the Human Body

An understanding of the behaviour of ^{210}Pb and ^{210}Po in the human body is an important pre-requisite for elucidating the distribution of the likely radiation dose from the ingestion of foodstuffs containing these radionuclides. Both behave differently in the environment (as outlined above) and in the human body. Although the major route into the human body would be by direct intake some would also be present from the decay of ^{226}Ra since they are the end members of the decay chain. This would only be important in occupational exposure such as in uranium mining where there is the possibility of enhanced ingestion by inhalation of dust particles.

^{210}Pb is a bone seeker being retained mainly in bone mineral for long periods. Approximately 70% of the body burden is found within the skeleton (UNSCEAR, 1982). Jaworowski (1969) reported levels of ^{210}Pb in the UK population in the range 0.67 - 0.96 Bq kg^{-1} although more recent work (Bradley and Fry, 1989) has found specific activities in femur samples of 1.3 ± 0.5 Bq kg^{-1} and 1.2 ± 0.6 Bq kg^{-1} in rib samples. That which is not incorporated into the bone is fairly evenly distributed throughout the rest of the body.

On the other hand ^{210}Po tends to accumulate in the soft tissue with higher levels particularly in the liver, kidney and, spleen. Typical specific activities found in the liver are in the range 0.5 - 1.0 Bq kg^{-1} with that of kidney being slightly lower at 0.3 - 0.8 Bq kg^{-1}. The ^{210}Po:^{210}Pb activity ratio in these organs can be considerably greater than 1 reflecting the preferential uptake of ^{210}Po. This has been attributed to the distribution of ^{210}Po being similar to that of sulphur and thus ^{210}Po may be able to replace sulphur in sulphur containing compounds in the body (UNSCEAR, 1988). In the other organs ^{210}Po and ^{210}Pb specific activities are similar resulting in ^{210}Po:^{210}Pb activity ratios in the range 0.8 - 1.0 (Bennett and Sandalls, 1991). The biological half-life of ^{210}Po has been estimated as 50 days (ICRP 30, 1979).

For inhabitants of the Arctic Region, ^{210}Po and ^{210}Pb specific activities can be considerably higher than other population groups, reflecting their increased dietary intake of these radionuclides through consumption of reindeer and caribou meat. Levels in Arctic populations can be 2 - 3 times higher for ^{210}Po and ^{210}Pb in bone, and 10 times higher in soft tissue for ^{210}Po (UNSCEAR, 1982).

1.f Objectives

There are two primary objectives in this study, these are:

a) To characterise the specific activities of ^{210}Po and ^{210}Pb in terrestrial foodstuffs cultivated in areas within England and Wales, which have the potential for enhancement from technological activity, and

b) To estimate the likely doses to the UK population through consumption of these foodstuffs.

Implicit in these primary objectives are a number of related activities.

To be able to decide if enhancement has occurred, comparisons must be made to foodstuffs which have been cultivated in areas which are considered to be background areas away from technological activity. Similarly, comparisons must be made with foodstuffs grown on areas known to be enhanced in natural radioactivity. It is also important to provide a sound statistical basis for these decisions but for this to be successful many samples are required.

Likewise in estimating the radiation dose through ingestion, data on consumption habits of different age groups are essential. Ideally, site specific data on consumption habits would be used to give the most accurate assessment of radiation dose, but this is not available. In this case national statistics on food consumption data will be used in the dose calculations.

Chapter 2 Sampling and Analysis

2.a Sampling

2.a.i Site Selection

As has been outlined above, one of the objectives of this study, was to investigate the potential for enhancement of ^{210}Pb and ^{210}Po in a variety of foodstuffs. Thus the selection of sites had to encompass those areas where the potential for enhancement of these two nuclides from natural and technological sources could occur. Within England and Wales, a survey of sites of natural and technological sources of ^{210}Pb and ^{210}Po was carried out, from known and published data. These data were used to pinpoint sites within the local area of the potential sources of TENR.

It is important that sampling points are chosen to be in close proximity to the site of possible enhancement. From modelling studies of atmospheric deposition, it has been found that the ground level airborne concentrations of pollutants reach baseline levels by about 10 km from a point source. In addition , there is an area close to the source, known as the skip distance, where the ground level airborne concentrations are little affected by discharges. This is up to about 500 m from the source. After this distance, the concentrations decline to baseline levels. The position of maximum ground level airborne concentration depends on a large number of factors such as weather conditions, stack height, the complexity of the terrain, coastal effects, the proximity of buildings, and the physical and chemical form of the pollutant (UKADMS, 1995). The modelling of atmospheric processes has become increasingly sophisticated and complicated, but the very general description in the preceding sentences, has outlined the typical results of these modelling exercises.

When selecting sites for sampling it was decided to choose areas which were a distance of between 0.5-5 km from the expected source of possible enhancement. This was considered to maximise the chance of finding enhanced levels in foodstuffs if they were present. It was also important to select sites from areas where it was unlikely to find enhanced levels of ^{210}Pb and ^{210}Po, so that comparisons could be made as to the levels found, thus giving a measure of the effect or not of any enhancement. These control sites were selected from areas of high and low rainfall in case the natural levels of ^{210}Pb and ^{210}Po varied with the effect of increased wet deposition in areas of high rainfall.

Table 7 Sampling Sites and Annual Rainfall Data

Site	Source	Nearest weather station	Rainfall (mm)	Percentage of the 1951-1980 average
Drax	Coal fired power station	Linton-on-Ouse	422	
Penrith	High rainfall control	Newton Rigg	1000	113
Newmarket	Low rainfall control	Cambridge	452	82
Sellafield	Nuclear facility	Sellafield	995	
Whitehaven	Phosphate ore processing	Sellafield	995	
Warminster	Low rainfall control	Larkhill	692	86
Brecon	High rainfall control	Bronydd Mawr	1424	
Avonmouth	Metal smelter	Bristol weather centre	739	
Holyhead	Metal smelter	Amlwch Anglesey	766	
Didcott	Coal fired power station	Benson	529	
Helston	High natural radon area	Lizard	740	
Romford	Metal smelter	Greenwich	507	85

Nearest weather station and annual rainfall data were taken from the Monthly Weather Summary (The Meteorological Office, 1991).

2.a.i.ii Control Sites

Four control sites were selected on the basis of their rainfall. The high rainfall areas were at Penrith in Cumbria and Brecon in Wales, and the low rainfall areas were Newmarket in Suffolk and Warminster in Wiltshire.

2.a.i.iii Sites of Natural Sources of ^{210}Pb and ^{210}Po

An indication of areas where there are likely to be enhanced levels of naturally occurring radionuclides can be found by measuring the atmospheric concentration of radon gas. An extensive study of the radon levels in dwellings in England and Wales has been carried out by the National Radiological Protection Board, and they have mapped its distribution by county and by postal code area. Samples for radon from 92000 dwellings were taken and it was found that the average concentration of radon was 21 Bq m^{-3} with some values several hundred times higher. Of the dwellings measured about 12000 were above the Government Action Level of 200 Bq m^{-3} and some were considerably higher (Green et al., 1992). Most of the high radon results were in Cornwall and Devon, with Derbyshire, Northamptonshire and Somerset being the next most affected counties.

Similarly, measurement of the uranium and thorium concentrations in soils would also be a useful indicator of the possibility of enhanced levels of natural series radionuclides in an area. In a survey of soils from Cornwall and Sutherland, mean levels of uranium and thorium were 3.5 and 1.5 times higher than the UK average. Uranium concentrations in Cornwall soils were variable and fell into two groups. Most were in the range 2.3 - 13 ppm but two samples were greater than 120 ppm. Samples taken in close proximity to a disused mine were high and variable, with uranium concentrations in the range 265 - 2020 ppm (Nicholson et al., 1990).

Cornwall, historically, was an area of extensive mining of copper and tin and it was not unusual to find uranium bearing minerals in these mines (Dines, 1930). Considerable amounts of uranium ore were mined at Trenwith and St. Steven, and it has been reported that ‘ores have been sold by hundred of tons’ (Collins, 1912a, 1912b).

It was decided to sample within Cornwall in areas where radon levels were high, and where there was evidence of past mining activity.

2.a.i.iv Technological Sources of ^{210}Pb and ^{210}Po

There are numerous areas of industrial activity within England and Wales which are potential sources of ^{210}Pb and ^{210}Po. Of these the most important and of particular relevance to this study are, metal smelting, coal fired power stations, and phosphate ore processing plants.

As mentioned earlier, previous studies have shown that appreciable amounts of natural radioactivity are released into the atmosphere from the burning of fossil fuels in power stations, in particular coal fired stations. From Table 5 it can be seen that the two largest coal fired power stations in England and Wales are the Drax power station in North Yorkshire, and the Didcot power station in Oxfordshire, which were the obvious candidates for study.

Metal smelting processes also release considerable amounts of natural radionuclides into the environment. The metal smelter at North Ferriby in Humberside (which closed in the early 1990's) was the subject of a considerable study into its emissions to the atmosphere (Kelly et al., 1993, Baxter et al., 1990).

Other sites where metal smelting occurs are at Carless Refining and Marketing Ltd, at Romford in Essex, Rio Tinto Zinc at Avonmouth near Bristol, and the Alcan aluminium smelter in Anglesey in Wales. Of these, Carless refining are authorised by HMIP, to discharge ^{210}Po into the atmosphere up to certain amounts. In 1993 this installation emitted 6.36X10^{8} Bq into the atmosphere which was just below its authorisation level of 6.78X10^{8} Bq.

No data were found for emissions from either the Alcan or Rio Tinto Zinc installations but, they were included in this study along with the Carless Smelter, since MAFF had previously sampled at these sites as part of its environmental monitoring program around non-nuclear sites.

Phosphate ore processing plants have been known to discharge large amounts of natural radioactivity through their liquid discharges into the marine environment (McCartney et al., 1990, McDonald et al., 1992, Carvalho, 1995, UNSCEAR, 1977, 1982, 1988). In West Cumbria, the Albright and Wilson phosphoric acid plant at Marchon has been studied as to its aqueous discharges to the Irish Sea, although nothing has been published as to its aerial discharges.

From the early 50's until 1992, the Marchon plant had been producing phosphoric acid using the hemihydrate wet phosphoric acid process from imported Moroccan phosphate ore which contained approximately 120ppm uranium (1440 Bq kg^{-1}). Phosphogypsum and acid wastes were discharged to the Irish Sea under authorisation from MAFF because of the elevated levels of radionuclides in the wastes (Poole et al., 1995). Over the period 1972-1992 the plant discharged 739 t of uranium into the Irish sea compared to 70t discharged by the nuclear fuel reprocessing plant at Sellafield operated by British Nuclear Fuels (BNF) (Poole et al., 1995). After 1992, the liquid discharges from this plant decreased considerably, since raw phosphate ores have stopped being used in the plant, because of the cessation of the wet process, to be replaced with crude phosphoric acid (known as green acid) instead.

There is anecdotal evidence to suggest that when the plant was using the wet process, considerable amounts of dust were dispersed into the surrounding area, whilst the ore was transported between Whitehaven docks and the plant (Stewart, pers comm). Considering the uranium concentration of the ore, there is the possibility of enhancement by this pathway. The area around this plant was sampled.

As a comparison it was decided that it would be useful to sample foodstuffs from an area close to a nuclear facility. Sellafield, is the largest nuclear fuel reprocessing plant in Western Europe and is operated by BNF. It was considered to be a suitable candidate since its discharges are higher than any other UK nuclear facility. Samples were taken from farms surrounding the Sellafield site.

The sites which were sampled, are tabulated in Table 7 along with the rainfall data from the closest weather station to each site, and their positions are marked on figure 3. Where possible, as many sample types as were available, were collected from one particular farm, although in many cases this was not possible, so other farms were visited.

Figure 3 Terrestrial foods sampling sites

2.a.ii Sample Type Selection

There has been one other study of the levels of ^{210}Pb and ^{210}Po in UK foodstuffs, and this was from total diet samples taken as part of a MAFF programme to evaluate the intake of trace metals and pesticides by the UK population (Smith-Briggs et al., 1984b). These foodstuffs were prepared as for consumption according to standard procedures, divided into twenty groups as determined by the National Food Survey, and, homogenised. Due to the length of time the samples were in storage, ^{210}Pb and ^{210}Po were in virtual equilibrium.

In this study, sample types were chosen according to their ability and potential to concentrate the radionuclides of interest, and their importance in the UK diet. Thus, liver was collected because of its ability to concentrate a variety of radionuclides including ^{210}Pb and ^{210}Po, cereal crops, fruit and milk, because of their importance in diet, particularly of the young, leafy green vegetables for their potential to concentrate radionuclides through atmospheric deposition to their leaf surfaces and, root vegetables because their radionuclide uptake is almost exclusively from the soil.

It was particularly important, when collecting liver samples, that the animal from which the liver came from, had lived on the farm since birth, or at least one year at the farm to equilibrate the input and output of ^{210}Pb and ^{210}Po from the animal. Also it was important to trace the chosen animal through the abattoir so that the liver could be sampled. The assistance of State Veterinary Officers is gratefully acknowledged in this respect. In remote areas many local butchers assisted in the procurement of suitable samples. Livers were collected from both cattle and sheep depending on what was available at the sites.

Table 8 Sample Types Collected at Sampling Sites

Site	Milk	Offal	Leafy Green Vegetables	Cereals	Fruit	Potatoes	Root Vegetables
Penrith	-	+	+	+	-	-	-
Whitehaven	-	-	-	+	+	-	+
Sellafield	+	-	+	+	+	+	-
Drax	-	-	-	+	+	-	-
Holyhead	-	+	+	+	+	-	-
Brecon	-	-	+	+	-	-	-
Newmarket		+	+	+	-	-	-
Avonmouth	-	-	-	-	-	+	-
Warminster	-	-	-	+	-	-	-
Didcot	-	-	-	+	-	+	-
Romford		-		+	-	+	-
Helston	-	+	+	-	-	-	+

It was not possible to collect a complete suite of samples from each site because of the different farming practices prevalent throughout the UK. Table 8 shows the samples which were collected from each site.

2.b Analysis of samples for ^{210}Pb and ^{210}Po

2.b.i.i Sample Preparation

Prior to chemical analysis of the samples for ^{210}Pb and ^{210}Po, some preparation of the samples was performed. On receiving samples in the laboratory after a sampling trip arrangements were made for their storage. Vegetation samples were stored in a cold store at 4^{0}C whilst offal and milk samples were frozen in a freezer at -20^{0}C until required for analysis.

Initial cleaning of the samples to remove dust and soil particles prior to sample digestion varied with sample type. For offal and milk samples no cleaning was required. Cereal products were cleaned by shaking the grains over a 2mm sieve to allow dust and small particles to fall through the mesh, larger particles such as stones and stalks were laboriously hand picked from the mass of the grains. The root vegetables potatoes, carrots, and turnips were rinsed under tap water to remove adhering soil particles and then thinly skinned (approximately 1-2mm). With leafy green vegetables the outer leaves were removed as these had invariably been attacked by insects or were diseased, or had soil particles contaminating the surface. The fruit samples, apples, pears, and blackberries were washed under tap water to remove dust particles on the skin and dried with non-coloured paper towels. With apples and pears, the fruits were deseeded but the skins were not removed prior to digestion.

The decision to remove skin, or not, was based on food practices in the household. Thus root vegetables and leafy green vegetables were skinned, or, the outer leaves removed, but fruits were not as these are commonly eaten whole.

Normally 4 to 5 times the analytical weight was taken for cleaning. This was then homogenised either by chopping into small pieces in the case of vegetation samples, or by mincing in a blender in the case of liver samples. Grain samples were homogenised by shaking, and milk, by mixing to ensure that the cream was evenly dispersed throughout the liquid.

2.b.i.ii Analytical Procedure

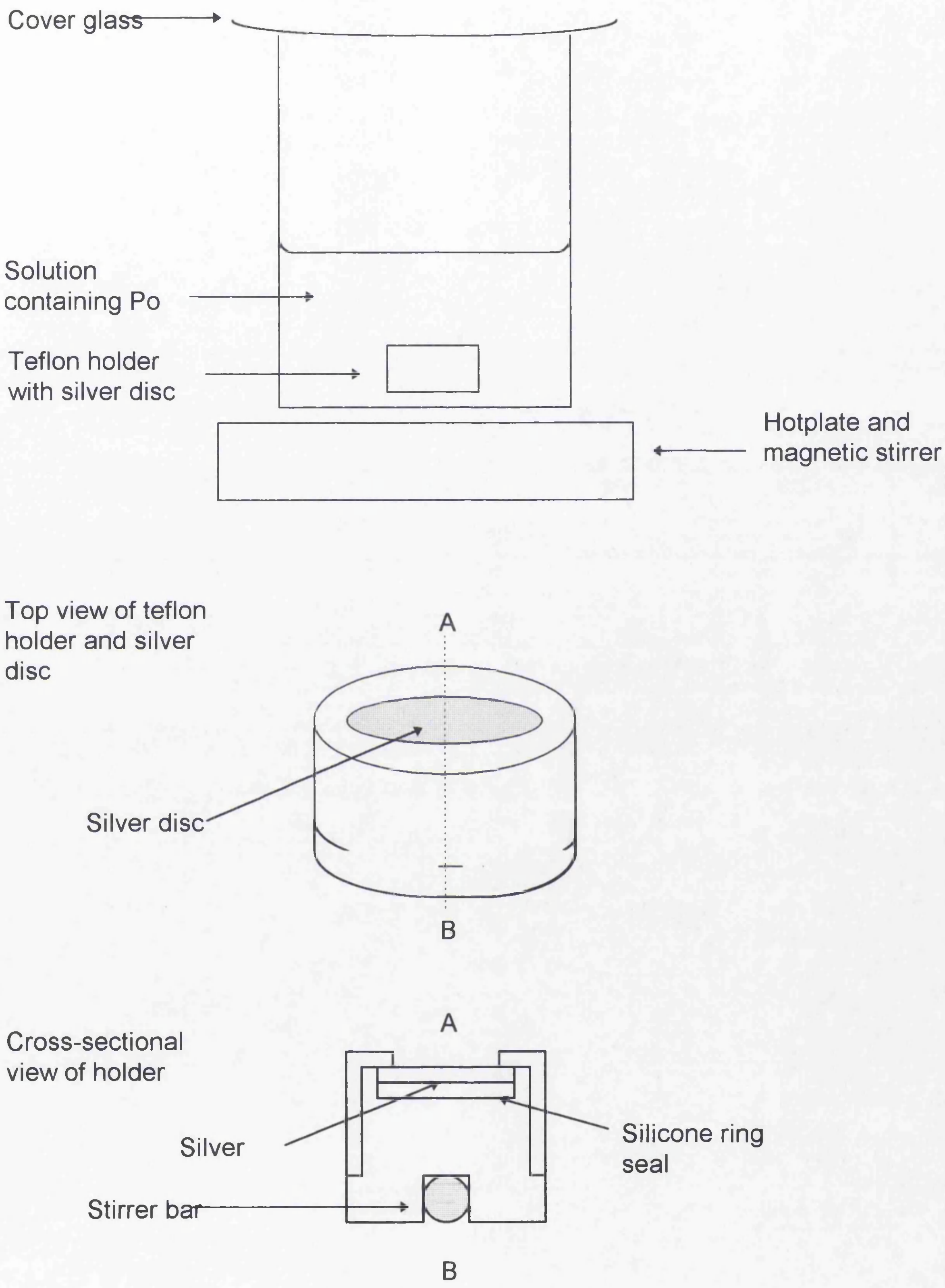
Most methods for the analysis of ^{210}Po and ^{210}Pb have been developed for samples of marine origin i.e. sediment and biota. In general this has entailed separating the ^{210}Po from the matrix followed by its detection using alpha spectroscopy. For ^{210}Pb, three other methods are available for its detection. These are either detection of the β emission of its daughter ^{210}Bi by either gas proportional counting (Church, et al., 1994, Moser, 1993) or liquid scintillation counting (Momoshima *et al.*, 1994), detection of its low energy γ emission using high purity germanium detectors (GeLi) (Lagarwaard and Woittiez, 1994), or by allowing the grand daughter product ^{210}Po to grow in after its initial separation. Marine samples have higher levels of ^{210}Po and ^{210}Pb than is expected to be found in terrestrial foodstuffs from the UK and therefore smaller amounts are required for analysis. Although the widely adopted method for the analysis of ^{210}Po by plating onto silver disks has been utilised in this study, (Flynn, (1968), Fleer and Bacon, (1984), MacKenzie et al., (1979)) the method of dissolution has had to be extended but at the same time kept as simple as possible.

An aliquot of the homogenised sample was weighed accurately into a 5 litre beaker and spiked with a known quantity of ^{208}Po spike. Sample weights were quite large and varied according to the expected level of ^{210}Po in the sample being analysed. Approximately 200g of cereals and liver were taken, whilst for the other sample types somewhere between 500 and 750g was used. With milk, normally 1.5 to 2 litres was taken, which was usually the complete sample. The ^{208}Po spike was prepared, by serial dilution, from a stock solution of activity 5.01±0.07 kBq g^{-1} purchased from AEA Technology. Alpha spectrometric purity of the solution was quoted as, ^{208}Po = 99.4%, ^{209}Po = 0.6%, ^{210}Po = < 0.01%.

500ml of 15.4M nitric acid "Analar" was added to the sample and left covered in a fume cupboard overnight without heating. This was because some sample types, particularly cereals, reacted violently if heated initially, producing large amounts of CO_2 and nitrous oxide fumes, with the froth overflowing from the beaker resulting in sample loss. Leaving the sample overnight obviated this effect, and by the next morning the partially digested sample could be safely heated and the digestion process taken further. The temperature of the hotplate was kept below 150^{0}C and the beaker covered throughout the digestion process to minimise loss of analyte.

As the reaction subsided more 15.4M nitric acid was added in 250 ml aliquots to continue the digestion process further, and once the majority of the organic material was destroyed 40% hydrogen peroxide was slowly added in 25 ml aliquots to decompose the more resistant organic compounds. This was performed until complete dissolution had occurred.

Figure 4 Plating apparatus



Sample types with large amounts of fat such as offal and milk were treated slightly differently. After the initial digestion with nitric acid a fatty layer was present on the surface of the solution. This fat was removed after cooling and further leached three times with 15.4 nitric acid and 40% hydrogen peroxide, and the leachate returned to the original solution for further processing. This was considered necessary to remove ^{210}Po and ^{210}Pb from the fat which was borne out by the high yields of the polonium spike recovered.

Once complete digestion was obtained, the sample was taken to almost dryness, and 10ml of 11M hydrochloric acid "Analar" was added to convert the residue to the chloride form, and, remove any residual nitric acid which could interfere with the plating process which followed.

The dissolved sample was transferred quantitatively to a 600 ml beaker and the volume reduced to between 100-150 ml. Approximately 1 g of hydroxylamine hydrochloride was added to reduce any iron present to iron(II). This held back the iron in solution whilst the pH was brought to between 1.5 and 2 using specific gravity 0.880 ammonia solution.

On reaching the required pH the sample was heated to about 95^{0}C and a polished 25 mm diameter silver disk in a teflon holder was placed into the solution. This holder (see Figure 4) had a magnetic stirring bar in its base so that the holder could be rotated rapidly on a magnetic/stirrer hotplate. Rapid stirring and high heating ensures even deposition of polonium onto the surface of the disk giving good resolution when the disk is alpha counted (Marckwald, 1905, Hamilton and Smith, 1986). Heating also improves the kinetics of the plating process. The beaker was positioned off-centre on the hotplate to disrupt the vortex caused by the rapid stirring, to ensure that the silver disk was always covered by solution.

After 4 hours, the teflon holder was removed from the solution, and disassembled. The silver disk was then rinsed with de-ionised water, then acetone, and left to air dry prior to alpha counting.

The plating solution was quantitatively transferred to a 250ml plastic bottle and stored for at least three months to allow ingrowth of ^{210}Po from ^{210}Pb in the solution. After this time, the plating process was repeated, after the addition of fresh spike, thus enabling the Pb-210 concentration to be calculated from its grand-daughter's ingrowth. Once this value is found, the original ^{210}Po concentration can be calculated, and corrections made for the decay and ingrowth of ^{210}Po between sampling and analysis. Blank determinations were carried out in tandem with the samples and all specific activities were blank corrected.

2.b.i.iii Alpha Spectroscopy

The α-particles emitted from the surface of the silver were detected using a silicon surface barrier detector. Because the α-particles are relatively massive and highly energetic, they interact strongly with matter giving up their energy over very small distances. The detectors have therefore to be housed in a vacuum chamber and the measurement done under vacuum.

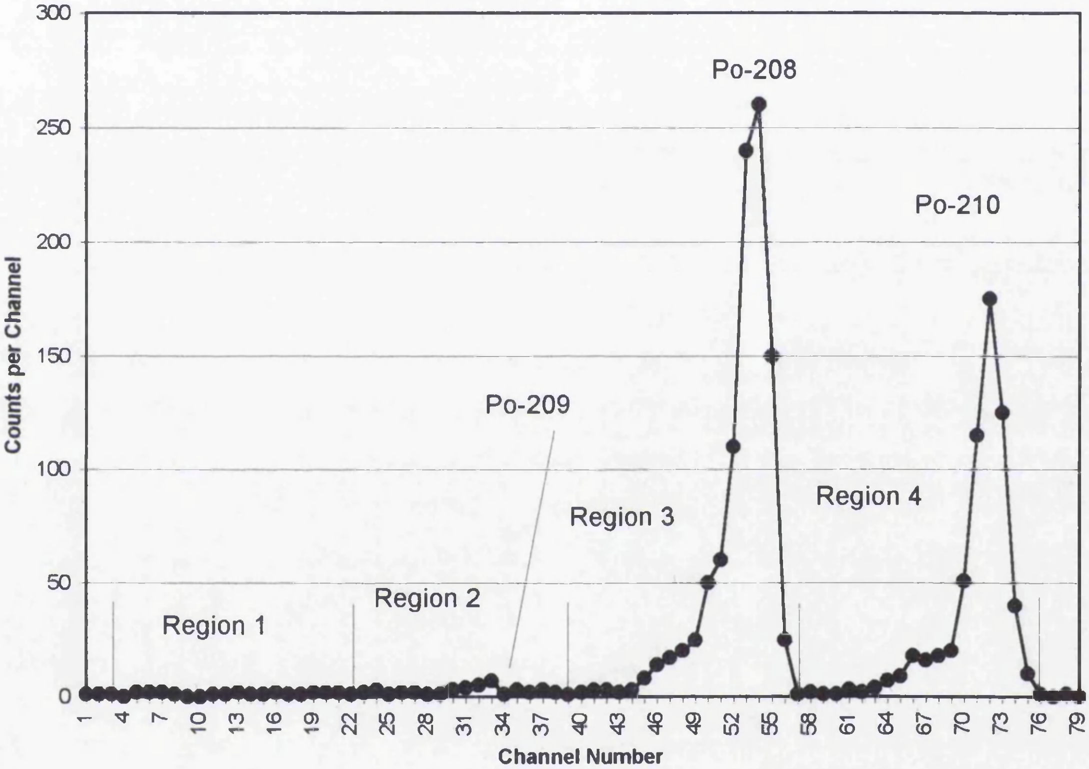
Surface barrier detectors consist of a wafer of n-type conducting material, which has been exposed to air and then coated with a thin gold film, forming a surface layer in which p-type conduction occurs. When a charged particle is incident on the detector, it dissipates its energy in the sensitive surface region of the detector, generating electron-hole pairs in the conduction and valence band. The number of promotions produced is proportional to the particle energy. Under an applied bias (10-100V) the charge carriers migrate to the electrodes forming a pulse.

An EG&G ORTEC OCTÊTE™ Octal Alpha Spectrometer with eight ORTEC ULTRA™ 450 mm^2 surface barrier detectors was used to count the polonium alpha particles. The alpha spectra were transferred to a Compuadd 325s computer and displayed using ORTEC Maestro II software.

Both ^{210}Po and ^{208}Po are mono-energetic alpha emitters of energies 5.304 MeV and 5.116 MeV respectively which leads to relatively simple α-spectra. A typical spectrum is shown in Figure 5.

The α-spectra generated generally had good separation because of the resolution between the polonium peaks. A correction was made to correct for tailing of the ^{210}Po peak into the ^{208}Po peak which was inevitable. In general, the correction was small in relation to the ^{208}Po spike peak, but, it was important in correcting the ^{210}Po peak.

Figure 5 Alpha spectrum of Po-208 and Po-210 showing Po-209 impurity and regions of interest.



The correction method used, was derived from Fleer and Bacon (1984) and is illustrated in Figure 5. It is assumed that the shapes of the pure ^{208}Po and ^{210}Po peaks are identical, and therefore the shape of the ^{208}Po tail can be used to infer the shape of the ^{210}Po tail. The sample spectrum is subdivided into 4 equal regions of interest designated 1-4. The minimum between the two peaks is first found and the number of channels between the minimum and the ^{208}Po maximum is noted. That number of channels is taken to the right of the ^{210}Po peak and is used to locate the upper boundary of region 4. The lower boundary is taken as the minimum of the two peaks, and the other three regions are taken to span the same number of channels. A further assumption is that the counts in regions two and three but not one are contributed by the low energy tail of the ^{210}Po peak.

Successive approximations are produced by iteration until the change in the estimated ^{210}Po was less than 0.1 counts. A small correction was made for the contribution of ^{209}Po which was an impurity in the ^{208}Po spike. The detector background was subtracted from each of the regions before the iteration process was carried out. Appendix 2 has the computer program which was used to carry out the iteration calculation and the calculation of the ^{208}Po and ^{210}Po concentrations.

No suitable reference materials were found for method validation, but during the study I participated in a blind intercomparison organised by AEA Technology (AEAT) on behalf of the Ministry of Agriculture Fisheries and Food (MAFF). The samples analysed consisted of spiked and unspiked freeze dried cabbage, milk and offal and the results were in very good agreement with the organising laboratory. These are reproduced in table 9 below.

Table 9 Summary of ^{210}Pb and ^{210}Po Intercomparison Results

	Spike Level	**AEAT Mean**	**AEAT SD**	**Westlakes**
		^{210}Pb (Bq kg^{-1}) Dry Weight		
Unspiked Milk	None	0.161	0.079	0.080
Unspiked Cabbage	None	0.205	0.093	0.247
Unspiked Liver	None	0.135	0.045	0.194
Spiked Milk	5.30	6.074	1.322	5.530
Spiked Cabbage	4.00	4.531	0.885	3.750
Spiked Liver	21.00	20.33	0.911	21.90
		^{210}Po (Bq kg^{-1}) Dry Weight		
Unspiked Milk	None	0.116	0.059	0.075
Unspiked Cabbage	None	0.226	0.060	0.206
Unspiked Liver	None	0.199	0.036	0.230
Spiked Milk	3.20	3.436	0.204	3.320
Spiked Cabbage	2.40	2.713	0.290	2.600
Spiked Liver	12.60	11.394	1.645	12.27

Chapter 3 ^{210}Pb and ^{210}Po Specific activities in foodstuffs

3.a Results of the Survey

With such an amount of data from a variety of sample types and sampling points the results are presented in a series of tables to aid comparison between sites, sample types and, activity ranges.

Table 10 ^{210}Pb and ^{210}Po Specific Activities According to Site (Bq kg^{-1} ± 1 σ)

Sample Site	Sample Type	^{210}Po	Error	^{210}Pb	Error	Po/Pb	Error
Avonmouth	Turnip	0.002	0.001	0.008	0.001	0.25	0.13
Brecon Beacon	Barley	0.230	0.002	0.42	0.005	0.55	0.01
Brecon Beacon	Barley	0.120	0.05	0.32	0.03	0.37	0.16
Brecon Beacon	Brussels Sprouts	0.034	0.001	0.1	0.003	0.34	0.01
Brecon Beacon	Cabbage	0.062	0.002	0.191	0.005	0.33	0.01
Cornwall	Bovine Liver	2.956	0.006	0.515	0.016	5.20	0.18
Cornwall	Brocolli	0.027	0.001	0.066	0.004	0.40	0.03
Cornwall	Cabbage	0.070	0.001	0.078	0.002	0.89	0.03
Cornwall	Cabbage	0.0035	0.0002	0.0041	0.003	0.85	0.63
Cornwall	Turnip	0.003	0.001	0.022	0.002	0.16	0.05
Didcot	Barley	0.192	0.003	0.215	0.007	0.89	0.03
Didcot	Potato	0.002	0.001	0.007	0.001	0.26	0.15
Didcot	Wheat	0.273	0.04	0.08	0.003	3.40	0.52
Drax	Apple	0.026	0.001	0.101	0.004	0.25	0.01
Drax	Barley	0.120	0.004	0.486	0.015	0.41	0.01
Drax	Pear	0.061	0.001	0.152	0.004	0.40	0.01
Drax	Pumpkin	0.001	0.0003	0.012	0.001	0.11	0.03
Drax	Wheat	0.031	0.001	0.078	0.002	0.41	0.02
Holyhead	Barley	0.303	0.004	0.23	0.009	1.32	0.05
Holyhead	Barley	0.379	0.007	0.49	0.01	0.78	0.02
Holyhead	Blackberry	0.470	0.002	0.238	0.007	1.97	0.06
Holyhead	Blackberry	0.376	0.002	0.278	0.008	1.35	0.04
Holyhead	Cabbage	0.003	0.001	0.013	0.001	0.24	0.08
Holyhead	Ovine Liver	0.740	0.006	1.6	0.02	0.46	0.01
Holyhead	Ovine Liver	0.586	0.003	0.99	0.01	0.59	0.01
Newmarket	Barley	0.421	0.003	0.39	0.01	1.08	0.03
Newmarket	Bovine Liver	0.794	0.004	0.186	0.001	4.28	0.03
Newmarket	Lettuce	0.136	0.003	0.138	0.004	0.98	0.04
Newmarket	Wheat	0.009	0.002	0.111	0.007	0.08	0.02
Penrith	Barley	0.008	0.003	0.448	0.004	0.02	0.01
Penrith	Bovine Liver	0.866	0.004	0.548	0.012	1.58	0.04
Penrith	Cabbage	0.044	0.002	0.223	0.007	0.20	0.01
Romford	Barley	0.197	0.002	0.19	0.01	1.02	0.06
Romford	Potato	0.001	0.0001	0.005	0.001	0.20	0.04
Sellafield	Apple	0.055	0.001	0.119	0.005	0.46	0.02
Sellafield	Apple	0.092	0.002	0.091	0.008	1.01	0.09
Sellafield	Barley	0.631	0.006	0.68	0.02	0.92	0.03
Sellafield	Cabbage	0.148	0.003	0.267	0.006	0.55	0.02
Sellafield	Cabbage	0.090	0.001	0.16	0.006	0.56	0.02
Sellafield	Milk	0.010	0.001	0.003	0.001	3.10	1.16
Sellafield	Potato	0.013	0.001	0.01	0.001	1.30	0.16
Sellafield	Wheat	0.116	0.003	0.35	0.012	0.33	0.01
Warminster	Barley	0.092	0.003	0.12	0.011	0.76	0.07
Whitehaven	Barley	0.427	0.005	0.689	0.018	0.62	0.02
Whitehaven	Blackberry	0.016	0.001	0.153	0.006	0.11	0.01
Whitehaven	Turnip	0.006	0.001	0.006	0.001	1.00	0.24

Table 10 lists the specific activities in foodstuffs by site, Table 11 lists the specific activities in foodstuffs by type and Table 12 lists the ranges of specific activities by type.

Table 11 ^{210}Pb and ^{210}Po Specific Activities According to Food Type (Bq kg^{-1} ± 1 σ)

Sample Site	Sample Type	^{210}Po	Error	^{210}Pb	Error	Po/Pb	Error
Drax	Apple	0.026	0.001	0.101	0.004	0.25	0.01
Sellafield	Apple	0.055	0.001	0.119	0.005	0.46	0.02
Sellafield	Apple	0.092	0.002	0.091	0.008	1.01	0.09
Brecon Beacon	Barley	0.23	0.002	0.42	0.005	0.55	0.01
Brecon Beacon	Barley	0.12	0.05	0.32	0.03	0.37	0.16
Didcot	Barley	0.192	0.003	0.215	0.007	0.89	0.03
Drax	Barley	0.12	0.004	0.486	0.015	0.41	0.01
Holyhead	Barley	0.303	0.004	0.23	0.009	1.32	0.05
Holyhead	Barley	0.379	0.007	0.49	0.01	0.78	0.02
Newmarket	Barley	0.421	0.003	0.39	0.01	1.08	0.03
Penrith	Barley	0.008	0.003	0.448	0.004	0.02	0.01
Romford	Barley	0.197	0.002	0.19	0.01	1.02	0.06
Sellafield	Barley	0.631	0.006	0.68	0.02	0.92	0.03
Warminster	Barley	0.092	0.003	0.12	0.011	0.76	0.07
Whitehaven	Barley	0.427	0.005	0.689	0.018	0.62	0.02
Holyhead	Blackberry	0.47	0.002	0.238	0.007	1.97	0.06
Holyhead	Blackberry	0.376	0.002	0.278	0.008	1.35	0.04
Whitehaven	Blackberry	0.016	0.001	0.153	0.006	0.11	0.01
Cornwall	Bovine Liver	2.956	0.006	0.515	0.016	5.2	0.18
Newmarket	Bovine Liver	0.794	0.004	0.186	0.001	4.28	0.03
Penrith	Bovine Liver	0.866	0.004	0.548	0.012	1.58	0.04
Cornwall	Brocolli	0.027	0.001	0.066	0.004	0.4	0.03
Brecon Beacon	Brussels Sprouts	0.034	0.001	0.1	0.003	0.34	0.01
Brecon Beacon	Cabbage	0.062	0.002	0.191	0.005	0.33	0.01
Cornwall	Cabbage	0.07	0.001	0.078	0.002	0.89	0.03
Cornwall	Cabbage	0.0035	0.0002	0.0041	0.003	0.85	0.63
Holyhead	Cabbage	0.003	0.001	0.013	0.001	0.24	0.08
Penrith	Cabbage	0.044	0.002	0.223	0.007	0.2	0.01
Sellafield	Cabbage	0.148	0.003	0.267	0.006	0.55	0.02
Sellafield	Cabbage	0.09	0.001	0.16	0.006	0.56	0.02
Newmarket	Lettuce	0.136	0.003	0.138	0.004	0.98	0.04
Sellafield	Milk	0.01	0.001	0.003	0.001	3.1	1.16
Holyhead	Ovine Liver	0.74	0.006	1.6	0.02	0.46	0.01
Holyhead	Ovine Liver	0.586	0.003	0.99	0.01	0.59	0.01
Drax	Pear	0.061	0.001	0.152	0.004	0.4	0.01
Didcot	Potato	0.002	0.001	0.007	0.001	0.26	0.15
Romford	Potato	0.001	0.0001	0.005	0.001	0.2	0.04
Sellafield	Potato	0.013	0.001	0.01	0.001	1.3	0.16
Drax	Pumpkin	0.001	0.0003	0.012	0.001	0.11	0.03
Avonmouth	Turnip	0.002	0.001	0.008	0.001	0.25	0.13
Cornwall	Turnip	0.003	0.001	0.022	0.002	0.16	0.05
Whitehaven	Turnip	0.006	0.001	0.006	0.001	1	0.24
Didcot	Wheat	0.273	0.04	0.08	0.003	3.4	0.52
Drax	Wheat	0.031	0.001	0.078	0.002	0.41	0.02
Newmarket	Wheat	0.009	0.002	0.111	0.007	0.08	0.02
Sellafield	Wheat	0.116	0.003	0.35	0.012	0.33	0.01

Table 12 Ranges of Specific Activities of ^{210}Pb and ^{210}Po Found in the Foodstuffs

	^{210}Po (Bq kg^{-1})			^{210}Pb (Bq kg^{-1})			
Sample Type	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Count
Apple	0.026	0.092	0.058	0.091	0.119	0.104	3
Barley	0.008	0.631	0.260	0.120	0.689	0.390	12
Blackberry	0.016	0.470	0.287	0.153	0.278	0.223	3
Bovine Liver	0.794	2.956	1.539	0.186	0.548	0.416	3
Brocolli	0.027	0.027	0.027	0.066	0.066	0.066	1
Brussels Sprouts	0.034	0.034	0.034	0.100	0.100	0.100	1
Cabbage	0.003	0.148	0.060	0.004	0.267	0.134	7
Lettuce	0.136	0.136	0.136	0.138	0.138	0.138	1
Milk	0.010	0.010	0.010	0.003	0.003	0.003	1
Ovine Liver	0.586	0.740	0.663	0.990	1.600	1.295	2
Pear	0.061	0.061	0.061	0.152	0.152	0.152	1
Potato	0.001	0.013	0.005	0.005	0.010	0.007	3
Pumpkin	0.001	0.001	0.001	0.012	0.012	0.012	1
Turnip	0.002	0.006	0.004	0.006	0.022	0.012	3
Wheat	0.009	0.273	0.107	0.078	0.350	0.155	4

The specific activities of ^{210}Pb and ^{210}Po in the samples ranged over three orders of magnitude. For ^{210}Pb, the lowest specific activity found was 0.003 Bq kg^{-1} in milk and the highest 1.600 Bq kg^{-1} in ovine liver, and for ^{210}Po the lowest was 0.001 Bq kg^{-1} in potato and pumpkin, and the maximum being 2.956 Bq kg^{-1} in bovine liver.

Within the individual sample types the ranges of specific activities of ^{210}Pb and ^{210}Po found were different. For vegetation samples there is a clear distinction in the levels found between foodstuffs grown below ground and those grown above. Below ground foodstuffs have low specific activities reflecting the low concentration factors for plants as discussed in Chapter 1. In above ground foodstuffs this would still apply but with the added input of ^{210}Pb and ^{210}Po from atmospheric sources. This effect is likely to be enhanced on those foodstuffs which present a high surface area on which particles can be trapped. This is particularly true for foodstuffs with coarse or fibrous surfaces e.g. cereals, leafy vegetables and some fruits. Although pumpkin is grown above ground its specific activities of ^{210}Pb and ^{210}Po are low even though there is the possibility of atmospheric input. Transfer through the skin is unlikely since pumpkin has a very thick and waxy skin which would provide an effective barrier to uptake through this route.

For the animal products milk and liver, specific activities of ^{210}Pb and ^{210}Po in these samples lie at ether end of the spectrum. Uptake in the liver samples would be expected to be high in the light of the high transfer factors f_m for ^{210}Pb and ^{210}Po as discussed in Chapter 1 and indeed since the f_m for ^{210}Po is higher than that for ^{210}Pb the activity ratio ^{210}Po: ^{210}Pb should be greater than 1. This is seen in samples for bovine liver but not for ovine liver. Although only one milk sample was assayed some comment can be made on the levels of ^{210}Pb and ^{210}Po found. Again with reference to Chapter 1 the milk transfer factor f_l is very low so it would be reasonable to assume that the specific activities of ^{210}Pb and ^{210}Po would be low.

With the exception of cabbage, the ranges of specific activity for ^{210}Pb were less than an order of magnitude with the smallest being a factor of approximately two for blackberry, potato, and ovine liver, whereas for ^{210}Po the ranges of specific activity were almost two orders of magnitude, except for ovine liver which was just greater than a factor of one. This is illustrated in Figures 6 and 7.

For the activity ratios of ^{210}Po:^{210}Pb, a large range of values is found in the sample types except for ovine liver. This sample type was only collected at one site, Holyhead, and the closeness of the ratio may be due to both animals being reared in close proximity to each other. Since the ^{210}Pb and ^{210}Po specific activities are also relatively close this may reflect the likely variability in these measurements at this site. The ranges are tabulated in Table 13 and shown in Figure 8.

Figure 6 Ranges of ^{210}Pb Specific activities in Different Sample Types

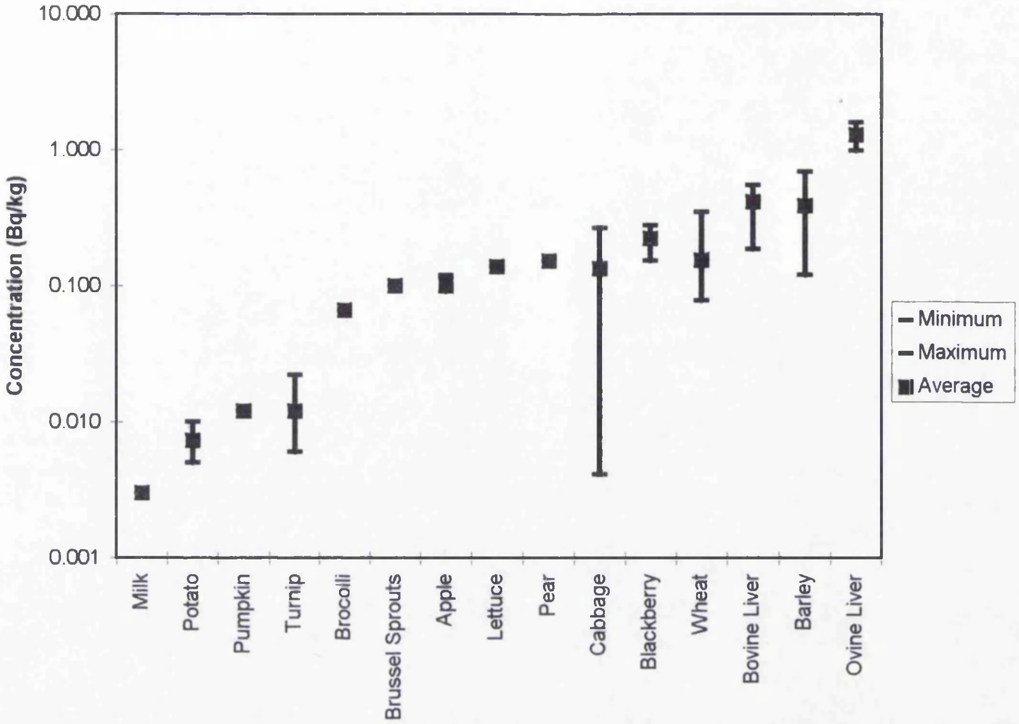


Figure 7 Ranges of ^{210}Po in Different Sample Types

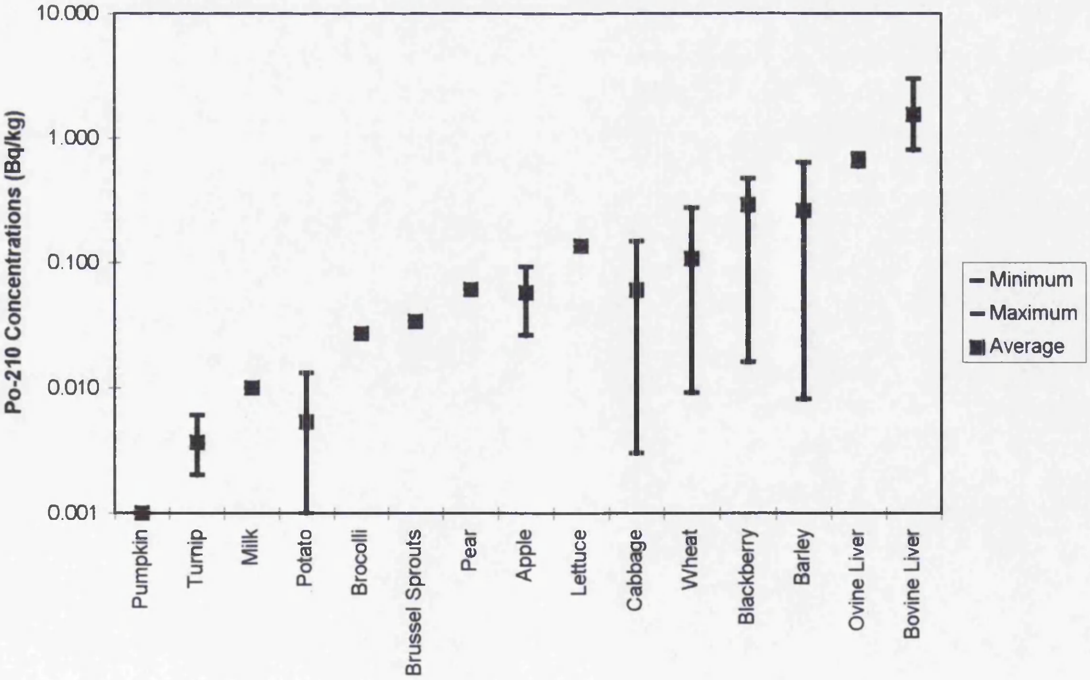


Figure 8 Ranges of ^{210}Po:^{210}Pb Activity Ratios in the Different Sample Types

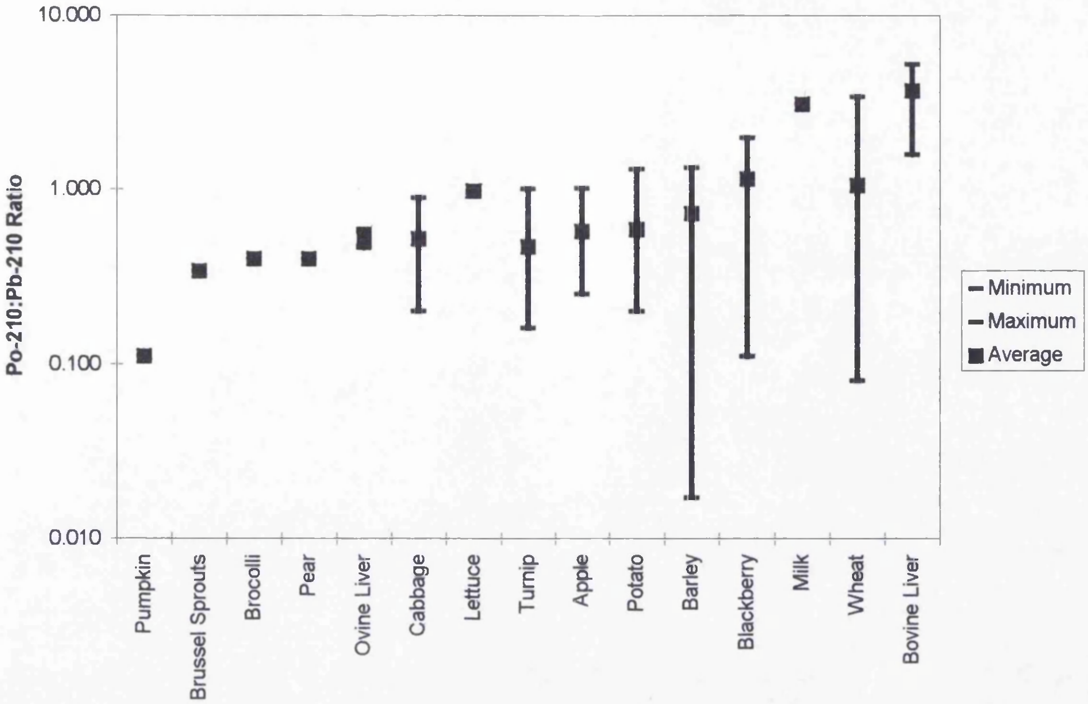
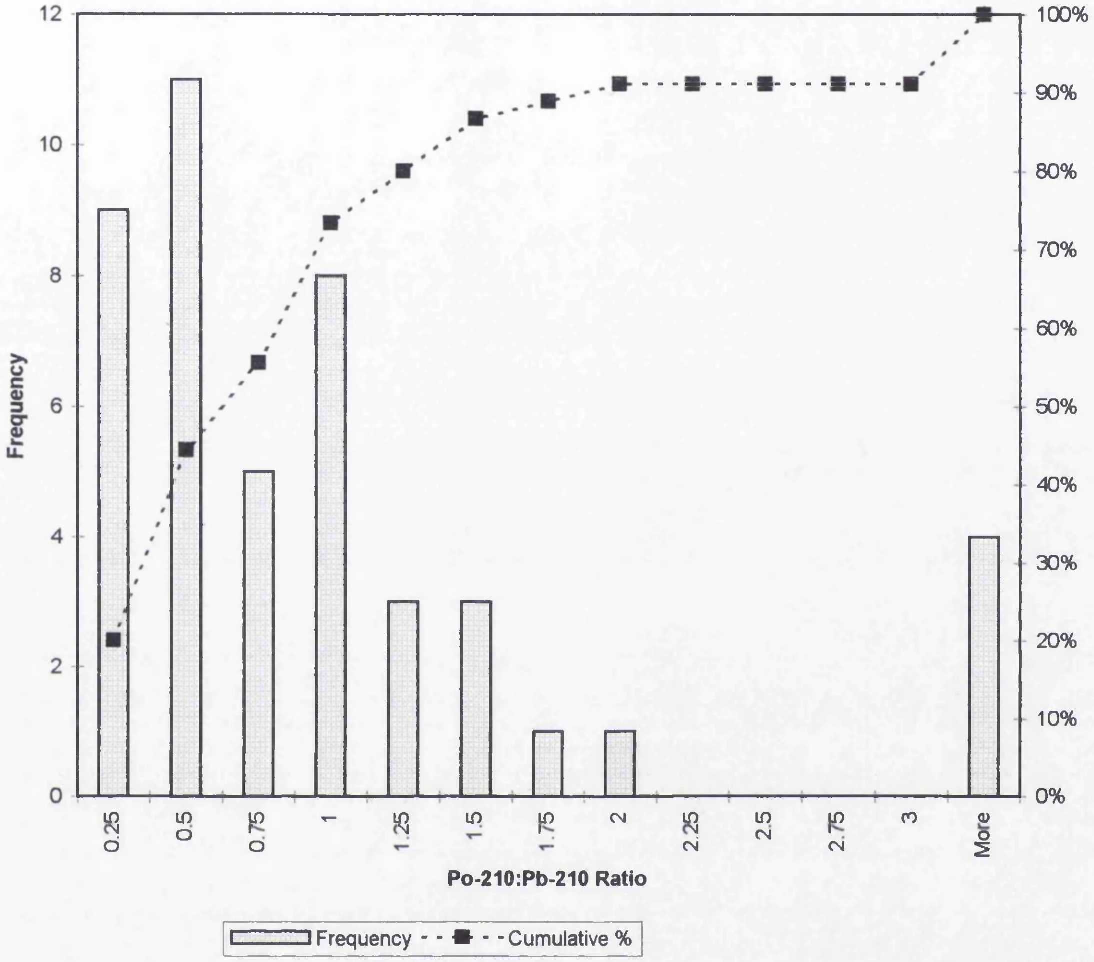


Table 13 Range of ^{210}Po:^{210}Pb Activity Ratios in Different Sample Types

Sample Type	Minimum	Maximum	Average
Apple	0.250	1.010	0.573
Barley	0.017	1.320	0.728
Blackberry	0.110	1.970	1.143
Bovine Liver	1.580	5.200	3.687
Broccoli	0.40	0.40	0.40
Brussels Sprouts	0.34	0.34	0.34
Cabbage	0.200	0.890	0.517
Lettuce	0.98	0.98	0.98
Milk	3.10	3.10	3.10
Ovine Liver	0.460	0.590	0.525
Pear	0.40	0.40	0.40
Potato	0.200	1.300	0.587
Pumpkin	0.11	0.11	0.11
Turnip	0.160	1.000	0.470
Wheat	0.080	3.400	1.055

If we consider the overall distribution of the ratios we can see that it is positively skewed with a mode of 0.5, a median of 0.55, and the mean 0.92. We can also see that 26% of the samples have a ratio greater than 1. This can be interpreted as a measure of the degree of disequilibrium within the sample types, particularly the liver samples, but care must be taken with this interpretation because of the various routes of the uptake of ^{210}Pb and ^{210}Po by plants and animals, and ultimately the concentration and transfer factors as discussed in Chapter 1. A frequency histogram of the ratios is plotted in Figure 9.

Figure 9 Frequency Histogram of ^{210}Pb:^{210}Po Ratios of all the Samples



3.b Statistical analysis

In attempting a statistical analysis of the results it is difficult to come to firm conclusions due to the limitations in the sampling strategy. The ideal situation would have been a number of replicate samples for each foodstuff at each site but this was not possible particularly as not all foodstuffs were cultivated at each site. Nevertheless some comparisons can be made between sample sites and sample types. The maximum specific activities of each of the different foodstuffs by site are tabulated in Table 14 below.

Table 14 Maximum ^{210}Pb and ^{210}Po Specific Activities (mBq kg^{-1}) in Foodstuffs by Site

Sample Type	Brecon		Newmarket		Penrith		Warminster	
	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb
Apple								
Barley	0.230	0.420	0.421	0.390	0.008	0.448	0.092	0.120
Blackberry								
Bovine Liver			0.794	0.186	0.866	0.548		
Broccoli								
Brussels Sprouts	0.034	0.100						
Cabbage	0.062	0.191			0.044	0.223		
Lettuce			0.136	0.138				
Milk								
Ovine Liver								
Pear								
Potato								
Pumpkin								
Turnip								
Wheat			0.009	0.111				

Sample Type	Avonmouth		Cornwall		Didcot		Drax	
	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb
Apple							0.026	0.101
Barley					0.192	0.215	0.120	0.486
Blackberry								
Bovine Liver			2.956	0.515				
Broccoli			0.027	0.066				
Brussels Sprouts								
Cabbage			0.070	0.078				
Lettuce								
Milk								
Ovine Liver								
Pear							0.061	0.152
Potato					0.002	0.007		
Pumpkin							0.001	0.012
Turnip	0.002	0.008	0.003	0.022				
Wheat					0.273	0.080	0.031	0.078

Sample Type	Holyhead		Romford		Sellafield		Whitehaven	
	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb
Apple					0.092	0.119		
Barley	0.379	0.490	0.197	0.190	0.631	0.680	0.427	0.689
Blackberry	0.470	0.278					0.016	0.153
Bovine Liver								
Broccoli								
Brussels Sprouts								
Cabbage					0.148	0.267		
Lettuce								
Milk					0.010	0.003		
Ovine Liver	0.740	1.600						
Pear								
Potato			0.001	0.005	0.013	0.010		
Pumpkin								
Turnip							0.006	0.006
Wheat					0.116	0.350		

This Table serves not only as a useful comparison of the specific activities of ^{210}Pb and ^{210}Po between sites but also as a comparison of the specific activities themselves in particular food types.

Although the data are sparse because of sampling difficulties and availability at each of the sites, we can see that there are definite variations in specific activities in particular food types between sites (see also Figures 6 and 7).

An attempt was made to rank the sites according to the specific activities of ^{210}Pb and ^{210}Po in the foodstuffs analysed. The sites were ranked according to the maximum specific activity of ^{210}Pb and ^{210}Po in each food type and are tabulated below (Table 15) for food types which were collected at more than two sites.

Although this method was less than satisfactory in ranking the sites according to maximum ^{210}Pb and ^{210}Po specific activities some observations can be drawn from this exercise. The method shows that for both ^{210}Pb and ^{210}Po the control sites are not necessarily ranked low, as might be expected. For ^{210}Po in cabbage, wheat, and bovine liver, the control sites were ranked the lowest, but with barley, Penrith and Warminster were ranked first and second, whilst Brecon Beacon and Newmarket were ranked sixth and eighth, out of the ten sites which were sampled. For ^{210}Pb the situation was not so clear cut. For barley Warminster, Newmarket, Brecon Beacon, and Penrith, were ranked first, fourth, fifth and, sixth respectively. Of the three sites sampled for bovine liver Newmarket and Penrith were ranked first and third, whilst, of the four sites sampled for cabbage, Brecon Beacon and Penrith were second and third respectively. With wheat, Newmarket was ranked third out of four sites.

It is difficult to explain this variation in the ranking of the sites by the above method but it may be useful in determining the enhancement or otherwise of ^{210}Pb and ^{210}Po in the particular foodstuffs at the sites sampled. This will be discussed further in Section 3.d.

Table 15 Ranking of Sites According to Increasing Maximum Specific Activity in Particular Food Types

a)^{210}Po

Barley	Cabbage	Wheat	Bovine Liver	Potato	Turnip	Apple	Blackberry
Penrith	Penrith	Newmarket	Newmarket	Romford	Avonmouth	Sellafield	Whitehaven
Warminster	Brecon Beacon	Drax	Penrith	Didcot	Cornwall	Drax	Holyhead
Drax	Cornwall	Sellafield	Cornwall	Sellafield	Whitehaven		
Didcot	Sellafield	Didcot					
Romford							
Brecon Beacon							
Holyhead							
Newmarket							
Whitehaven							
Sellafield							

Note: Increasing Maximum Specific Activity going down the column in both tables

b) ^{210}Pb

Barley	Cabbage	Wheat	Bovine Liver	Potato	Turnip	Apple	Blackberry
Warminster	Cornwall	Drax	Newmarket	Romford	Whitehaven	Sellafield	Whitehaven
Romford	Brecon Beacon	Didcot	Cornwall	Didcot	Avonmouth	Drax	Holyhead
Didcot	Penrith	Newmarket	Penrith	Sellafield	Cornwall		
Newmarket	Sellafield	Sellafield					
Brecon Beacon							
Penrith							
Drax							
Holyhead							
Sellafield							
Whitehaven							

3.c Comparison with Previous UK and World-wide Data

Over the years there has been considerable interest in the specific activities of ^{210}Pb and ^{210}Po in terrestrial and marine foodstuffs because of the dose received by humans through the ingestion pathway. Thus a considerable amount of data has been accumulated which is distributed about the scientific literature. A search of the literature was carried out and the data on terrestrial foodstuffs found are tabulated in appendix 1 along with the references.

To view comparable data with that found in this study a series of tables have been prepared below which list the ^{210}Pb and ^{210}Po specific activities according to food type. The data in each Table are sorted according to country of origin with that from this study first, followed by previous UK data, then world-wide data. If known, the source of the ^{210}Pb and ^{210}Po is also cited. The following mnemonics are used in the Source column of the tables:

ANUA: A Natural Uranium Area An area high in naturally occurring uranium isotopes (Zhu, 1990)	HNRA: High Natural Radiation Area An area with a number of radioactive anomalies due to different radionuclides.
CA: Control Area	UTA: Uranium Tailings Area
RPA: Reclaimed Phosphate Area	CPS: Coal Power Station

3.c.i Fruit Comparisons

Only two countries were found from the literature search with data on the specific activities in fruit, these were the UK and Germany. These data found are shown below in Table 16.

The only other UK data for fruit, was that determined by Smith-Briggs (1984), and this only for ^{210}Pb. Its value of 18 mBq kg^{-1} was lower than that found in any of the fruits in this study

The specific activities of ^{210}Po in apple from Germany are at the lower range of those found in this study, whereas the ^{210}Pb data of Globel are about a factor of five lower. Weisshar's data for ^{210}Pb on the other hand, extend over a larger range than that found in this study. The range of ^{210}Po:^{210}Pb activity ratios found by Globel overlap with that found here but with a higher upper boundary. For the other soft fruits, ^{210}Pb specific activities (Weisshar, 1993) were higher for blackberry and cranberry, and, lower for blueberry, redcurrent, and strawberry compared to that for blackberry in this study.

Table 16 ^{210}Pb and ^{210}Po data (mBq kg^{-1}, fresh weight) for fruit from this study, and other sources.

Location	Sample Type	^{210}Po	^{210}Pb	210Po:210Pb	Source	Reference
UK	Apple	26 - 92	91 - 119	0.25 - 1.01		This Study
UK	Blackberry	16 - 470	153 - 278	0.11 - 1.97		This Study
UK	Pear	61	152	0.4		This Study
UK	Fresh fruit		18			Smith-Briggs (1984)
Germany	Apple	19-34	21-30	0.90-1.41		Globel (1989)
Germany	Apple		20-140			Weisshar (1993)
Germany	Blackberry		490			Weisshar (1993)
Germany	Blueberry		120			Weisshar (1993)
Germany	Cranberry		340			Weisshar (1993)
Germany	Red currant		90			Weisshar (1993)
Germany	Strawberry		30-60			Weisshar (1993)

3.c.ii Cereals Comparisons

The data for wheat, barley, maize, rye, rye flour, rice, manioc, pulse, and, bread have been tabulated below in Table 17. The world-wide data for ^{210}Pb specific activities in barley are all within that found in this study, with the range of that from Germany (Weisshar 1993) being very similar. In comparing wheat with the other data, bread is also taken into account, thus, the results of the studies by Smith-Briggs (1984, 1986) are similar to that found here but, for ^{210}Pb the upper range in this study is about a factor of two higher whereas for ^{210}Po it is an order of magnitude higher. Hill's (1965) data for ^{210}Po in bread and cereals are similar to that found in this study.

For the rest of the world the ^{210}Pb specific activities overlap with those found here although the wheat flour ^{210}Po data of Globel (1989) has a higher upper range, which is about a factor of three higher than this study. It is tempting to attribute this difference in specific activities to the milling process, particularly since the lower boundary is elevated in relation to that found in this study, but, further evidence is required before this can be ascribed to a technological process.

Table 17 Data for Cereals (mBq kg^{-1}, fresh weight) from this study, and other Sources

Location	Sample Type	^{210}Po (mBq kg^{-1})	^{210}Pb (mBq kg^{-1})	^{210}Po:^{210}Pb	Source	Reference
UK	Barley	8 - 631	120 - 689	0.017 - 1.320		This Study
UK	Wheat	9 - 273	78 - 350	0.08 - 3.40		This Study
UK	Bread		116			Smith-Briggs (1984)
UK	Bread	27	61			Smith-Briggs (1986)
UK	Bread, Cereal	37-259				Hill (1965)
Argentina	Rice	110				Colangelo *et al.*, (1992)
Brazil	Manioc	205	153	1.34	CA	Santos *et al.,* (1993)
Brazil	Manioc	232-254	169-175	1.37-1.45	HNRA	Santos *et al.,* (1993)
China	Rice	470	570	0.82	HNRA	Zhu (1990)
China	Rice	2500	2400	1.04	ANUA	Zhu (1990)
Germany	Barley		120-680			Weisshar (1993)
Germany	Maize		60			Weisshar (1993)
Germany	Rye		120			Weisshar (1993)
Germany	Rye flour	296-481	222-555	0.80-1.33		Globel (1989)
Germany	Wheat		70-240			Weisshar (1993)
Germany	Wheat flour	192-740	241-666	0.80-1.18		Globel (1989)
India	Pulse		41-240			Lalit (1980)
India	Rice		30-488			Lalit (1980)
India	Wheat		55-481			Lalit (1980)
USSR	Barley	189	244			Ladinskaya (1973)
USSR	Wheat	104	178			Ladinskaya (1973)
	Cereals		74 - 185			UNSCEAR (1977)
	Grain	37 - 370				Parfenov (1974)

The cereal with the highest specific activity is rice from a natural uranium area in China (Zhu, 1990) with values in the Bq kg^{-1} range. The other cereal products had specific activities comparable to those in this study.

In comparing ratios, wheat flour from Germany (Globel, 1989) is within the range of that found here and is much narrower in magnitude, 0.38 compared to 3.32. Most of the other ratios were close to unity except for Manioc from Brazil whose ratios from areas of high natural radioactivity and control areas were consistently above 1.

3.c.iii Milk Comparisons

Although only one milk was assayed in this study it is used in comparison with milk from other countries even though the conclusions may not be definitive. The specific activity found for milk in this study is at the low end of the range for both UK and world-wide data (see Table 18). In particular the ^{210}Pb value was the lowest of all of the data found in the literature search.

In comparison to other countries, milk from cows at a reclaimed phosphate area in the USA showed the highest specific activities. The cows from this area were fed on forage which was grown on land reclaimed from a phosphate mining area which suggests that the milk may be undergoing enhancement in its ^{210}Pb and ^{210}Po specific activities in comparison to other world-wide data.

Table 18 ^{210}Pb and ^{210}Po Data for Milk (mBq l^{-1}) from this study, and other Sources

Location	Sample Type	^{210}Po (mBq l^{-1})	^{210}Pb (mBq l^{-1})	^{210}Po:^{210}Pb	Source	Reference
UK	Milk	10	3	3.1		This Study
UK	Dried Milk	7.4-22.2				Hill (1965)
UK	Milk		35			Smith-Briggs (1984)
UK	Milk		22			Smith-Briggs (1986)
Brazil	Milk		5.0-60		HNRA	Amaral et al., (1988)
Germany	Milk	5.1-35	5.2-32	0.90-1.34		Globel (1989)
Germany	Milk	7 - 67	22 - 167			Schüttelkopf (1981)
Germany	Milk		20-50			Weisshar (1993)
USA	Milk	22.5-106	74-246		RPA	Staples *et al.*, (1994)
USSR	Milk	8.5	20			Ladinskaya *et al.*, (1971)

The activity ratio of ^{210}Po:^{210}Pb in the milk sample in this study is much larger than the only other ratio from the literature search, but it should be remembered that these specific activities are very small. The associated error for ^{210}Pb is 30% whilst that of ^{210}Po is 10% for that sample.

3.c.iv Root Crops Comparisons

As can be seen in Table 19, there is a striking difference in the specific activities in UK root crops in comparison to the rest of the world. The values in this and previous UK studies are, in some cases, an order of magnitude lower than those of comparable world-wide data.

A large proportion of the world-wide data has come from areas of high natural radioactivity or natural uranium areas. The root crops from these areas have high levels of ^{210}Pb and ^{210}Po compared to background areas which is good evidence of the enhancement of these nuclides. Interestingly most of these root crops have ^{210}Po:^{210}Pb activity ratios much smaller than unity which may reflect differences in uptake of these isotopes by the crops.

Table 19 ^{210}Pb and ^{210}Po Data for Root Crops (mBq kg^{-1}, fresh weight) from this study, and other sources

Location	Sample Type	^{210}Po (mBq kg^{-1})	^{210}Pb (mBq kg^{-1})	^{210}Po:^{210}Pb	Source	Reference
UK	Potato	1 - 13	5 - 10	0.2 - 1.3		This Study
UK	Turnip	2 - 6	6 - 22	0.16 - 1.0		This Study
UK	Potato		16			Smith-Briggs (1984)
UK	Carrot, Potato	37				Hill (1965)
Argentina	Potato	16				Colangelo *et al.*, (1992)
Brazil	Beetroot	133	178	0.75		Santos *et al.*, (1993)
Brazil	Beetroot	333-587	474-503	0.70-1.08	HNRA	Santos *et al.*, (1993)
Brazil	Carrot	267	278	0.95		Santos *et al.*, (1993)
Brazil	Carrot	333-445	280-400	1.11-1.19	HNRA	Santos *et al.*, (1993)
Brazil	Radish	288	562	0.58		Santos *et al.*, (1993)
Brazil	Radish	289-863	824-1418	0.35-0.6	HNRA	Santos *et al.*, (1993)
Brazil	Turnip	122	222	0.55		Santos *et al.*, (1993)
Brazil	Turnip	278-1130	470-1850	0.59-0.61	HNRA	Santos *et al.*, (1993)
China	Radish	100	240	0.42	ANUA	Zhu (1990)
China	Sweet potato	150	280	0.53	HNRA	Zhu (1990)
China	Sweet potato	510	540	0.94	ANUA	Zhu (1990)
Germany	Beetroot		100			Weisshar (1993)
Germany	Carrot	26-44	22-56	0.85-1.27		Globel (1989)
Germany	Carrot		40			Weisshar (1993)
Germany	Kohl rabi		30			Weisshar (1993)
Germany	Potato		30-60			Weisshar (1993)
Germany	Radish		20			Weisshar (1993)
Japan	Potato		100			Kanetani *et al.*,(1981)
	Potato	33 - 189	80 - 244			Bunzel *et al.*,(1979)
	Root Vegetables	37 - 111				Parfenov (1974)
	Root Vegetables		7.8			Morse and Welford (1971)

3.c.v Offal Comparisons

As can be seen in Table 20, the offal samples in this study, have the highest specific activities for both ^{210}Pb and ^{210}Po of all of the foodstuffs sampled. In comparison to previous UK studies these levels are in reasonable agreement with those published by Smith-Briggs (1984, 1986) but in the lower part of the range of that by Hill (1965).

Table 20 ^{210}Pb and ^{210}Po Data for Offal (mBq kg^{-1}, fresh weight) from this study, and other Sources

Location	Sample Type	^{210}Po (mBq kg^{-1})	^{210}Pb (mBq kg^{-1})	^{210}Po:^{210}Pb	Source	Reference
UK	Bovine Liver	794 - 2956	186 - 548	1.58 - 5.2		This Study
UK	Ovine Liver	586 - 740	990 - 1600	0.46 - 0.59		This Study
UK	Beef, Lamb kidney	1776-9990		0.05-1		Hill (1965)
UK	Beef, Lamb liver	148-3700		0.7		Hill (1965)
UK	Cattle liver	1900±400	700±400	2.7±1.6	CPS	Smith-Briggs (1984)
UK	Cattle liver	3600±600	500±300	7.2±4.5	CA	Smith-Briggs (1984)
UK	Lamb kidney	3330-66600		0.2		Hill (1965)
UK	Offal		651			Smith-Briggs (1984)
Canada	Caribou kidney	259000 ±18000	84000±8000	3.64±1.59	NBA	Thomas (1994)
Canada	Caribou liver	374000 ±25000	158000 ±10000	2.18±1.57	NBA	Thomas (1994)
Finland	Reindeer liver	37740-173530	10360-55870	3.1-5.0	CA	Kauranen et al. (1969)
Germany	Cattle liver		320-800		NS	Weisshar (1993)
Germany	Pig liver		200-280		NS	Weisshar (1993)
Germany	Pigs liver	3330-4540	3150-4370	0.95-1.08	CA	Globel (1989)
Israel	Chicken liver	210-1030			NS	Izak-Biran *et al.*,(1989)
USA	Cattle liver		592		RPA	Stricker *et al.*, (1994)
USA (New Mexico)	Cattle kidney	17000±5000	3000±600	5.7±2.0	CA	Lapham *et al.*, (1989)
USA (New Mexico)	Cattle kidney	31000-65000	2900-13000	2.38-22.4	UTA	Lapham *et al.*,(1989)
USA (New Mexico)	Cattle liver	12000-56000	380-3400	16.5-31.6	UTA	Lapham *et al.*,(1989)
USA (New Mexico)	Cattle liver	9000±2000	250±140	36±22	CA	Lapham *et al.*,(1989)
	Kidney	6605	1628			Bunzel *et al.*,(1979)
	Liver	1230	666			Bunzel *et al.*,(1979)
	Liver, kidney	185 - 37000				Parfenov (1974)

In Table 20, it can be seen that the world-wide data for the specific activities of ^{210}Pb and ^{210}Po in offal are much higher than that of the UK. Cattle offal ^{210}Po specific activities are an order of magnitude higher than those of the UK with ^{210}Pb being about a factor of 6. Within these animals there is a clear distinction between those which have high and low specific activities which seems to be reflected in the areas where they were reared. Control area animals have lower specific activities than uranium tailings area animals.

The highest specific activities for offal were in Caribou and Reindeer. These animals consume large amounts of lichen, approximately 3 - 4 kg day^{-1}, which can contain high specific activities of ^{210}Pb and ^{210}Po. Lichen, which is known to concentrate radionuclides as well as stable elements, is the main food source for these animals and this explains the high levels found. Offal also shows the largest range of ^{210}Po:^{210}Pb activity ratios compared to the other foodstuffs which is well reflected in Table 20 above. Ratios are in general very much larger than one, indicating preferred uptake of ^{210}Po.

3.c.vi Green Vegetable Comparisons

Table 21 below lists the results of the literature survey for this food type. Large variations in specific activity can be seen, with the values for this study, and previous studies by Smith-Briggs (1984, 1986), being in the lower part of the range of world-wide data. The ^{210}Po data from Hill (1965) are amongst the highest found. It would be tempting to attribute this high value to enhancement and, indeed it may be so, but unfortunately Hill does not give a sampling site for this foodstuff.

Elevated levels are found in areas of high natural radioactivity with the activity ratios of ^{210}Po:^{210}Pb in general being less than one. This suggests that cabbage takes up ^{210}Pb more efficiently than ^{210}Po.

In this study, ratios in cabbage tended to be very much lower than the world-wide data. This is difficult to explain, but it could be that the specific activities may be a function of the age of the plant and the rate of uptake such that at the time of sampling, the plants had not reached their equilibrium specific activities.

Table 21 ^{210}Pb and ^{210}Po Data for Green vegetables (mBq kg^{-1}, fresh weight) from this study, and other Sources

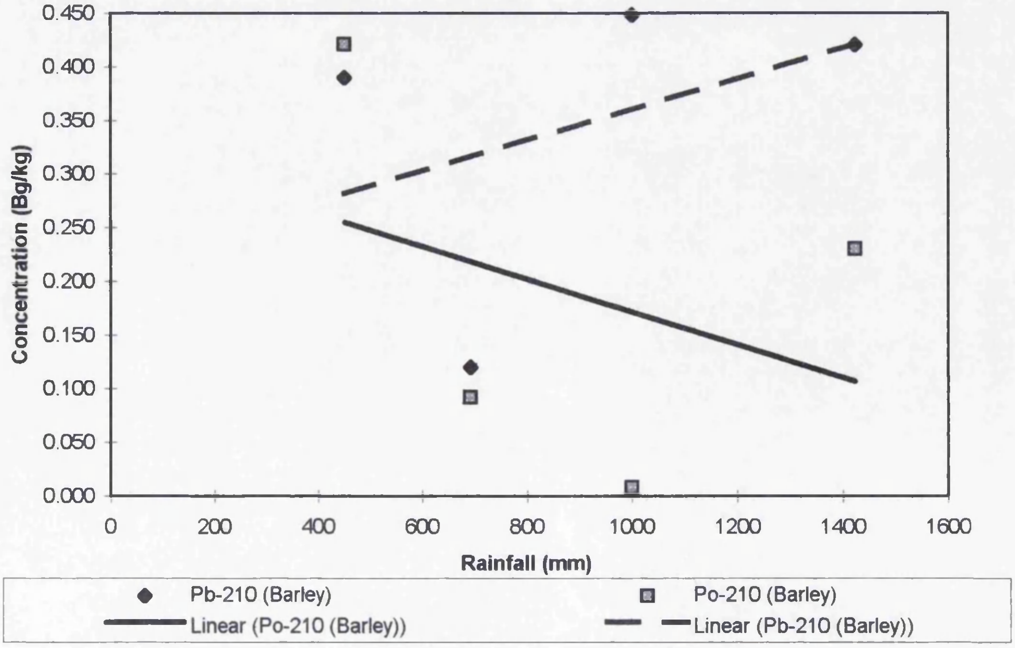
Location	Sample Type	^{210}Po (mBq kg^{-1})	^{210}Pb (mBq kg^{-1})	^{210}Po:^{210}Pb	Source	Reference
UK	Brocoli	27	66	0.4		This Study
UK	Brussels Sprouts	34	100	0.34		This Study
UK	Cabbage	3 - 148	4 - 267	0.2 - 0.89		This Study
UK	Lettuce	136	138	0.98		This Study
UK	Green vegetables	222-3330		1-3		Hill (1965)
UK	Green vegetables		31			Smith-Briggs (1984)
UK	Vegetables	40	49			Smith-Briggs (1986)
Argentina	Cauliflower	22				Colangelo *et al.*, (1992)
Argentina	Spinach	320				Colangelo *et al.*, (1992)
Brazil	Broccoli	447	467	0.96	CA	Santos *et al.*, (1993)
Brazil	Broccoli	457-761	539-774	0.89-0.98	HNRA	Santos *et al.*, (1993)
Brazil	Cabbage	227	518	0.75	CA	Santos *et al.*, (1993)
Brazil	Cabbage	398-578	557-659	0.71-0.88	HNRA	Santos *et al.*, (1993)
Brazil	Cauliflower	167	278	0.6	CA	Santos *et al.*, (1993)
Brazil	Cauliflower	279-375	538-894	0.39-0.48	HNRA	Santos *et al.*, (1993)
Brazil	Chicory	944	1144	0.76	CA	Santos *et al.*, (1993)
Brazil	Chicory	522-1320	988-1790	0.52-0.74	HNRA	Santos *et al.*, (1993)
Brazil	Forage (grass)		1040-9220		HNRA	Amaral *et al.*, (1988)
Brazil	Kale	467	667	0.7	CA	Santos *et al.*, (1993)
Brazil	Kale	455-560	450-700	0.69-.82	HNRA	Santos *et al.*, (1993)
Brazil	Lettuce	722	783	0.92	CA	Santos *et al.*, (1993)
Brazil	Lettuce	882-1453	954-1978	0.73-0.92	HNRA	Santos *et al.*, (1993)
Brazil	Swiss chard	462	0.54	0.85	CA	Santos *et al.*, (1993)
Brazil	Swiss chard	474-1357	539-1117	0.88-1.21	HNRA	Santos *et al.*, (1993)
Germany	Broccoli		190-220			Weisshar (1993)
Germany	Brussels sprouts		100-170			Weisshar (1993)
Germany	Cabbage	4.1-7.9	3.8-8.3	0.95-1.10		Globel (1989)
Germany	Cabbage (red/white)		30-890			Weisshar (1993)
Germany	Cabbage (savoy)		240			Weisshar (1993)
Germany	Cauliflower		20-40			Weisshar (1993)
Germany	Cucumber		70			Weisshar (1993)
Germany	Kale		90-1600			Weisshar (1993)
Germany	Leek		40-410			Weisshar (1993)
Germany	Lettuce	8-20	10-18	0.71-1.17		Globel (1989)
Germany	Lettuce		200-880			Weisshar (1993)
Germany	Parsley		630			Weisshar (1993)
Germany	Spinach	6.5-9.9	7.4-9.1	0.79-1.11		Globel (1989)
Germany	Spinach		260-930			Weisshar (1993)
Germany	Vegetables+salad	333 - 1628	555 - 31820			Schüttelkopf (1981)

3.d A Potential for Technological Enhancement

One of the primary objectives of this thesis was to determine whether any of the sites have undergone technological enhancement of the specific activities of ^{210}Pb and ^{210}Po in the foodstuffs sampled. It was therefore important to compare the specific activities found at sites which may have the potential of being enhanced, with control sites distant from sources of ^{210}Pb and ^{210}Po. Care was taken in choosing control sites according to this criterion and they were also chosen on the basis of their annual rainfall. Thus pairs of sites were chosen with high rainfall and low rainfall respectively which were also at some distance from each other where possible. Rainfall data for 1991 are tabulated in Table 7 and the site locations are depicted in figure 3.

In the preceding discussion on ranking of the sites it was noted that the control sites were not necessarily ranked low in all food types. A possible explanation for this could be the effect of rainfall causing washout of ^{210}Pb and ^{210}Po from the atmosphere. To test this hypothesis the average barley specific activities for the control sites were plotted against annual rainfall data for 1991. This is shown below in figure 10.

Figure 10 Variation of Rainfall with Average Specific Activity in Barley at the Control Sites



The figure shows that although both radionuclides have opposite trends, the correlation coefficients are low (0.16 for ^{210}Pb and, 0.12 for ^{210}Po) indicating no significant effect of rainfall in 1991 on the average specific activities at the control sites.

Another criterion for assessing enhancement or not is by assuming that the specific activities are normally distributed, and taking any specific activity above an upper percentile as being enhanced. This has been done using the complete data set of foodstuffs, which has been collected at more than one site, for the 97.5th percentile specific activities, and also for the control set itself. Similarly, one could define a minimum level of enhancement which is equal to the mean plus three times the standard deviation of the specific activities of particular foodstuffs at the control sites. The results are shown below in tables 22, 24 and 26.

Comparing the calculated percentiles with the specific activities of the foodstuffs at the individual sites, we can see which sites are enhanced for which foodstuff. This is shown below in Tables 23 and 25.

Table 22 97.5th Percentiles of the ^{210}Pb and ^{210}Po Specific Activities of Bovine Liver, Barley, and Cabbage from the control sites

	^{210}Po (Bq kg^{-1})	**^{210}Pb (Bq kg^{-1})**
	97.5th Percentile	**97.5th Percentile**
Bovine Liver	0.864	0.539
Barley	0.402	0.445
Cabbage	0.062	0.222

On the basis of the results of the analysis based on the levels of the percentiles at the control sites, shown in Table 22 above, we can see that for ^{210}Po, there is one site of possible natural enhancement (Cornwall), one site of possible technological enhancement (Whitehaven), and one nuclear site (Sellafield) above the percentile criteria. Similarly for ^{210}Pb, there is one site of possible enhancement by coal power electricity generation (Drax), one site of possible enhancement by metal smelting (Holyhead), one site of possible enhancement by phosphate ore processing (Whitehaven), and, one nuclear site (Sellafield).

Table 23 Sites with ^{210}Pb and ^{210}Po Specific Activities above the 97.5th Percentiles of the Control Sites

	^{210}Po	^{210}Pb
	97.5th Percentile	**97.5th Percentile**
Bovine Liver	Cornwall	-
Barley	Sellafield	Drax
Barley	Whitehaven	Holyhead
Barley	-	Sellafield
Barley	-	Whitehaven
Cabbage	Cornwall	Sellafield
Cabbage	Sellafield	-

On the criterion of the percentiles based on all of the sites from Table 24 it can be seen that for ^{210}Pb, Sellafield shows enhancement for four, Whitehaven for one, Holyhead for one, and, Penrith for one food type. Whereas for ^{210}Po, Sellafield shows enhancement for three, Holyhead for one, Cornwall for one, Whitehaven for one, and, Didcot for one food type.

Table 24 97.5th Percentiles of the ^{210}Pb and ^{210}Po Specific activities (Bq kg^{-1}, fresh weight) of the Food Types from all sites

	^{210}Pb	^{210}Po
Sample type	**97.5th Percentile**	**97.5th Percentile**
Apple	0.119	0.090
Barley	0.687	0.585
Blackberry	0.275	0.459
Bovine Liver	0.546	2.852
Cabbage	0.264	0.142
Potato	0.010	0.061
Turnip	0.021	0.006
Wheat	0.332	0.261

Using the mean plus three times the standard deviation of the specific activities of the foodstuffs from the control sites criterion, a slightly different picture emerges. As can be seen from Table 26 below, based on the specific activities calculated using the three criteria, this criterion is a more rigorous test for possible enhancement as compared to the percentile criteria for control sites, but is less rigorous than the food type percentiles for ^{210}Po in bovine liver and cabbage and, for ^{210}Pb in cabbage.

Table 25 Sites with ^{210}Pb and ^{210}Po Specific Activities above the 97.5th Percentiles by Sample Type

	^{210}Pb	^{210}Po
Sample Type	**97.5th Percentile**	**97.5th Percentile**
Apple	Sellafield	Sellafield
Barley	Whitehaven	Sellafield
Blackberry	Holyhead	Holyhead
Bovine Liver	Penrith	Cornwall
Cabbage	Sellafield	Sellafield
Potato	Sellafield	Sellafield
Turnip	Cornwall	Whitehaven
Wheat	Sellafield	Didcot

On the basis of this criterion, Sellafield, Whitehaven, and, Cornwall show enhancement of ^{210}Pb and ^{210}Po in some sample types (see Table 27 below).

Table 26 Comparison of the Calculated Specific Activities of ^{210}Pb and ^{210}Po Using the Three Criteria for Technological Enhancement

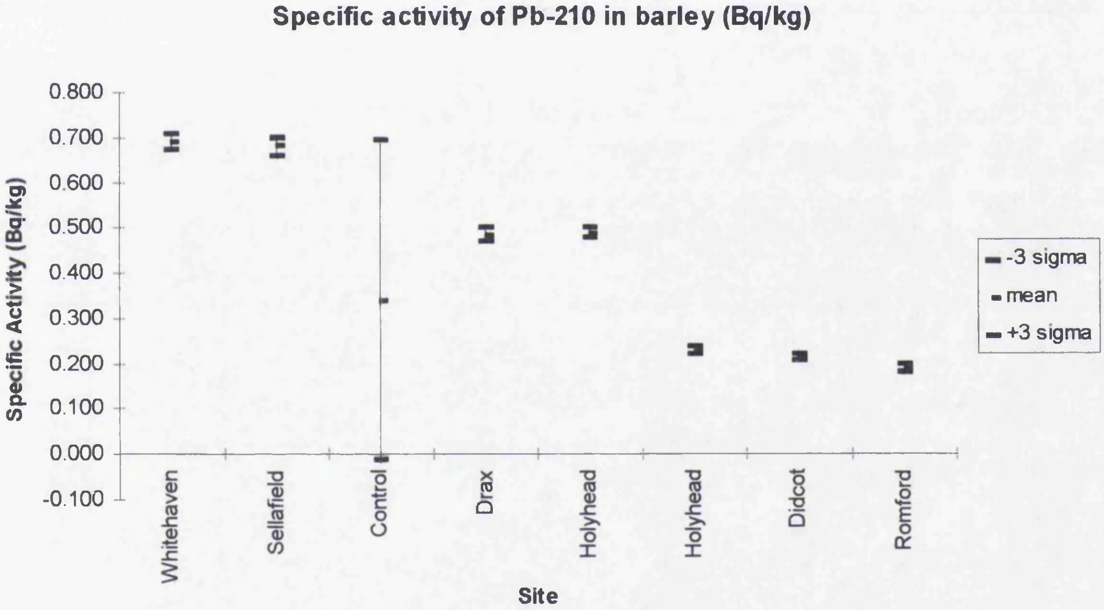
	^{210}Po Bq kg^{-1}			**^{210}Pb Bq kg^{-1}**		
Site Criterion	**97.5th Percentile All**	**97.5th Percentile Control**	**Mean + 3σ Control**	**97.5th Percentile All**	**97.5th Percentile Control**	**Mean + 3σ Control**
Barley	0.585	0.402	0.601	0.687	0.445	0.693
Cabbage	0.142	0.062	0.080	0.264	0.222	0.255
Bovine Liver	2.852	0.864	0.938	0.546	0.539	0.910

Table 27 Sites with ^{210}Pb and ^{210}Po Specific Activities above the Mean plus Three Standard Deviation Specific activity Criteria for Control Sites

Sample Type	^{210}Po	^{210}Pb
Barley	Sellafield	Whitehaven
Bovine Liver	Cornwall	-
Cabbage	Sellafield	Sellafield

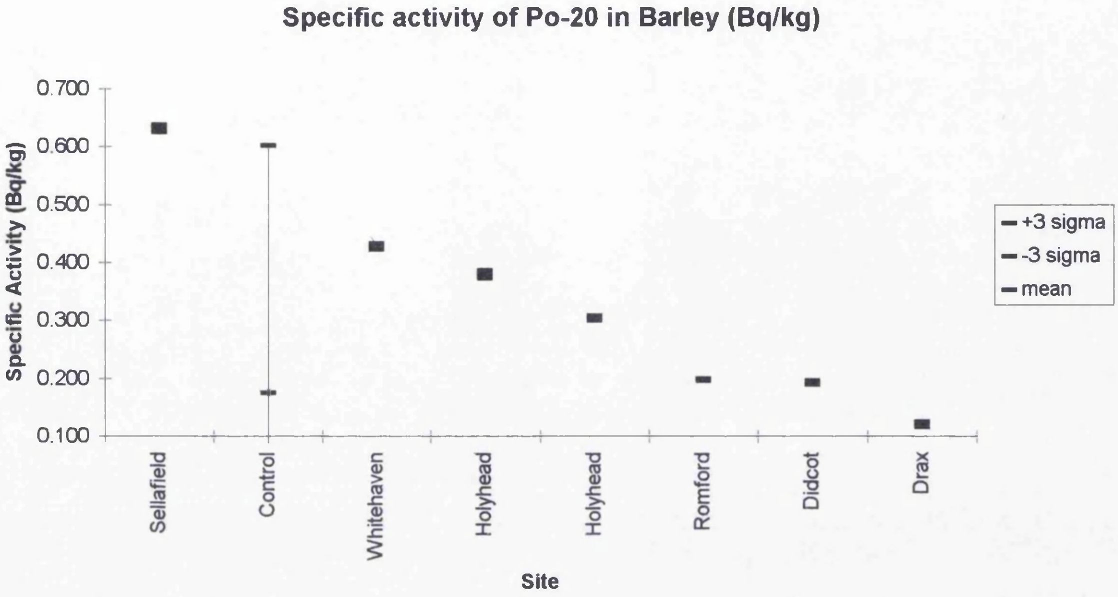
The above attempts at using a statistical basis to identify enhancement have had some degree of success in that the criteria have more or less consistently identified the same sites for possible enhancement. Both Sellafield and Cornwall were identified by all criteria as being enhanced in ^{210}Po, and, Whitehaven and Sellafield as being enhanced in ^{210}Pb. Using the percentile criterion for all of the sample types, further sites are identified as being enhanced although it should be remembered that these sites were identified because they have particular sample types which lie close to the upper limit of the distribution for that type. This is presented graphically in figures 11 and 12. Enhanced sites are to the left of the control sites in these figures.

Figure 11 Comparison of mean plus three standard deviation criteria with sample data for ^{210}Po at enhanced sites



It is easy to understand the cause of the enhancement at Cornwall as being of natural origin, since this is an area known to have high levels of natural radionuclides as discussed in Chapter 2. The causes of enhancement at Sellafield and Whitehaven are more difficult to explain. In the past both areas had been subjected to industrial activities which are known to have discharged large amounts of ^{210}Po into the environment (see Chapter 2).

Figure 12 Comparison of mean plus three standard deviation criteria with sample data for ^{210}Pb at enhanced sites



At Sellafield, the fire in the Windscale piles, in 1957, caused a large amount of ^{210}Po to be released into the environment. Almost a hundred half-lives have elapsed since the fire to the time of sampling so it can safely be said that this is not the cause of the enhancement. This leaves two other possibilities, either, Sellafield is routinely discharging ^{210}Po into the atmosphere, or, there is a natural source. To decide the cause it would require a survey of the atmospheric and soil activities of ^{210}Pb and ^{210}Po at the sites sampled, since there are no data available.

At Whitehaven there is the possibility that spillage of phosphatic ores that were supplied to the Marchon plant could be the cause of the enhancement, but again a soil survey of the activities of ^{210}Pb and ^{210}Po and phosphorous concentrations would be required to confirm this.

It should be noted that in comparison to world-wide data, the UK enhanced sites would be classed as relatively minor in magnitude of enhancement, or not at all.

Chapter 4. Radiological Assessment of ^{210}Pb and ^{210}Po in Terrestrial Foodstuffs.

4.a The Critical Group and Food Consumption Data.

There are four routes through which the human population can receive a radiation dose from radioactivity:

a) External irradiation from a remote source.
b) Absorption of radionuclides through the skin.
c) Inhalation of radioactivity in the atmosphere.
d) Ingestion of radionuclides from foodstuffs.

Absorption of radionuclides is an important route of occupational exposure, as is exposure by external radiation, particularly, for workers in the nuclear industry. For non-occupational exposure the inhalation and ingestion routes are the main routes of exposure.

In the UK, monitoring of foodstuffs for radionuclides has been ongoing since the 1950's, when it was first undertaken by the Atomic Energy Authority at the Windscale site (renamed Sellafield) in Cumbria. Interest has expanded to include contamination of foodstuffs due to atomic weapons fallout, site specific and accident related effects on the food chain and, natural radioactivity in food (MAFF, 1994). Monitoring programmes are run by plant operators, government departments and agencies as well as local authorities. Also, information on radionuclide concentrations comes from research projects carried out by universities and other bodies, of which this thesis is one.

In a review of the radiation exposure of the UK population, (Hughes, and O'Roirdan, 1993) it was stated that the average annual dose to any member of the UK population was about 2,600 µSv, of which, 2,210 µSv was from exposure to natural sources. More than 50 %, 1,300 µSv, of this dose was due to exposure to radon and its progeny, with the next highest being 350 µSv via external irradiation from terrestrial sources. Internal irradiation, mainly through consumption of foodstuffs, accounted for 300 µSv, and cosmic radiation 260 µSv. Table 28 summarises the annual dose to the UK public from all sources of radiation.

Table 28 Annual exposure of the UK population from all sources of radiation

Source	Annual Collective Dose (man Sv)	Average Annual Dose (µSv)
Natural		
Cosmic	15,000	260
Gamma	20,200	350
Internal	17,300	300
Radon	74,900	1,300
Artificial		
Medical	21,400	370
Occupational[a]	430	7
Fallout	290	5
Discharges[b]	20	0.4
Products	20	0.4
Total (rounded)	150,000	2,600

[a]Some 80% from Natural Sources [b]Some 20% from Natural Sources

In any assessment of the dose equivalent to man from radioactivity, the objective is to estimate the highest dose that could possibly be received by any member of the public. Thus, it is important to identify the individual, or individuals, who are likely to receive the highest exposure from a given practice. These individuals are defined as the critical group, and they will vary according to the particular practice involved. The ICRP (ICRP, 1984) has defined the critical group as:

> 'The [critical] group should be representative of those individuals in the population expected to receive the highest dose equivalent; the group should be small enough to be relatively homogeneous with respect to age, diet and those aspects of behaviour that effect the doses received. The Commission believes that it will be reasonable to apply the mean dose equivalent for individual members of the public to the mean dose equivalent in the critical group.'

ICRP also recommends that:

> 'the critical group would not consist of one individual nor would it be very large for then homogeneity would be lost. The size of the critical group will usually be a few tens of persons.'

In dose assessment studies of radioactivity from the ingestion of foodstuffs, there are two methods used for the selection of critical groups (NRPB, 1993) these are:

a) Site specific methods. Where a local habit survey of the consumption of foodstuffs produced in the proximity of a source of radioactivity is carried out. These data are then used in dose calculation for the ingestion pathway.

b) Generalised methods. Whereby habit data based on national statistics on food consumption are used in the calculation of the dose via the ingestion pathway.

In this study the Generalised method is used.

In any assessment of exposure to radiation through the ingestion of foodstuffs, it is important to have accurate information on the food consumption habits of the population under study. In the UK many data have been accumulated on the food consumption habits of the different age groups of the population. Food consumption surveys have been performed on infants (Mills and Tyler, 1992), schoolchildren (Department of Health, 1989), and adults (Gregory *et al.,* 1990). The survey by Gregory was done under the auspices of the Office of Population Censuses and Surveys which brought together data on extreme consumption rates, and social and regional differences of consumption. The report contains the initial findings of the above study which was carried out between October 1986 and August 1987. A representative sample of adults in the age group 16 to 64 living in private households was recruited, with pregnant women being excluded. Participants were asked to record their diet over a seven day period, and a variety of anthropometric and blood pressure measurements were made, along with a 24 hour urine collection and a specimen of venous blood.

Byrom, *et al.,* (1995) have brought together the above studies to produce an agreed standard set of data which could be used by UK national regulatory bodies, nuclear operators, local authorities, and other interested parties for the calculation of radiation dose from the ingestion pathway.

In Byrom *et al.,* (1995), twenty-six food groups were chosen to be representative of a whole diet of the UK population. Food consumption data were tabulated for each of four age groups; infants aged 6 to 12 months; schoolchildren aged 10 to 11 years; schoolchildren aged 14 to 15 years; and adults aged 16 to 64 years. Depending on the type of assessments required different measures of consumption were calculated for each of the age groups. These were; the average (mean per capita) consumption over the whole population, the mean and median for the consumers, and, the 95th and 97.5th percentile for the consumers.

In defining the critical group, an upper percentile of the habits of the population under study is usually chosen. In the past this has been either the 95th or 97.5th percentile.

In assessing the dose from food consumption, it is considered appropriate to sum the dose from the two food groups which provide the highest doses, calculated using the 97.5th percentile consumption rate, plus the remainder of the diet using food average consumption rates (Byrom, *et al.,* 1995). In this study the 97.5th consumption rate is used in the calculation for the critical group.

To find the two foodstuffs which give the largest CED based on 97.5th consumption rates, the calculation is performed for each foodstuff, for each of the age groups at each site. The two largest are then selected for each age group for each site and the total dose for each age group at each site is then found by summing the 97.5th percentile consumption rate CED with the CED received from the remaining foodstuffs based on average consumption rates.

The two foodstuffs which contribute the largest dose based on 97.5th consumption rates for the critical groups used in the dose assessment at each of the sites studied are tabulated below.

Table 29 97.5th Percentile foodstuffs used in critical group calculations

Site	Pb-210			Po-210		
Brecon Beacon	All age groups	Barley	Cabbage	All age groups	Barley	Cabbage
Holyhead	6 -12 months	Barley	Blackberry	All age groups	Barley	Blackberry
	All other ages	Barley	Ovine Liver			
Newmarket	All age groups	Barley	Wheat	All age groups	Barley	Bovine Liver
Cornwall	All age groups	Bovine Liver	Cabbage	All age groups	Bovine Liver	Cabbage
Romford	All age groups	Barley	Potato	All age groups	Barley	Potato
Penrith	All age groups	Barley	Bovine Liver	All age groups	Bovine Liver	Cabbage
Didcot	All age groups	Barley	Wheat	All age groups	Barley	Wheat
Sellafield	All age groups	Barley	Wheat	All age groups	Barley	Wheat
Warminster	All age groups	Barley		All age groups	Barley	
Drax	All age groups	Barley	Pear	All age groups	Barley	Pear
Whitehaven	All age groups	Barley	Blackberry	All age groups	Barley	Blackberry

4.b Dose Calculations and Annual Intakes of ^{210}Pb and ^{210}Po

To calculate the annual committed effective dose (CED) we require information on the concentration of the radionuclides in the foodstuff of interest, the dose coefficient of the radionuclides, and the annual consumption rate of the foodstuff.

In mathematical terms this is defined as,

$$H_r(\text{Sv yr}^{-1}) = C_{r,f}(\text{Bqkg}^{-1}) \times D_r(\text{SvBq}^{-1}) \times R_f(\text{kg yr}^{-1})$$

where H_r = annual commited effective dose equivalent for radionuclide *r*,

$C_{r,f}$ = concentration of radionuclide *r* in foodstuff *f*,

D_r = dose per unit intake for radionuclide *r*,

R_f = annual consumption rate for foodstuff *f*.

Recommended values of the dose coefficients for ^{210}Pb and ^{210}Po for each of the different age groups, were taken from three sources. These values were based on biokinetic models which have been progressively improved over the years and extended to different age groups (ICRP,1979,1989,1990,1993). Of particular importance in these biokinetic models is the gut absorption factor or the f_1 value.

For ^{210}Po, the f_1 value has attracted considerable attention, because of the dose received from the ingestion of ^{210}Po in foodstuffs. ICRP had initially recommended a value of 0.1 (ICRP, 1979) but this was based on few data. The

Nuclear Energy Agency (NEA) in 1988 reviewed the available data at that time, for a large number of radionuclides (NEA,1988) and made specific recommendations as to f_1 values following intake by ingestion. They recommended an f_1 value of 0.3 for ^{210}Po. In a report by the NRPB (1990) recent data on rats were considered and NRPB advised the retention of the ICRP value of 0.1 for adults, and 0.2 for children. More recently, Hunt and Allington (1993) in a series of experiments in which crab meat of a known ^{210}Po concentration was fed to a number of adult volunteers, found a value for the f_1 factor of 0.8. Also, in a study of the transfer of ^{210}Po from caribou to wolves (Thomas, 1994b), data were obtained that indicated an f_1 value of about 0.4 although strictly this may not apply to humans. The most recent recommendation from ICRP (1993) for the f_1 value is 1.0 for children up to 1 year and 0.5 for adults and children over 1 year.

It can be seen from above that considerable variation has been found in the f_1 value for ^{210}Po. This could in part be due to differences in the speciation of ^{210}Po administered to the animals, and humans, in the laboratory experiments performed, compared to that which is ingested in foodstuffs. Recent experiments (Bulman, et al., 1995) on the extractability of ^{210}Po using a number of sulphur containing ligands and solvent systems, from lamb's liver, pig's kidneys, mussel flesh and, brown crab meat, has shown that ^{210}Po is present as different forms in these materials. This important result shows that the f_1 value found by Hunt and Allington (1993) by eating crab meat is not applicable to the consumption of all foodstuffs and therefore requires further study.

To be consistent with current radiological practice, the most recent ingestion dose coefficient values and corresponding f_1 value, are used in this study. These have been taken from three sources, IAEA (1996), ICRP, and the European Union (1996) and are tabulated in Table 30. The dose coefficient for ^{210}Pb also includes a contribution from the decay of its daughter radionuclides which would be produced by its decay within the body. In this study, the values published by the European Union (1996) and the International Atomic Energy Agency (1996) were used since these are the most recent compilations to date. Both of these compilations provide dose coefficients for the less than or equal to 1 year, and 1 - 2 years age groups. The consumption data of Byrom provide data for the 6 - 12 month age group so a decision must be made as to which dose coefficient to use since this can make a large difference in the dose calculation. This is discussed later.

Table 30 Committed Effective Dose Per Unit Intake e(g) via ingestion (Sv/Bq)

Age<=1		**f_1 for**	**Age 1-2**	**Age 2-7**	**Age 7-12**	**Age 12-17**	**Age >17**
^{210}Pb:Dose per unit intake for the age groups above							
f_1	**^{210}Pb**	**Upper row IAEA and European Union, lower row ICRP. f_1 for 1-15 age 0.4**					
0.6	$8.4\text{X}10^{-6}$	0.2	$3.6\text{X}10^{-6}$	$2.2\text{X}10^{-6}$	$1.9\text{X}10^{-6}$	$1.9\text{X}10^{-6}$	$6.9\text{X}10^{-7}$
0.6		0.2	$3.6\text{X}10^{-6}$	$2.2\text{X}10^{-6}$	$1.9\text{X}10^{-6}$	$1.9\text{X}10^{-6}$	$7.07\text{X}10^{-7}$

^{210}Po:Dose per unit intake for the age groups above							
f_1	**^{210}Po**	**Upper row IAEA and European Union, lower row ICRP.**					
1.0	$2.6\text{X}10^{-5}$	0.5	$8.8\text{X}10^{-6}$	$4.4\text{X}10^{-6}$	$2.6\text{X}10^{-6}$	$1.6\text{X}10^{-6}$	$1.2\text{X}10^{-6}$
1.0	$2.1\text{X}10^{-5}$	0.5	$8.8\text{X}10^{-6}$	$4.4\text{X}10^{-6}$	$2.6\text{X}10^{-6}$	$1.6\text{X}10^{-6}$	$1.2\text{X}10^{-6}$

The annual effective committed dose equivalent has been calculated for each of the age groups using the dose coefficients above and the consumption rates of Byron et al.(1996) and the maximum specific activities found of the foodstuffs (see Chapter 3). These calculated doses are tabulated below. The proportion of the mean and critical group doses at each site are also tabulated (see table 32).

It must be stressed that the doses calculated are those received from the consumption of the foodstuffs collected from each particular site and as such each site is not strictly comparable with all others because not all foodstuffs were available at each site.

However, if the contributions to the dose from those foodstuffs which make up the 97.5th percentile consumption rates are compared to those which were calculated using average consumption rates within each critical group, we can see that the 97.5th percentile consumption doses are about 90% of the overall dose, with the minimum being 89% for the 10 - 11 age group at Drax. Moreover since the vast majority of the critical groups have barley as one of the 97.5th percentile consumption foods, it therefor seems reasonable to make comparisons based on the two foodstuffs which are calculated using this consumption rate.

Barley has one of the largest 97.5th consumption rates, and has specific activities of ^{210}Pb and ^{210}Po near the upper range of the foodstuffs assayed (see figures 6 and 7, Chapter 3).

Table 31 Calculated Committed Effective Dose (μSv yr^{-1}) From the Ingestion of ^{210}Pb and ^{210}Po in Foodstuffs at the Sites Studied

Site	Age group	Pb-210 Dose	Po-210 Dose	Total Dose	^{210}Po Dose Fraction	^{210}Pb Dose Fraction
Brecon Beacon	6 - 12 months	125	198	323	0.61	0.39
Brecon Beacon	10 - 11 years	69	45	114	0.39	0.61
Brecon Beacon	14 - 15 years	87	36	123	0.29	0.71
Brecon Beacon	16 - 64 years	36	29	64	0.45	0.55
Holyhead	6 - 12 months	219	743	962	0.77	0.23
Holyhead	10 - 11 years	109	141	249	0.57	0.43
Holyhead	14 - 15 years	127	100	227	0.44	0.56
Holyhead	16 - 64 years	60	93	153	0.61	0.39
Newmarket	6 - 12 months	132	458	590	0.78	0.22
Newmarket	10 - 11 years	74	106	180	0.59	0.41
Newmarket	14 - 15 years	94	80	174	0.46	0.54
Newmarket	16 - 64 years	37	73	110	0.66	0.34
Cornwall	6 - 12 months	34	444	477	0.93	0.07
Cornwall	10 - 11 years	14	81	95	0.85	0.15
Cornwall	14 - 15 years	15	51	66	0.77	0.23
Cornwall	16 - 64 years	11	76	86	0.88	0.12
Romford	6 - 12 months	50	155	204	0.76	0.24
Romford	10 - 11 years	28	39	67	0.58	0.42
Romford	14 - 15 years	36	31	66	0.47	0.53
Romford	16 - 64 years	14	24	38	0.63	0.37
Penrith	6 - 12 months	145	139	284	0.49	0.51
Penrith	10 - 11 years	77	26	103	0.25	0.75
Penrith	14 - 15 years	96	17	112	0.15	0.85
Penrith	16 - 64 years	41	24	65	0.37	0.63
Didcot	6 - 12 months	75	364	439	0.83	0.17
Didcot	10 - 11 years	43	91	134	0.68	0.32
Didcot	14 - 15 years	54	71	125	0.57	0.43
Didcot	16 - 64 years	21	56	77	0.73	0.27
Sellafield	6 - 12 months	281	655	936	0.70	0.30
Sellafield	10 - 11 years	155	156	311	0.50	0.50
Sellafield	14 - 15 years	196	121	317	0.38	0.62
Sellafield	16 - 64 years	76	97	173	0.56	0.44
Warminster	6 - 12 months	31	72	102	0.71	0.29
Warminster	10 - 11 years	18	18	35	0.51	0.49
Warminster	14 - 15 years	22	14	36	0.39	0.61
Warminster	16 - 64 years	9	11	20	0.55	0.45
Drax	6 - 12 months	185	168	353	0.48	0.52
Drax	10 - 11 years	94	36	130	0.28	0.72
Drax	14 - 15 years	113	27	139	0.19	0.81
Drax	16 - 64 years	46	23	68	0.34	0.66
Whitehaven	6 - 12 months	219	349	568	0.61	0.39
Whitehaven	10 - 11 years	113	86	199	0.43	0.57
Whitehaven	14 - 15 years	139	67	206	0.33	0.67
Whitehaven	16 - 64 years	56	53	109	0.49	0.51

Table 32 Calculated Committed Effective Dose From the Ingestion of ^{210}Pb and ^{210}Po in Foodstuffs Separated According to Mean and 97.5th percentile consumption.

	Committed Effective Dose (μSv y^{-1})							
	16 - 64 years		14 -15 years		10 - 11 years		6 - 12 months	
Site	**Mean**	**97.5th**	**Mean**	**97.5th**	**Mean**	**97.5th**	**Mean**	**97.5th**
Holyhead	9	144	12	214	14	233	33	929
Sellafield	12	161	17	299	19	293	94	843
Newmarket	5	105	7	167	6	172	22	569
Cornwall	2	85	2	88	2	94	6	472
Didcot	0.3	77	1	123	0.8	133	2	438
Brecon Beacon	2	63	2	120	2	112	6	317
Drax	7	62	14	125	15	115	37	317
Penrith	3	62	5	107	4	100	10	274
Whitehaven	0.1	108	0.2	205	0.2	199	2	237
Romford	-	38	-	65	-	67	-	204
Warminster	-	20	-	35	-	35	-	102

As can been seen in Table 33 below, the largest range of total dose due to the 97.5th percentile consumption foodstuffs is in the 6 - 12 month age group, with the lowest range being the 16 - 64 years age group. Both the 10 - 11 years and 14 - 15 years age groups are virtually identical, and are intermediate between the 6 - 12 months and 16 - 64 years age groups.

In Table 33 below the proportions of the dose due to ^{210}Pb and ^{210}Po in the 97.5th percentile foodstuffs are presented. These data have also been graphed to aid comparison and are presented in figures 13 to 18.

Table 33 Committed effective dose (μSv yr^{-1}) due to ^{210}Pb and ^{210}Po from consumption of the largest 97.5th percentile consumption foodstuff and its associated dose ratio

	16-64 yr			14-15yr			10-11yr			6-12months		
Site	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio
Sellafield	76	47	1.6	96	123	0.8	123	97	1.3	492	171	2.9
Holyhead	45	34	1.3	58	88	0.7	74	70	1.1	428	123	3.5
Cornwall	71	7	10.0	47	10	4.7	77	10	7.7	423	24	17.6
Whitehaven	51	47	1.1	65	124	0.5	83	98	0.8	333	174	1.9
Newmarket	50	27	1.8	64	70	0.9	82	56	1.5	328	98	3.3
Didcot	33	15	2.2	41	38	1.1	53	31	1.7	213	54	3.9
Brecon Beacon	25	29	0.9	33	76	0.4	42	60	0.7	179	106	1.7
Romford	24	13	1.8	30	34	0.9	38	27	1.4	154	48	3.2
Penrith	21	31	0.7	14	81	0.2	22	64	0.4	124	113	1.1
Drax	14	33	0.4	18	88	0.2	23	69	0.3	94	122	0.8
Warminster	11	8	1.4	14	22	0.6	18	17	1.1	72	30	2.4

Table 34 Committed effective dose (μSv yr^{-1}) due to ^{210}Pb and ^{210}Po from consumption of the second largest 97.5th percentile consumption foodstuff and its associated dose ratio

	16-64 yr			14-15yr			10-11yr			6-12months		
Site	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio	^{210}Po	^{210}Pb	Dose ratio
Holyhead	45	22	2.0	58	30	1.9	74	30	2.5	296	82	3.6
Didcot	23	5	4.6	29	14	2.1	37	11	3.4	150	20	7.5
Newmarket	19	8	2.4	13	20	0.6	24	16	1.5	113	28	4.0
Sellafield	14	24	0.6	18	63	0.4	23	50	0.5	90	88	1.0
Drax	5	8	0.6	5	14	0.4	8	14	0.6	55	45	1.2
Cornwall	4	2	2	3	4	0.7	4	3	1.3	18	7	2.6
Brecon Beacon	3	6	0.5	2	9	0.2	2	7	0.3	15	16	0.9
Whitehaven	1	8	0.1	1	14	0.1	2	14	0.1	15	45	0.3
Penrith	2	8	0.2	2	11	0.2	2	10	0.2	11	25	0.4
Romford	0.1	0.4	0.2	0.2	1.2	0.2	0.2	0.8	0.2	0.9	1.5	0.6

Figure 13 Committed Effective Dose of ^{210}Po from the largest 97.5th foodstuff in the critical group

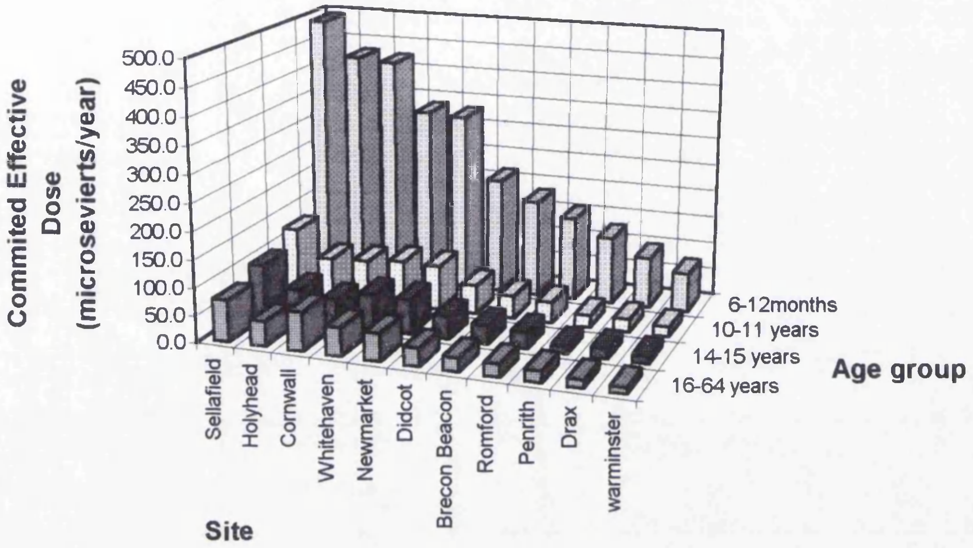
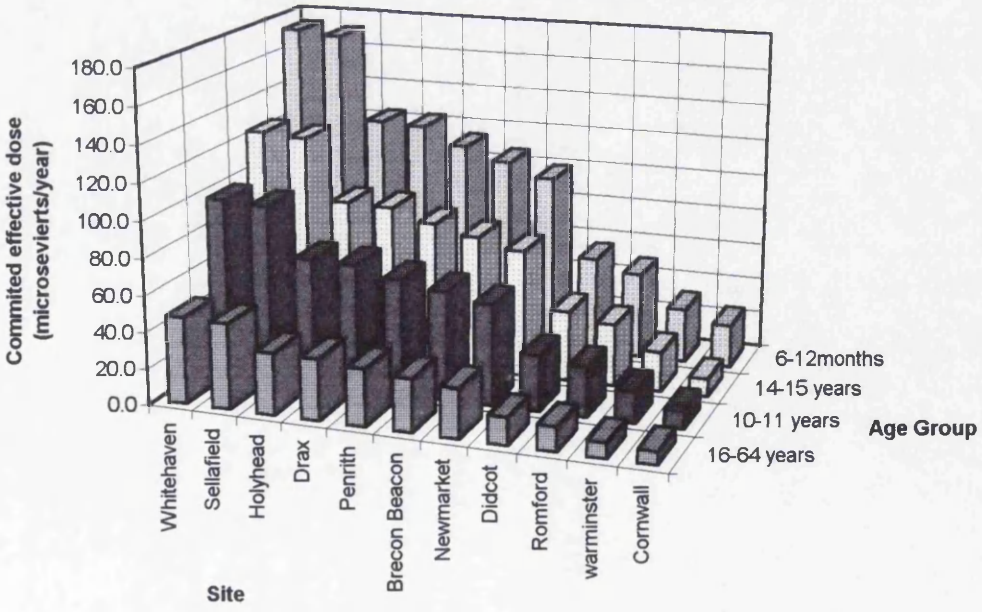


Figure 14 Committed Effective Dose of ^{210}Pb from the largest 97.5th foodstuff in the critical group



Examination of figures 13,14 and 17 for the largest 97.5th percentile consumption rate foodstuff shows that in general for most age groups, ^{210}Po makes up the greater contribution to the CED. This is particularly so at Cornwall because of the high ^{210}Po:^{210}Pb activity ratio in bovine liver. Indeed, for the 6 - 12 month age group, only at Drax does ^{210}Po contribute less than 50% of the dose.

For the second largest 97.5th percentile foodstuffs there is a similar trend in the dose (see figures15, 16 and 18), although this is not so marked as for the largest 97.5th percentile foodstuff, but the proportion of the dose due to ^{210}Po is still of some importance particularly in the 6 - 12 month age group.

Figure 15 Committed Effective Dose of ^{210}Po from the second largest 97.5th foodstuff in the critical group

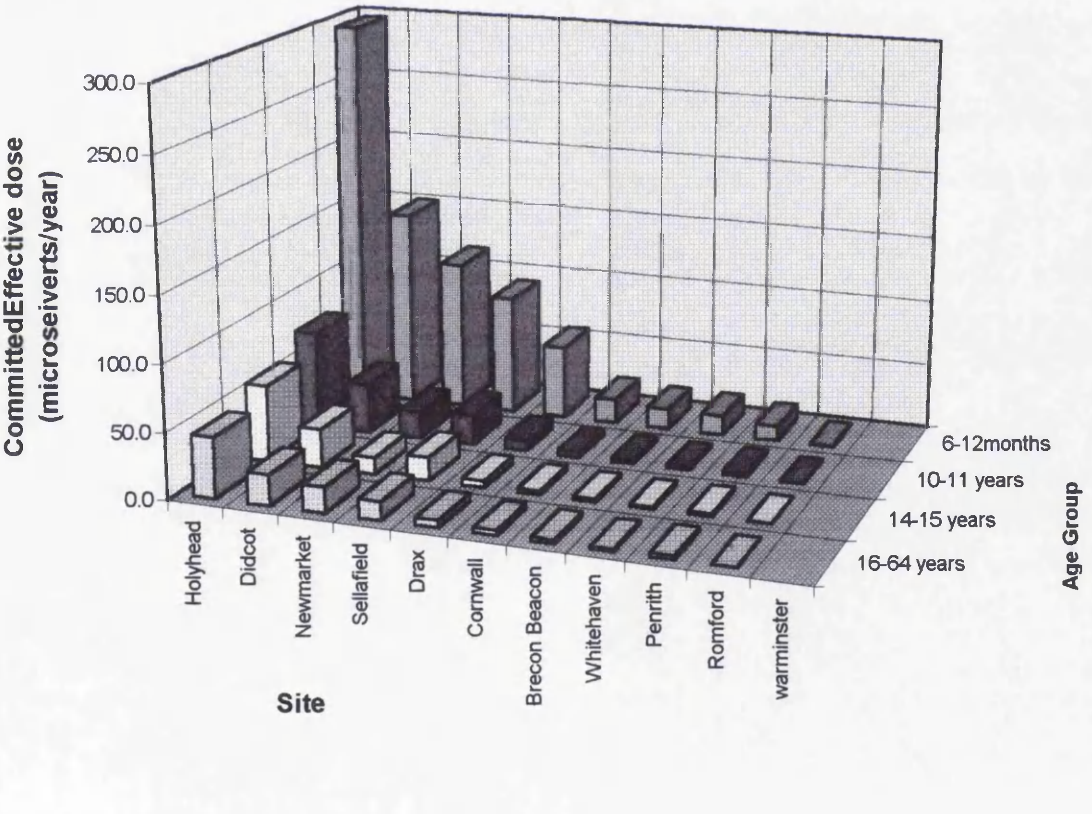
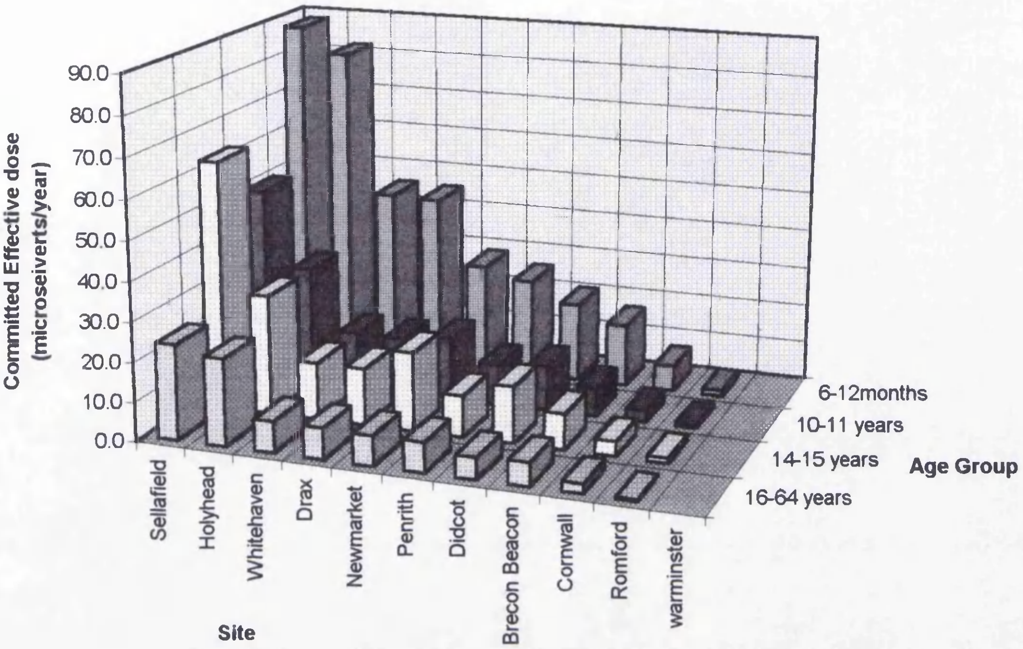


Figure 16 Committed effective dose from ^{210}Pb from the second largest food group in the critical group



It would be wrong to assume that equal specific activities of ^{210}Po and ^{210}Pb would give equal dose, as examination of the dose coefficients shows. It is possible to calculate the activity ratio of ^{210}Po: ^{210}Pb in foodstuffs which would give an equal dose from ^{210}Po and ^{210}Pb by simple algebraic manipulation of the equation to calculate the committed effective dose. This has been done and is presented in Table 35 below.

Table 35 The activity ratio of ^{210}Po: ^{210}Pb in foodstuffs which would give equal dose from ^{210}Pb and ^{210}Po, for different age groups.

Age Group (years)	<= 1	1-2	2-7	7-12	12-17	>17
^{210}Po: ^{210}Pb	0.323	0.409	0.5	0.731	1.187	0.575

Except for the 12 - 17 year age group this ratio is less than 1 with the lowest being the 6 - 12 month age group. Thus when the activity ratio is greater than the above values, the contribution to the dose from ^{210}Po becomes more important. These calculated activity ratios can be compared to those presented in figure 8 and Table 13 of Chapter 3. The CED ^{210}Po: ^{210}Pb ratios are plotted in figures 17 and 18 below.

Figure 17 Ratio of ^{210}Po:^{210}Pb CED in the largest food group

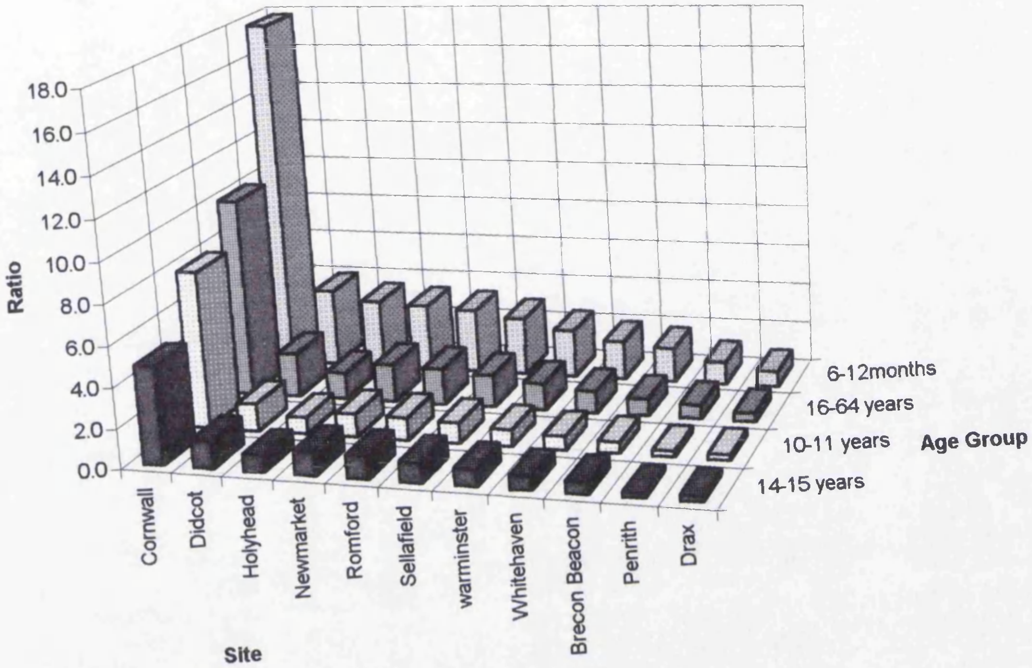
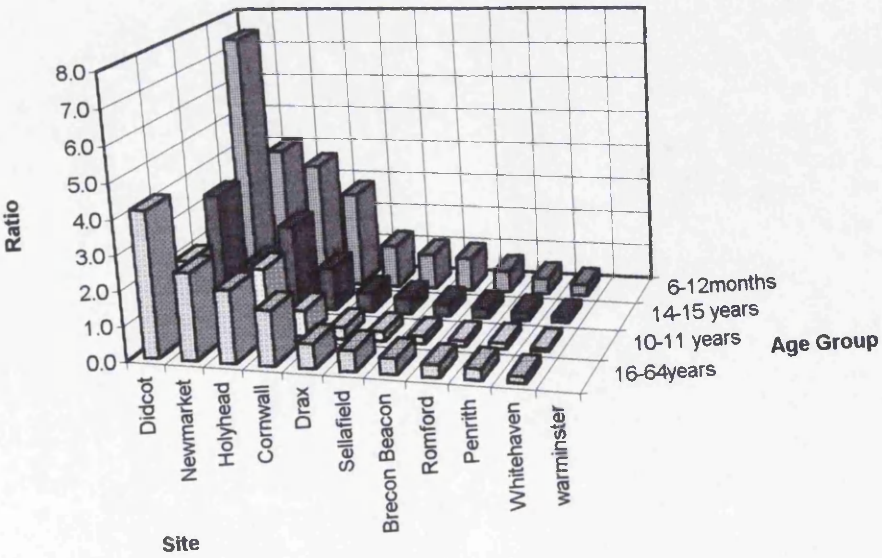


Figure 18 Ratio of ^{210}Po:^{210}Pb CED in the second largest food group



The levels of the doses received are all within the 1 mSv annual limit laid down by Her Majesty's Government although those for 6 - 12 month age group at Holyhead and Sellafield are very close to this limit. For this age group the choice of dose coefficient used has a large effect on the calculated dose. For ^{210}Pb the dose coefficient for the less than or equal to 1 year group is 2.3 times larger than the 1 - 2 year group. Also for ^{210}Po the effect is even larger, here the difference is a factor of 2.9. Thus, using the dose coefficients for the 1 - 2 year age group with consumption rates for the 6 - 12 month age group, would reduce the dose received to the 6 - 12 month age group by a factor of between 2.3 and 2.9.

It should be remembered, that these calculated doses are those which would be received, if the foodstuffs were consumed immediately after harvesting, and if no losses had occurred during processing, or through any preparation or cooking steps. This is particularly unlikely as most foodstuffs are stored for some time before consumption, and thus, depending on the extent of equilibrium between ^{210}Pb and ^{210}Po, the dose from ^{210}Po can decrease if the ratio is greater than one or vice versa. Cooking on the other hand could affect the specific activity of ^{210}Po due to its increased volatility, although this could be strongly affected by its speciation. The processing of foodstuffs could also change the specific activities of ^{210}Pb and ^{210}Po, in particular in the production of flour where the outer husk is removed from the gain before milling. If the distribution of ^{210}Pb and ^{210}Po within the grain is not homogeneous this will again affect the specific activities and the dose from consumption of the foodstuffs.

It must be stressed that the doses calculated are a conservative estimate and are likely to be evened out due to the above effects and through the distribution of foodstuffs throughout the United Kingdom.

It is interesting to calculate the daily intake of ^{210}Pb and ^{210}Po in each age group from the specific activities found in this study and compare them with previous work. Table 36 shows the annual intake by ingestion of ^{210}Pb and ^{210}Po for each of the sites studied.

The intakes have been calculated as follows:

$I_{ING,f} = SP_{f,r} \times R_f$ where

$I_{ING,f}$ = Intake by ingestion of foodstuff *f*

$SP_{f,r}$ = Specific Activity of radionuclide *r* in foodstuff *f*

R_f = Annual intake of foodstuff *f*

From this, the total annual intake of each radionuclide can be found by summing the individual activities of each radionuclide by ingestion for each foodstuff, as shown below.

$I_{ING,r,s} = \Sigma I_{ING,f}$ where

$I_{ING,r,s}$ = Total annual intake by ingestion at site *s* of radionuclide *r*

$I_{ING,f}$ = Annual intake by ingestion of radionuclide *r* in foodstuff *f*

The specific activities of the annual intake of ^{210}Pb and ^{210}Po at each site and their ranges are tabulated below. These tables can be compared with Table 38 below which has data from other countries throughout the world.

Care must be taken in comparing the data from this study and world-wide data. The world-wide studies were undertaken using "market basket" studies or duplicate sample methods and, as such, we are not comparing like with like. This is because the foodstuffs in these studies have undergone some kind of processing or cooking stage and there is likely to have been a storage time before consumption whereas in this study the foodstuffs were analysed unprocessed and uncooked. Still, some differences can be identified.

For ^{210}Po, with the exception of the 6 - 12 month age group, the upper range of intake is significantly higher than that of other European countries and the United States. The Laplanders of Finland are the only Europeans with a higher intake (by an order of magnitude) but this is due to the high concentrations in reindeer meat which forms a large part of their diet. The higher intake of the Marshallese and Japanese populations reflects the higher consumption of seafoods with elevated levels in comparison to the consumption of terrestrial foodstuffs by the UK population.

Table 36 Annual intake (Bq) of ^{210}Pb and ^{210}Po for different age groups at the sites studied

Site	Age group	^{210}Pb Intake	^{210}Po Intake	Total Intake	^{210}Po Intake Fraction	^{210}Pb Intake Fraction
Brecon Beacon	6 - 12 months	14.9	7.6	22.5	0.34	0.66
	10 - 11 years	35.9	17.2	53.1	0.32	0.68
	14 - 15 years	45.6	22.3	67.8	0.33	0.67
	16 - 64 years	52.0	23.4	75.4	0.31	0.69
Holyhead	6 - 12 months	26.0	28.6	54.6	0.52	0.48
	10 - 11 years	56.9	54.2	111.0	0.49	0.51
	14 - 15 years	66.7	62.1	128.9	0.48	0.52
	16 - 64 years	86.5	77.3	163.8	0.47	0.53
Newmarket	6 - 12 months	15.7	17.6	33.3	0.53	0.47
	10 - 11 years	38.9	40.7	79.7	0.51	0.49
	14 - 15 years	49.5	49.6	99.1	0.50	0.50
	16 - 64 years	53.2	60.5	113.7	0.53	0.47
Cornwall	6 - 12 months	4.0	17.1	21.0	0.81	0.19
	10 - 11 years	7.3	31.2	38.4	0.81	0.19
	14 - 15 years	7.8	31.6	39.4	0.80	0.20
	16 - 64 years	15.1	62.7	77.7	0.81	0.19
Romford	6 - 12 months	5.9	5.9	11.8	0.50	0.50
	10 - 11 years	14.7	14.8	29.5	0.50	0.50
	14 - 15 years	18.7	18.9	37.6	0.50	0.50
	16 - 64 years	19.6	19.8	39.4	0.50	0.50
Penrith	6 - 12 months	17.2	5.3	22.6	0.24	0.76
	10 - 11 years	40.4	9.9	50.3	0.20	0.80
	14 - 15 years	50.1	10.2	60.2	0.17	0.83
	16 - 64 years	59.1	19.7	78.8	0.25	0.75
Didcot	6 - 12 months	8.9	14.0	22.9	0.61	0.39
	10 - 11 years	22.4	35.0	57.4	0.61	0.39
	14 - 15 years	28.4	44.3	72.7	0.61	0.39
	16 - 64 years	29.9	46.6	76.4	0.61	0.39
Sellafield	6 - 12 months	33.4	25.2	58.6	0.43	0.57
	10 - 11 years	81.4	60.0	141.4	0.42	0.58
	14 - 15 years	102.9	75.6	178.5	0.42	0.58
	16 - 64 years	110.1	80.3	190.5	0.42	0.58
Warminster	6 - 12 months	3.6	2.8	6.4	0.43	0.57
	10 - 11 years	9.0	6.9	15.9	0.43	0.57
	14 - 15 years	11.4	8.8	20.2	0.43	0.57
	16 - 64 years	12.0	9.2	21.2	0.43	0.57
Drax	6 - 12 months	22.0	6.4	28.5	0.23	0.77
	10 - 11 years	49.2	13.8	63.0	0.22	0.78
	14 - 15 years	59.3	16.4	75.7	0.22	0.78
	16 - 64 years	66.1	18.7	84.8	0.22	0.78
Whitehaven	6 - 12 months	26.1	13.4	39.5	0.34	0.66
	10 - 11 years	59.4	32.8	92.2	0.36	0.64
	14 - 15 years	73.2	41.4	114.6	0.36	0.64
	16 - 64 years	80.4	44.0	124.4	0.35	0.65

Table 37 The Overall Ranges of Annual Intake (Bq) for different age groups of ^{210}Pb and ^{210}Po

	Annual Intake Ranges (Bq)		
Age group	Total Intake	^{210}Pb	^{210}Po
6 - 12 months	6.4 - 58.8	3.6 - 33.4	2.8 - 28.6
10 - 11 years	15.9 - 141.4	7.3 - 81.4	6.9 - 60.0
14 - 15 years	20.2 - 178.5	7.8 -102.9	8.8 - 75.6
16 - 64 years	21.2 - 190.5	12.0 - 110.1	9.2 - 80.3

Table 38 Comparable World-wide Data on Annual Intake (Bq) by Ingestion of ^{210}Pb and ^{210}Po

Country	^{210}Pb	^{210}Po	Reference
Bulgaria	21.9		Keslev et al. (1975)
France	18.3		Servant et al. (1981)
Italy	40.2	40.2	Clemete et al. (1980)
West Germany	62.1		Globel et al. (1966)
	40.2	40.2	UNSCEAR (1988)
	25.6		Weisshaar (1993)
USSR	84.0	54.8	Ladinskya et al. (1973)
		51.1	Yeremolayeva et al. (1969)
UK		43.8	Hill (1965)
	30.0	28.5	Smith-Briggs et al. (1986)
	16.4		Chamberlain (1983)
	4.2 - 93.2		MAFF (1994)
India		20.5	Khandekar (1977)
(Bombay 1971 - 1978)	21.9 - 73.0		Shukla (1994)
Argentina		17.5	UNSCEAR (1972)
Finland (Lapland)	116.2	932.5	Kauranen et al. (1969)
USA (estimated mean)	19.0	21.9	Holtzman (1980)
	16.8	21.9	Spencer et al. (1977)
Japan	36.5 - 73.0		Tanaka et al. (1968)
	73.0	219.1	Yamamaoto (1993)
	80.4		Kanetani et al. (1981)
	190 - 310		Holtzman (1980)
(Marine foods only)	13.6		Shimizu (1983)
(Marine foods only)	8.0 - 15.3	175.3 - 252.0	Yamamoto et al. (1994)
Marshall Islands (mainly marine diet)	131.5	796.2	Noshkin et al. (1994)

For ^{210}Pb, for the 14 -15 years and 16 - 64 years age groups, the ranges of intake are similar to those of other European countries but less than that of the

Laplanders. In comparison with the Japanese population the intakes fall within their range but it should be noted that the estimates published in the literature cover a large range (8.0 - 310 Bq yr^{-1}).

Table 39 Calculated Annual Effective Doses from Published Ingestion Intakes

Country	^{210}Pb (μSv y^{-1})	^{210}Po (μSv y^{-1})	Total (μSv y^{-1})	Reference
Bulgaria	15			Keslev et al. (1975)
France	13			Servant et al. (1981)
Italy	28	48	76	Clemete et al. (1980)
West Germany	43			Globel et al. (1966)
	28	48	76	UNSCEAR (1988)
	18			Weisshaar (1993)
USSR	58	66	124	Ladinskya et al. (1973)
		61		Yeremolayeva et al. (1969)
UK		53		Hill (1965)
	21	34	55	Smith-Briggs et al. (1986)
	11			Chamberlain (1983)
	3 - 64			MAFF (1994)
India		25		Khandekar (1977)
(Bombay 1971 - 1978)	15			Shukla (1994)
	50			
Argentina		21		UNSCEAR (1972)
Finland (Lapland)	80	1119	1199	Kauranen et al. (1969)
USA (estimated mean)	13	26	39	Holtzman (1980)
	12	26	38	Spencer et al. (1977)
Japan	25			Tanaka et al. (1968)
	50			Yamamaoto (1993)
	50	263	313	
	55			Kanetani et al. (1981)
	131			Holtzman (1980)
	214			Shimizu (1983)
(Marine foods only)	9			Shimizu (1983)
(Marine foods only)	6	210	216	Yamamoto et al. (1994)
	11	302	313	Yamamoto et al. (1994)
Marshall Islands (mainly marine diet)	91	955	1046	Noshkin et al. (1994)

The intakes found from previous UK studies are lower than those found in this study apart from the MAFF (1994) data which are in good agreement. MAFF collected data on ^{210}Pb, as well as a range of artificial radionuclides, as part of a duplicate diet study performed at Sizewell, the site of a nuclear power station. The other data were based on foods that had undergone some kind of processing, storage, or cooking stage, and as such are not strictly comparable.

Using the ingestion intake data above, the dose from ingestion was calculated using the most recent dose coefficients for adults. The result of this calculation is shown in table 39.

For ^{210}Pb, the range of doses covers 3 - 213 μSv y^{-1}, with the median value being 26, and the average 43 μSv y^{-1}. The distribution is positively skewed due to the higher dose received by those populations which have a high proportion of marine foods or reindeer and caribou in the diet. Similarly for ^{210}Po, the distribution of dose is positively skewed with the range covering 21 - 1119 μSv y^{-1}, with the average being 217 and the median being 53 μSv y^{-1}. Again, the upper range of doses reflecting the higher proportion of marine foods or reindeer and caribou in the diet.

In comparison with the doses for adults found in this study, the world-wide data for ^{210}Pb have a larger range with the upper value being approximately twice that found here. Similarly for ^{210}Po the range of the world-wide data is larger, with the upper value over an order of magnitude higher than that found in this study. Again these reflect the higher proportion of marine foods or reindeer and caribou in the diet. Those populations (Laplanders and Marshall Islanders) at the upper end of the range would receive doses in excess of the UK Government guidelines.

Summary and Suggestions for Future Work

Although the data set is limited due to the sampling difficulties and availability of the sample type at each site, some conclusions can be formulated. As was to be expected, different foodstuffs contained different specific activities of ^{210}Pb and ^{210}Po. Similarly, the same foodstuffs from different sites contained different specific activities of ^{210}Pb and ^{210}Po. One problem in deciding whether enhancement had occurred at suspected TENR sites was that the control sites which were chosen (hopefully) to reflect background levels, in some cases had higher specific activities than the suspected TENR sites. The control sites were also chosen in areas of high and low rainfall in case this had an affect on the source term of ^{210}Pb and ^{210}Po but, no correlation was found. This effect is difficult to explain and thus merits further study.

A large range of ^{210}Pb: ^{210}Po activity ratios were found showing that these nuclides were not in secular equilibrium, again this should be expected considering the growth periods of the foodstuffs, the source term of the nuclides, and the dynamics of uptake of the nuclides by the foodstuffs. It is obvious that the dynamics of uptake by the foodstuffs plays an important role in the final specific activities of ^{210}Pb and ^{210}Po and this certainly merits further study in relation to the problem of TENR.

Using a variety of statistical techniques, attempts were made to determine if enhancement of the specific activities of ^{210}Pb and ^{210}Po in foodstuffs, was evident at any of the sites studied. Based on a comparison with the control sites, barley from Sellafield (nuclear Site), bovine liver from Cornwall (natural radioactivity site), and cabbage from Sellafield showed enhancement for ^{210}Po and, barley from Whitehaven (phosphate processing site) and cabbage from Sellafield showed enhancement for ^{210}Pb. With less rigorous criteria based on data from all of the sites, blackberries from Holyhead (metal smelting site), bovine liver from Penrith (control site) can be added to the enhancement list for ^{210}Pb, and, blackberries from Holyhead and wheat from Didcot (coal fired power station) can be added to the enhancement list for ^{210}Po. There is a definite need to re-sample at these site to confirm these conclusions. Multiple samples should be taken at each site so that the distribution of specific activities of ^{210}Pb and ^{210}Po in foodstuffs can be categorised. This is important so that the statistical analysis can be put on a sound basis.

Relative to world-wide data the enhanced UK sites would be classed as relatively minor in magnitude of enhancement, or not at all.

From a radiological point of view, the doses received through the consumption of the foodstuffs at the sites studied, are all within the UK Government limit of 1 mSv y^{-1}, although the dose to the 6 -12 month age group at Holyhead and Sellafield are close to this limit. If the dose coefficient for the 1 - 2 year age group rather than for the <1 year age group is used for the calculations of the dose to the 6 - 12 months age group, then the dose received is reduced dramatically. The Generalised method was used in the calculation of the doses. In this method national statistics on food consumption rates are used in the calculation. If the Site specific method was used then a survey of the consumption rates at each site may have found different consumption rates and have given some indication of the proportion of the consumption of locally produced foods at each site. This could have a large effect on the doses received at each site and would deserve further study.

Intake of ^{210}Pb and ^{210}Po in foodstuffs was calculated for each site and compared to world-wide data. In general the world wide data had a larger range than the UK data.

Appendix 1 Computer programme for the calculation of ^{210}Pb and ^{210}Po specific activities

```
10 PRINT"PROGRAMME TO CALCULATE PO-210 AND PB-210
CONCENTRATIONS"
20 PRINT"IN SAMPLES WHICH HAVE BEEN DEPOSITED TWICE ON SILVER"
30 PRINT"DISCS":PRINT:PRINT
31 L3=4.538E-07
40 INPUT"SAMPLE DESCRIPTION";A$
45 INPUT"WEIGHT OF SAMPLE IN GRAMS";W
46 Z=0
50 INPUT"HAS THE SAMPLE BEEN COUNTED TWICE (Y/N)";B$
60 INPUT"NUMBER OF DAYS TO FIRST PLATING";T1
70 INPUT"NUMBER OF DAYS TO FIRST COUNTING";T2
80 INPUT"ACTIVITY OF SPIKE USED (DPM) RE ACTIVITY ON SPIKE BOTTLE";PA
90 INPUT"NUMBER OF DAYS SPIKE DECAYED TO COUNTING";T5
100 INPUT"NUMBER OF COUNTS IN REGION A";A
110 INPUT"NUMBER OF COUNTS IN REGION B";B
120 INPUT"NUMBER OF COUNTS IN REGION C";C
130 INPUT"NUMBER OF COUNTS IN REGION D";D
140 INPUT"COUNTING TIME IN SECONDS";T6
150 GOTO 300
160 Z=1
170 PRINT"SECOND COUNTING DATA"
180 INPUT"NUMBER OF DAYS TO SECOND PLATING";T3
190 INPUT"NUMBER OF DAYS TO SECOND COUNTING";T4
200 INPUT"ACTIVITY OF SPIKE (TOTAL ACTIVITY IN DPM)";PA
210 INPUT"NUMBER OF DAYS SPIKE DECAYED TO COUNTING";T5
220 INPUT"NUMBER OF COUNTS IN REGION A";A
230 INPUT"NUMBER OF COUNTS IN REGION B";B
240 INPUT"NUMBER OF COUNTS IN REGION C";C
250 INPUT"NUMBER OF COUNTS IN REGION D";D
260 INPUT"COUNTING TIME IN SECONDS";T6
300 E=(A*C)/B:F=(A*D)/B
310 E1=0:F1=0
320 E1=(A*(C-F))/(B-E):F1=(A*D)/(B-C)
330 IF (E1-E)<.1 AND (F1-F)<.1 THEN GOTO 350
340 E=E1:F=F1:GOTO 320
350 PS=INT(B-E1-F1+D+C+.5):P0=INT(A+E1+F1+.5):PRINT"208-
PO";PS:PRINT"210-PO";P0
355 ES=SQR(PS)/T6:EO=((SQR(P0)/T6)*60)/2.22
360 PS=(PS/T6)*60:PS=PS*EXP(L3*T5*24*60):G=PS/PA
365 PRINT"OVERALL EFFICIENCY IS";G*100
366 INPUT"DETECTOR EFF IS-IN DECIMAL =";DE
367 PE=(G/DE)*100
368 PRINT"PLATING EFF IS";PE;"%"
370 P0=((P0/T6)*60)/(G*W*2.22):EO=(EO/W)/G
380 PRINT"ACTIVITY ON DATE OF COUNTING";P0*37;"+/-";EO*37;"BQ/KG"
```

```
381 DC=EXP(.005007*T2)
382 PP=P0*37*DC
383 EP=EO*37*DC
384 PRINT"210-Po ACTIVITY AT TIME OF SAMPLING=";PP;"+/-";EP;"BQ/KG"
400 IF B$="N" AND Z=0 THEN GOTO 40
410 IF Z=0 THEN C1=P0:C3=EO:GOTO 160
420 IF Z=1 THEN C2=P0:C4=EO
1120 L0=.00502
1130 L1=.000085
1140 A=0:B=0:C=0:D=0:E=0:F=0:G=0
1280 A=EXP(-L1*T1)*(1-EXP(-L0*(T3-T1)))*(EXP(-L0*(T4-T3)))
1290 B=C2*(1-EXP(-L0*T1))
1300 C=EXP(-L0*T1)
1310 D=C1/EXP(-L0*(T2-T1))
1320 PO=(D-(B/A))/C
1330 P1=C2/A
1335 P3=C4/A
1336 P41=EXP(-L0*T1)
1337 P42=EXP(-L0*(T2-T1))
1340 P4=((P41/P42)*C3)^2
1350 P4=SQR(P4+((((1-(EXP(-L0*T1)))*(P41))/A)*C4)^2)
1370 PRINT"SAMPLE ";A$
1380 PRINT"==========================="
1390 PRINT
1400 PRINT "210-PB = ";P1*37,"+/-";P3*37,"BQ/KG"
1410 PRINT
1420 PRINT "210-PO = ";PO*37,"+/-";P4*37,"BQ/KG"
1430 PRINT
1440 PRINT "210-PO/210-PB = ";PO/P1,"+/-";PO/P1*SQR((P4/PO)^2+(P3/P1)^2)
1680 GOSUB 2000
1999 END
2000 PRINT"DO YOU WISH TO CALCULATE ANOTHER SAMPLE"
2010 INPUT"ANSWER 1 OR 0";X
2011 IF X=1 THEN GOTO 10
2070 RETURN
```

Appendix 2 Food Consumption Data from Byrom et al., 1996

Consumption of food by infants aged 6 to 12 months (kg person-1y-1).

Food group	% Consumers	Average (mean per caput)	Mean (Consumers)	Median (Consumers)	95th percentile (Consumers)	97.5th percentile (Consumers)
Imported fruit	88.5	9.5	10	6	35	45
Domestic fruit	84.8	7.5	9	6	25	35
Nuts	4	0	1	1	2	2
Potatoes	95.4	10	10	9	25	35
Root vegetables	93	4.5	5	4.5	10	15
Potatoes and root vegetables	**95.9**	**15**	**15**	**15**	**35**	**45**
Other imported vegetables	55.7	1.5	3	2	8.5	15
Green vegetables	53.6	2	3.5	2.5	9.5	10
Other domestic vegetables	91.8	3	3	2.5	9	10
Green and other domestic vegetables	**93.4**	**5**	**5**	**4**	**15**	**15**
Mushrooms	4.3	0	0.6	0.6	1.5	1.5
Sugar	96.9	3	3	2.5	7.5	8.5
Honey	6.9	0.2	2	1	7.5	7.5
Pig meat	79.5	1	1.5	1	4	5.5
Cattle meat	86.6	3	3	2.5	8	10
Sheep meat	65.7	0.6	0.8	0.6	2	3
Offal	33.8	0.4	1	0.6	3.5	5.5
Poultry	61.2	1	2	1.5	5	5.5
Game	-a	-a	-a	-a	-a	-a
Oil (non-dairy)	96.1	2	2	1.5	5.5	6.5
Milk	97.9	120	130	120	290	320
Butter	25.8	0.3	1	1	3	3.5
Cheese	62.7	1	2	1	5	7
Other milk products	71.9	10	15	15	40	45
Butter, cheese and other milk products	**90.3**	**15**	**15**	**10**	**40**	**45**
Eggs	84.6	4.5	5	4	15	15
Fish	55.7	2	3.5	2.5	10	15
Shellfish	-a	-a	-a	-a	-a	-a
Cereals	99.7	15	15	15	30	30

a No data available

Combined consumptions are shown in bold type.

Appendix 2 Food Consumption Data from Byrom et al., 1996

Consumption of food by 10 to 11 years age group(kg person-1y-1).

Food group	% Consumers	Average (mean per caput)	Mean (Consumers)	Median (Consumers)	95th percentile (Consumers)	97.5th percentile (Consumers)
Imported fruit	77.3	10	15	8.5	55	65
Domestic fruit	86.4	15	15	10	40	50
Nuts	23.3	0.4	1.5	1.5	5	7
Potatoes	99.9	45	45	40	75	85
Root vegetables	90.2	5.5	6	5	15	20
Potatoes and root vegetables	**100**	**50**	**50**	**45**	**85**	**95**
Other imported vegetables	72.8	5.5	7.5	6	20	20
Green vegetables	74.1	4.5	6	4.5	15	20
Other domestic vegetables	94.1	7.5	8	7	20	25
Green and other domestic vegetables	**96.6**	**10**	**15**	**10**	**30**	**35**
Mushrooms	17.1	0.3	1.5	1	3.5	4.5
Sugar	99.9	20	20	15	35	35
Honey	3.9	0.1	2	1	5.5	7.5
Pig meat	94.6	8	8.5	7.5	20	25
Cattle meat	96.9	10	15	10	25	30
Sheep meat	41	1.5	4	3	10	10
Offal	36.6	1	3	2.5	9	10
Poultry	68	3.5	5.5	4	15	15
Game	0.3	0	4	4	7.5	7.5
Oil (non-dairy)	100	10	10	10	20	20
Milk	99.9	110	110	110	220	240
Butter	73	2	3	2	8	9.5
Cheese	71.4	2.5	4	3	10	10
Other milk products	86.9	10	10	8.5	30	40
Butter, cheese and other milk products	**98.3**	**15**	**15**	**10**	**40**	**45**
Eggs	96.1	6.5	6.5	5.5	20	20
Fish	69.3	4	6	4.5	15	20
Shellfish	3.2	0.1	2.5	1.5	5	7
Cereals	100	45	45	45	70	75

Appendix 2 Food Consumption Data from Byrom et al., 1996

Consumption of food by 14 to 15 years age group(kg person-1y-1).

Food group	% Consumers	Average (mean per caput)	Mean (Consumers)	Median (Consumers)	95th percentile (Consumers)	97.5th percentile (Consumers)
Imported fruit	72.9	10	15	9	55	65
Domestic fruit	73.5	10	15	10	40	50
Nuts	21.2	0.5	2	1.5	7.5	9.5
Potatoes	99.4	55	60	55	110	130
Root vegetables	91.4	6.5	7.5	6	20	20
Potatoes and root vegetables	**99.9**	**65**	**65**	**60**	**120**	**130**
Other imported vegetables	69.5	8	10	9	30	35
Green vegetables	77.9	7	9	7	20	25
Other domestic vegetables	95.3	10	10	10	25	30
Green and other domestic vegetables	**97.5**	**15**	**20**	**15**	**40**	**45**
Mushrooms	14.3	0.3	2	1.5	5	5.5
Sugar	99.6	20	20	20	40	45
Honey	3.7	0.1	2	1	4.5	5
Pig meat	92.9	10	10	10	25	30
Cattle meat	93	10	15	10	30	35
Sheep meat	41.8	2.5	5.5	4	15	15
Offal	39.5	1.5	3.5	2.5	10	10
Poultry	66.1	4	6.5	5	15	20
Game	0.5	0	6	6	10	10
Oil (non-dairy)	100	10	10	10	20	25
Milk	99.8	110	110	95	220	260
Butter	77.1	3	4	3	10	15
Cheese	70.6	3.5	5	4	15	15
Other milk products	67.2	6.5	10	7	30	40
Butter, cheese and other milk products	**97.3**	**15**	**15**	**10**	**35**	**45**
Eggs	93.3	7	7	5.5	20	25
Fish	62.8	4	6.5	5	20	20
Shellfish	3.9	0.1	2.5	1.5	5.5	6
Cereals	100	50	50	50	90	95

Appendix 2 Food Consumption Data from Byrom et al., 1996

Consumption of food by 16 - 64 years age group (kg person-1y-1).

Food group	% Consumers	Average (mean per caput)	Mean (Consumers)	Median (Consumers)	95th percentile (Consumers)	97.5th percentile (Consumers)
Imported fruit	79.1	25	30	20	90	110
Domestic fruit	76.3	15	20	15	60	75
Nuts	27.9	0.8	3	1.5	9.5	10
Potatoes	98.9	50	50	45	100	120
Root vegetables	94.9	10	10	10	30	40
Potatoes and root vegetables	**99.9**	**60**	**60**	**55**	**110**	**130**
Other imported vegetables	57	5	9	7	25	30
Green vegetables	90.7	15	15	15	35	45
Other domestic vegetables	98.2	20	20	15	40	50
Green and other domestic vegetables	**99.2**	**30**	**35**	**30**	**70**	**80**
Mushrooms	37.3	1	3	2.5	8	10
Sugar	98.4	15	15	10	40	45
Honey	7.9	0.2	2.5	1.5	7.5	9.5
Pig meat	92.4	15	15	10	35	40
Cattle meat	89.7	15	15	15	40	45
Sheep meat	35.1	3	8	6	20	25
Offal	10	2	5.5	4.5	15	20
Poultry	72	7.5	10	8.5	25	30
Game	0.7	0	6	4	10	15
Oil (non-dairy)	99.9	10	10	10	25	25
Milk	99	95	95	85	210	240
Butter	68.3	3	4.5	3	15	15
Cheese	84	7	8	6.5	20	25
Other milk products	70.9	9.5	15	8.5	40	55
Butter, cheese and other milk products	**96.5**	**20**	**20**	**15**	**50**	**60**
Eggs	94.1	8	8.5	6.5	20	25
Fish	73.1	9.5	15	10	30	40
Shellfish	16.5	0.6	3.5	2.5	9	10
Cereals	99.9	50	50	50	90	100

Appendix 3 World-wide Data on ^{210}Pb and ^{210}Po Specific Activities in Terrestrial Foodstuffs

Location	Sample Type	^{210}Po Concentration (mBq kg^{-1})	^{210}Pb Concentration (mBq kg^{-1})	^{210}Po:^{210}Pb	Source of radionuclides	Reference
Argentina	Tomatoes	35			NS	Colangelo et al., (1992)
Argentina	Cauliflower	22			NS	Colangelo et al., (1992)
Argentina	Rice	110			NS	Colangelo et al., (1992)
Argentina	Spinach	320			NS	Colangelo et al., (1992)
Argentina	Potato	16			NS	Colangelo et al., (1992)
Brazil	Milk		5.0-60		HNRA	Amaral et al., (1988)
Brazil	Forage (grass)		1040-9220		HNRA	Amaral et al., (1988)
Brazil	Beetroot	333-587	474-503	0.70-1.08	HNRA	Santos et al., (1993)
Brazil	Beetroot	133	178	0.75	CA	Santos et al., (1993)
Brazil	Swiss chard	474-1357	539-1117	0.88-1.21	HNRA	Santos et al., (1993)
Brazil	Swiss chard	462	0.54	0.85	CA	Santos et al., (1993)
Brazil	Turnip	278-1130	470-1850	0.59-0.61	HNRA	Santos et al., (1993)
Brazil	Turnip	122	222	0.55	CA	Santos et al., (1993)
Brazil	Kale	455-560	450-700	0.69-.82	HNRA	Santos et al., (1993)
Brazil	Kale	467	667	0.7	CA	Santos et al., (1993)
Brazil	Cauliflower	279-375	538-894	0.39-0.48	HNRA	Santos et al., (1993)
Brazil	Cauliflower	167	278	0.6	CA	Santos et al., (1993)
Brazil	Cabbage	398-578	557-659	0.71-0.88	HNRA	Santos et al., (1993)
Brazil	Cabbage	227	518	0.75	CA	Santos et al., (1993)
Brazil	Broccoli	457-761	539-774	0.89-0.98	HNRA	Santos et al., (1993)
Brazil	Broccoli	447	467	0.96	CA	Santos et al., (1993)

Location	Sample Type	^{210}Po Concentration (mBq kg^{-1})	^{210}Pb Concentration (mBq kg^{-1})	^{210}Po:^{210}Pb	Source of radionuclides	Reference
Brazil	Chicory	522-1320	988-1790	0.52-0.74	HNRA	Santos et al., (1993)
Brazil	Chicory	944	1144	0.76	CA	Santos et al., (1993)
Brazil	Carrot	333-445	280-400	1.11-1.19	HNRA	Santos et al., (1993)
Brazil	Carrot	267	278	0.95	CA	Santos et al., (1993)
Brazil	Lettuce	882-1453	954-1978	0.73-0.92	HNRA	Santos et al., (1993)
Brazil	Lettuce	722	783	0.92	CA	Santos et al., (1993)
Brazil	Manioc	232-254	169-175	1.37-1.45	HNRA	Santos et al., (1993)
Brazil	Manioc	205	153	1.34	CA	Santos et al., (1993)
Brazil	Radish	289-863	824-1418	0.35-0.6	HNRA	Santos et al., (1993)
Brazil	Radish	288	562	0.58	CA	Santos et al., (1993)
Canada	Caribou kidney	259000±18000	84000±8000	3.64±1.59	NBA	Thomas et al., (1994)
Canada	Caribou liver	374000±25000	158000±10000	2.18±1.57	NBA	Thomas et al., (1994)
Canada	Caribou muscle	17000±2000			NBA	Thomas et al., (1994)
China	Rice	470	570	.82	HNRA	Zhu (1990)
China	Rice	2500	2400	1.04	ANUA	Zhu (1990)
China	Sweet potato	150	280	0.53	HNRA	Zhu (1990)
China	Sweet potato	510	540	0.94	ANUA	Zhu (1990)
China	Aubergine	300	280	1.07	HNRA	Zhu (1990)
China	Radish	100	240	0.42	ANUA	Zhu (1990)
China	Pork	460	20	23	HNRA	Zhu (1990)
China	Pork	140	330	0.42	ANUA	Zhu (1990)
Finland	Reindeer meat	2997-12395	118-296	11-42	CA	Kauranen et al. (1969)
Finland	Reindeer liver	37740-173530	10360-55870	3.1-5.0	CA	Kauranen et al. (1969)
Germany	Pigs liver	3330-4540	3150-4370	0.95-1.08	CA	Globel (1989)

Location	Sample Type	^{210}Po Concentration (mBq kg^{-1})	^{210}Pb Concentration (mBq kg^{-1})	^{210}Po:^{210}Pb	Source of radionuclides	Reference
Germany	Milk	5.1-35	5.2-32	0.90-1.34	CA	Globel (1989)
Germany	Apple	19-34	21-30	0.90-1.41	CA	Globel (1989)
Germany	Carrot	26-44	22-56	0.85-1.27	CA	Globel (1989)
Germany	Wheat flour	192-740	241-666	0.80-1.18	CA	Globel (1989)
Germany	Rye flour	296-481	222-555	0.80-1.33	CA	Globel (1989)
Germany	Pea	5.5-10.8	5.0-11.3	0.95-1.22	CA	Globel (1989)
Germany	Lettuce	8-20	10-18	0.71-1.17	CA	Globel (1989)
Germany	Spinach	6.5-9.9	7.4-9.1	0.79-1.11	CA	Globel (1989)
Germany	Cabbage	4.1-7.9	3.8-8.3	0.95-1.10	CA	Globel (1989)
Germany	Milk		20-50		NS	Weisshar (1993)
Germany	Cattle muscle		20-50		NS	Weisshar (1993)
Germany	Cattle liver		320-800		NS	Weisshar (1993)
Germany	Pig muscle		20-40		NS	Weisshar (1993)
Germany	Pig liver		200-280		NS	Weisshar (1993)
Germany	Chicken muscle		20-130		NS	Weisshar (1993)
Germany	Egg		20-70		NS	Weisshar (1993)
Germany	Leek		40-410		NS	Weisshar (1993)
Germany	Cabbage (red/white)		30-890		NS	Weisshar (1993)
Germany	Kale		90-1600		NS	Weisshar (1993)
Germany	Cabbage (savoy)		240		NS	Weisshar (1993)
Germany	Cauliflower		20-40		NS	Weisshar (1993)
Germany	Kohl rabi		30		NS	Weisshar (1993)
Germany	Broccoli		190-220		NS	Weisshar (1993)
Germany	Brussel sprouts		100-170		NS	Weisshar (1993)

Location	Sample Type	^{210}Po Concentration (mBq kg^{-1})	^{210}Pb Concentration (mBq kg^{-1})	^{210}Po:^{210}Pb	Source of radionuclides	Reference
Germany	Beetroot		100		NS	Weisshar (1993)
Germany	Carrot		40		NS	Weisshar (1993)
Germany	Radish		20		NS	Weisshar (1993)
Germany	Cucumber		70		NS	Weisshar (1993)
Germany	Lettuce		200-880		NS	Weisshar (1993)
Germany	Spinach		260-930		NS	Weisshar (1993)
Germany	Bean		90-210		NS	Weisshar (1993)
Germany	Onion		20-50		NS	Weisshar (1993)
Germany	Parsley		630		NS	Weisshar (1993)
Germany	Strawberry		30-60		NS	Weisshar (1993)
Germany	Blueberry		120		NS	Weisshar (1993)
Germany	Cranberry		340		NS	Weisshar (1993)
Germany	Redcurrant		90		NS	Weisshar (1993)
Germany	Blackberry		490		NS	Weisshar (1993)
Germany	Apple		20-140		NS	Weisshar (1993)
Germany	Potato		30-60		NS	Weisshar (1993)
Germany	Wheat		70-240		NS	Weisshar (1993)
Germany	barley		120-680		NS	Weisshar (1993)
Germany	Rye		120		NS	Weisshar (1993)
Germany	Maise		60		NS	Weisshar (1993)
India	Rice		30-488		NS	Lalit et al., (1980)
India	Wheat		55-481		NS	Lalit et al., (1980)
India	Pulse		41-240		NS	Lalit et al., (1980)
Israel	Chicken muscle	70-180			NS	Izak-Biran et al., (1989)

Location	Sample Type	^{210}Po Concentration (mBq kg^{-1})	^{210}Pb Concentration (mBq kg^{-1})	^{210}Po:^{210}Pb	Source of radionuclides	Reference
Israel	Chicken liver	210-1030			NS	Izak-Biran et al., (1989)
Israel	Egg	140-170			NS	Izak-Biran et al., (1989)
UK	Cattle liver	1.9±0.4	0.7±0.4	2.7±1.6	CPS	Smith-Briggs (1984)
UK	Cattle liver	3.6±0.6	0.5±0.3	7.2±4.5	CA	Smith-Briggs (1984)
UK	Green vegetables	222-3330		1-3	NS	Hill (1965)
UK	Carrot, Potato	37				Hill (1965)
UK	Bread, Cereal	37-259				Hill (1965)
UK	Dried Milk	74-222				Hill (1965)
UK	Beef muscle	111				Hill (1965)
UK	Lamb muscle	111				Hill (1965)
UK	Beef, Lamb liver	148-3700		0.7		Hill (1965)
UK	Beef, Lamb kidney	1776-9990		0.05-1		Hill (1965)
UK	Lamb kidney	3330-66600		0.2		Hill (1965)
USA	Milk	22.5-106	74-246		RPA	Staples et al., (1994)
USA	Cattle liver		592		RPA	Stricker et al., (1994)
USA (New Mexico)	Cattle muscle	310-3400	80	3.9-42.5	UTA	Lapham et al., (1989)
USA (New Mexico)	Cattle muscle	520±130	90±60	5.8±.96	CA	Lapham et al., (1989)
USA (New Mexico)	Cattle liver	12000-56000	380-3400	16.5-31.6	UTA	Lapham et al., (1989)
USA (New Mexico)	Cattle liver	9000±2000	250±140	36±22	CA	Lapham et al., (1989)
USA (New Mexico)	Cattle kidney	31000-65000	2900-13000	2.38-22.4	UTA	Lapham et al., (1989)
USA (New Mexico)	Cattle kidney	17000±5000	3000±600	5.7±2.0	CA	Lapham et al., (1989)

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