An investigation of the behaviour and distribution of radionuclides in the urban environment of Glasgow

By

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Abstract

The densely populated nature of urban areas makes them an important area of study with respect to environmental contamination, but these areas have often not received the attention they deserve. This thesis describes a study of the distribution of natural and manmade radionuclides within the City of Glasgow. The environment within Glasgow will not only contain natural radionuclides (which are ubiquitous in the environment), but it will also receive direct input of anthropogenic radionuclides from atmospheric fallout (weapons testing, satellite burn-up and Chernobyl) and possible point sources of both anthropogenic and natural radionuclides may arise from universities, research establishments and hospitals within the city. This initial investigation of the behaviour and distribution of radionuclides in the urban environment of Glasgow incorporates a city-wide investigation of radionuclide activities in a range of environmental materials and a more detailed study of radionuclide distributions and inventories in soil cores from a park within the city.

The city-wide investigation included sampling sites both close to, and remote from possible point sources of radionuclides and, over the three sampling times in July 2000 (Peurou, 2000), November 2000 and April 2001, found that ^{40}K specific activities generally decreased in the order grass> street dust > soil. The variability in the ^{40}K specific activities also decreased in this order. The ranges observed in ^{40}K specific activities were 197-2115 (Bq kg^{-1}), 225-479 (Bq kg^{-1}) and 189-1000 (Bq kg^{-1}), in grass, soil and street dust, respectively. Some of this variation has been attributed to environmental management of selected sites.

^{137}Cs specific activities ranged from 1.2- 22.5 (Bq kg^{-1}) in grass, 2.4-85.1 (Bq kg^{-1}) in soil and from below the detection limit to 211 (Bq kg^{-1}) in street dust. The ^{137}Cs street dust data population showed a broadly exponential trend with 2 outliers being observed, both near potential point sources of ^{137}Cs.

^{7}Be, a cosmic ray- produced radionuclide, was detected in grass with specific activities in the range 46-530 (Bq kg^{-1}) and in street dust in the range 44-595 (Bq

kg^{-1}). Considerable variation was observed both spatially and temporally, despite having a uniform atmospheric production rate and fairly uniform rainfall over the study area.

The highest specific activities of ^{226}Ra , a daughter nuclide of ^{238}U, were observed in soil (11.9- 44.4 Bq kg^{-1}), with fairly consistent results being observed both spatially and temporally.

^{131}I was detected in the Clyde Estuary, down stream of a hospital with a nuclear medicine facility. This, and the ^{137}Cs outliers in street dust, highlighted that this survey was sensitive enough to detect radionuclides that may be linked to a point source.

From the investigation of three cores of contrasting soil types from Pollok Park, a reference site for the city-wide investigation, the coniferous soil was found to have the lowest pH and highest organic content with a ^{210}Pb inventory which was comparable to other ^{210}Pb inventories in the literature for Central Scotland. The deciduous soil, which also had a low pH and high organic content, had a ^{210}Pb inventory approximately a factor of three lower than the coniferous soil. An exponential decrease in unsupported ^{210}Pb specific activities with increasing depth in both the coniferous and deciduous soils could be attributed either to accumulation of the soils or to downward migration of the ^{210}Pb. However, the depth distributions of ^{239,240}Pu in both cases were inconsistent with the apparent ^{210}Pb accumulation rates, indicating that the presence of natural and manmade radionuclides at depth in these profiles is almost certainly due to downward migration. The grassland soil, which had a much higher pH and lower organic content than both the forest soils, had a ^{210}Pb inventory comparable with the deciduous soil core. The grassland core exhibited a profile indicative of mixing, and accumulation of this soil is unlikely. All three cores suggest the downward movement of ^{137}Cs.

Plutonium was found to be associated with the organic phase of the forest soils but considerable revision of the BCR sequential extraction technique would be needed in future for use on such organic soils.

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BCR	Bureau of Certified References
DIPE	Di-isopropyl Ether
GDL	Generalised Derived Limits
GCSS	Glasgow City Scientific Services
LET	Linear energy transfer
LEGe	Low energy germanium detector
MAFF	Ministry of Agriculture, Fisheries and Food
NRPB	National Radiological Protection Board
NORM	Naturally occurring radioactive materials
ROW	Reverse osmosis water
SUERC	Scottish Universities Environmental Research Centre
SNAP	Systems for Nuclear Auxiliary Power
TAG	Aggregated transfer factor
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
USEPA	United States Environmental Protection Agency
WOS	Web Of Science

Chapter 1. Introduction

1.1 Radioactivity in the environment

There have been comparatively few studies of radionuclide distributions and behaviour in urban environments and the focus of this project was to provide such information for the City of Glasgow.

It is important to understand the behaviour and distribution of natural and man-made radionuclides in order to:

1) Accurately assess levels of human radiation exposure,
2) Evaluate the impact of any future planned, or accidental, release of radionuclides to the environment,
3) Identify any point source inputs of radionuclides to the environment.

This knowledge can be applied to the study area, which will receive radionuclide inputs from a variety of natural and man made sources.

Natural radionuclides are ubiquitous in the environment and make a major contribution to background radiation. It is therefore important to understand the distribution of these natural radionuclides in the study area. This area also receives direct input of anthropogenic radionuclides from atmospheric fallout (weapons testing, satellite burn-up and Chernobyl) and possible point sources of both anthropogenic and natural radionuclides may arise from universities, research establishments and hospitals within the city. A number of nuclear facilities are located at greater distances (e.g. Sellafield nuclear fuel reprocessing plant, nuclear reactors at Hunterston, Torness and Chapelcross and naval bases on the Firth of Clyde), but radionuclides released from these sources are unlikely to have any detectable effect within Glasgow.

1.1.1 - Primordial radionuclides

Primordial radionuclides are naturally occurring species with half-lives comparable to the age of the earth (Choppin et al., 1995). As primordial radionuclides were present at the time of the earth's formation they are naturally present in rocks and, as they decay, their daughter products accumulate in the geological environment. In some cases, these daughter nuclides are stable and analysis of parent: daughter systematics can be used in geochronological applications. In other cases, as discussed below, the decay products may be radioactive, giving rise to decay chains. Primordial radionuclides can be divided into lighter nuclides that occur singly (e.g.^{40}K) and heavier nuclides, as mentioned, that occur in decay series. There are three main radioactive decay series: The uranium series, which begins with ^{238}U, the thorium series, which begins with ^{232}Th and the actinium series, which begins with ^{235}U (Figures 1-3) (Eisenbud and Gesell, 1987). These three families of heavy radioactive elements account for much of the background radiation to which humans are exposed.

In a closed system, the daughter nuclides produced by radioactive decay in each series eventually achieve a state called secular equilibrium with their parent radionuclide. This state is achieved when the half-life of the parent nuclide is much longer than those of the succeeding species, such that there is no significant change in the concentration of the parent during the time interval over which its shorter-lived descendants attain equilibrium. When this state is achieved, all nuclides within a given decay chain decay at the same rate. The ^{232}Th series comes to equilibrium in about 50 years, in contrast to the ^{238}U chain, which takes longer than 10^6 years to come to equilibrium. This state of secular equilibrium only occurs in a truly closed system. However, in the environment, disequilibrium can occur if the system changes and is no longer 'closed'. For example, if the members of the decay chain are being transported by ground water (migrating), the differing physicochemical behaviour of each element in the chain may lead to differing migration rates or the precipitation or dissolution of the different decay chain members, thus leading to disequilibrium. These disequilibria can be used as an investigatory tool in earth and environmental sciences (Ivanovich and Harmon, 1992; Choppin et al., 1995).

As previously mentioned, primordial nuclides can also occur singly and an important example of such a nuclide is ^{40}K. Of the three naturally occurring potassium isotopes, only ^{40}K is unstable and decays mainly by β emission (89.33 %) with the remaining 10.67 % decaying by electron capture (Brown and Firestone, 1986). The isotopic abundance of ^{40}K is 0.0117 %, its half life is 1.2 x 10^9 years and it contributes significantly to the background radiation received by humans (Choppin et al., 1995). The average contents of ^{40}K, ^{238}U and ^{232}Th in Bq kg^{-1} in various rocks are summarised in Table 1.

Type of rock	**^{238}U**	**^{232}Th**	**^{40}K**
Igneous	4.81 x 10^{-5}	4.81 x 10^{-5}	8.14 x 10^{-4}
Sandstones	1.48 x 10^{-5}	2.41 x 10^{-5}	3.25 x 10^{-4}
Shales	1.48 x 10^{-5}	4.07 x 10^{-5}	8.14 x 10^{-4}
Limestones	1.48 x 10^{-5}	5.2 x 10^{-6}	8.14 x 10^{-5}

Table-1. Average ^{238}U, ^{232}Th and ^{40}K specific activities (Bq kg^{-1}) in various rocks (Eisenbud and Gesell, 1987)

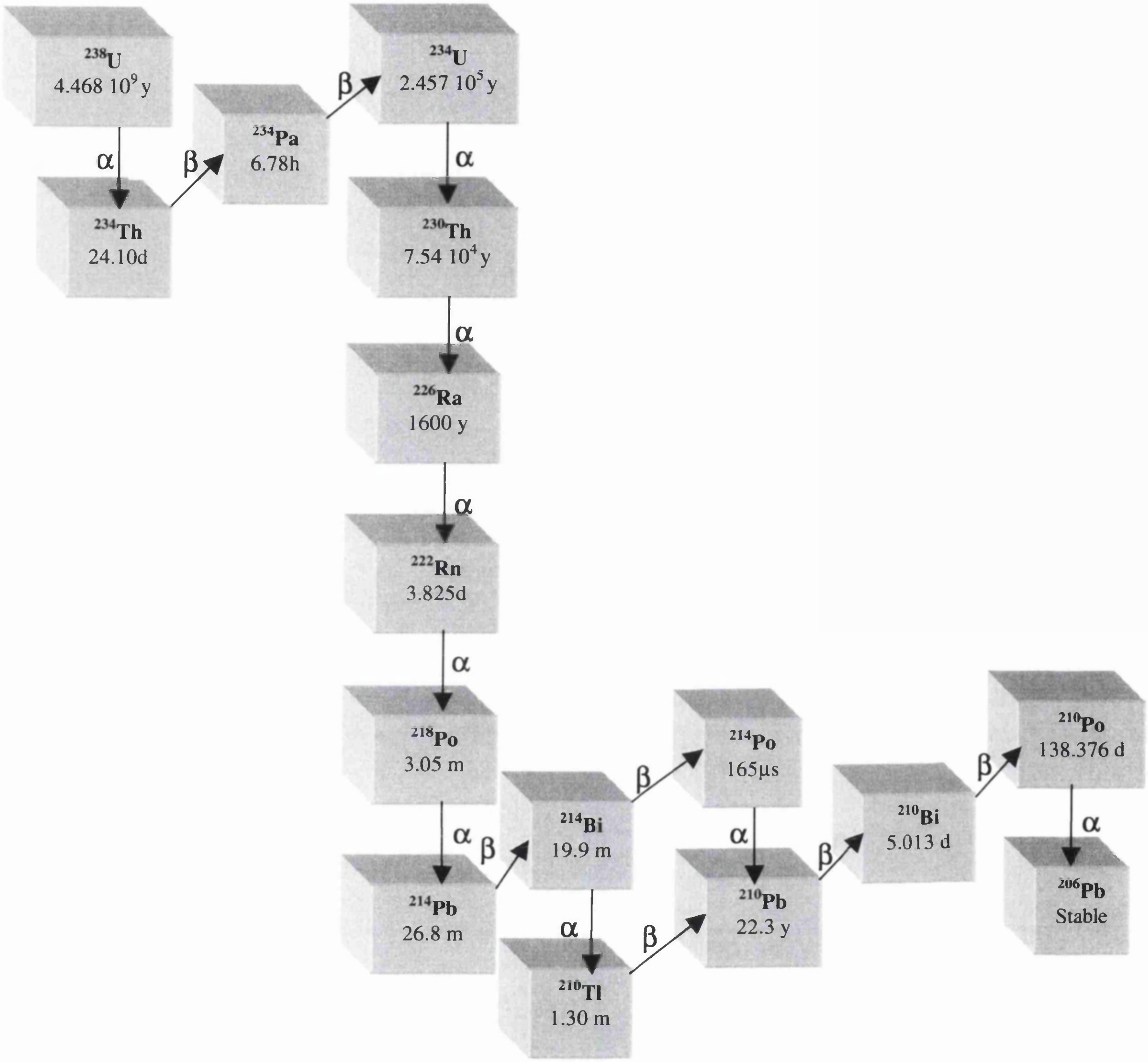


Figure 1. The Uranium Natural Radioactive Decay Series

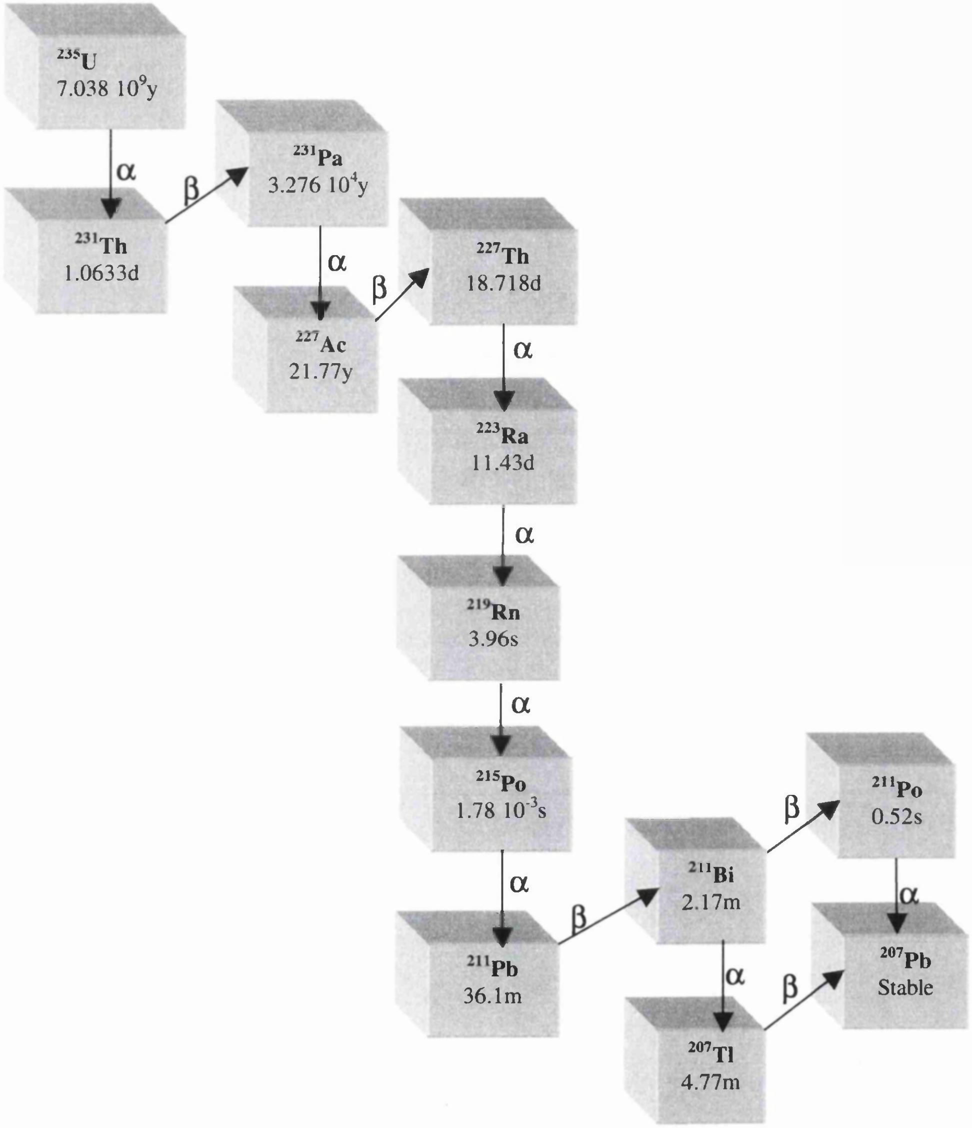


Figure 2. The Actinium Natural Radioactive Decay Series

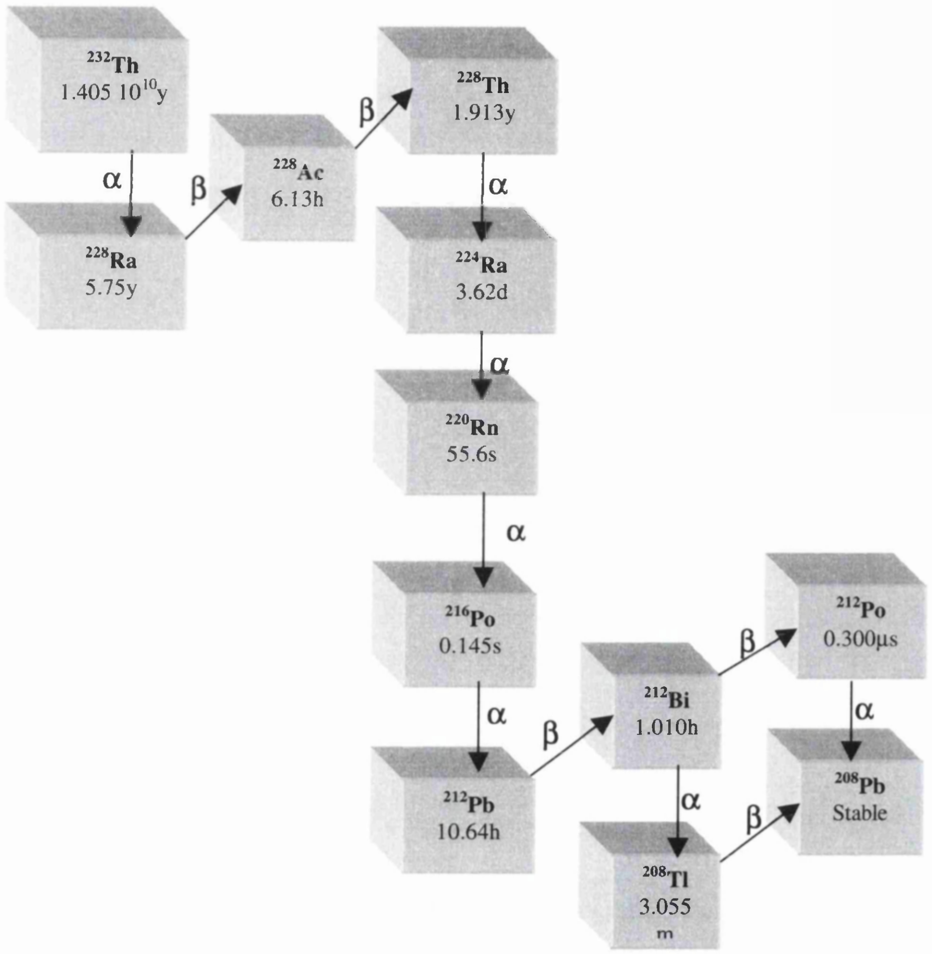


Figure 3. The Thorium Natural Radioactive Decay Series

1.1.2 - Cosmogenic radionuclides

Cosmogenic radionuclides are ubiquitous throughout the environment. They originate mostly in the upper atmosphere due to the interaction of the galactic cosmic ray flux with the earth's atmosphere. Interactions of primary cosmic rays and their products from interactions with nuclei can induce nuclear reactions in the atmosphere to generate radionuclides. Primary cosmic radiation comprises predominantly protons, with a smaller percentage of α particles, nuclei and electrons making up the remainder. The products of the interactions of cosmic rays with the atmosphere include electrons, gamma rays, neutrons and mesons. At sea level, mesons account for about 80% of the cosmic radiation and electrons about 20% (Eisenbud and Gesell, 1987). Both long and short-lived radionuclides are formed by these atmospheric interactions, a few of which are shown in Table 2.

Radionuclide	Half life	Decay mode and particle energy (Me V)	Atmospheric production rate (atoms $m^{-2}s^{-1}$)
^{7}Be	53.24 d	Electron Capture (0.862)	81
^{24}Na	14.965 h	β^- (1.389)	-
^{33}P	25.4 d	β^-(0.249)	-
^{3}H	12.33 y	β^-(0.0186)	2500
^{26}Al	$7.2*10^5$ y	β^+(1.16)	1.2
^{36}Cl	$3.02*10^5$ y	β^-(0.709)	60
^{14}C	5730 y	β^-(0.1565)	17000-25000

Table. 2- Examples of Cosmogenic radionuclides (Choppin et al., 1995; Longworth et al., 1998)

Equilibrium is assumed to be established between the production rate and the mean residence time of these cosmic radionuclides in environmental reservoirs (the atmosphere, the sea, lakes, soil, plants etc). The global inventory of ^{14}C is approximately 8 500 PBq and of this amount, 140 PBq remains in the atmosphere, whilst the rest is incorporated in terrestrial material (Eisenbud and Gesell, 1987). Tritium, which decays by weak β^- emission to ^{3}He, is also an important cosmic ray produced radionuclide with a global inventory of approximately 1.3 x 10^{18} Bq . Tritium is rapidly incorporated into water and enters the hydrological cycle (Eisenbud and Gesell, 1987).

1.1.3 - Anthropogenic radionuclides

Man-made radionuclides are produced predominantly by induced nuclear fission but also by neutron capture reactions and nuclear fusion. These reactions can occur both in nuclear reactors and in nuclear explosions (Choppin et al., 1995). A wide range of nuclides can be produced by these processes, some of which (notably isotopes of Cs, Pu and Am) occur as low level contaminants in the study area.

Atmospheric testing of nuclear weapons was carried out in two main series from 1952-1958 and in 1961-1962 and has contributed significantly to the global distribution of anthropogenic radionuclides. After 1962, only a limited number of atmospheric tests were carried out, the last having been in 1980 (Eisenbud and Gesell, 1987; Hardy, 1974). There have been an estimated 520 atmospheric tests, mainly in the Northern Hemisphere, with a total explosive yield of 524 Mt. Of these bomb-produced radionuclides that were deposited on the earth's surface as fallout, 12% were deposited locally, near the explosion site; 10% were dispersed via tropospheric dispersion in the latitude of the test and the remaining 78%, the stratospheric component, was dispersed on a global scale. These weapons testing fallout radionuclides have now largely decayed to negligible environmental levels, with ^{14}C, ^{90}Sr, ^{137}Cs and Pu isotopes being the most significant residual species (Eisenbud and Gesell, 1987; Choppin et al., 1995). The decay of these radionuclides has in time, introduced other radionuclides into the environment. For example, ^{241}Am was not generated in significant quantities during weapons tests, but has subsequently been produced in the environment by decay of fallout ^{241}Pu.

A number of accidents involving radionuclides have added to this atmospheric fallout. An important example of this was the atmospheric burn-up in 1964 of an American satellite which contained a System for Nuclear Auxiliary Power (SNAP) ^{238}Pu power source. This accident over the Indian Ocean released 629 TBq of ^{238}Pu and delivered to the world's population a collective dose of 2100 man Sv (UNSCEAR, 1993). Nuclear reactor accidents have also been responsible for catastrophically releasing radionuclides into the environment, the most serious of which was an explosion in a power reactor in Chernobyl, Ukraine in April 1986. This accident resulted in the release of noble gases and other volatile fission and activation products. An estimated 2 EBq of radionuclides were released from this accident (Warner and Harrison, 1993).

Plutonium has several isotopes, the most important with respect to environmental contamination being ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu. ^{239}Pu and ^{240}Pu have similar energies of α particle such that they are irresolvable by α spectrometry. Thus, ^{239,240}Pu is used to express a composite activity for the two nuclides. ^{239}Pu has a half-life of 2.41 x 10^4 years and ^{240}Pu has a half-life of 6.56 x 10^3 years, with both isotopes being produced in nuclear reactors by neutron irradiation of ^{238}U and ^{235}U. ^{239}Pu is fissile and can be used in nuclear bombs and reactors while ^{240}Pu is not fissile and therefore cannot be used in bombs and reactors (Eisenbud and Gesell, 1987). Pu isotopes were important products of neutron capture reactions in weapons testing and have therefore been distributed worldwide. ^{238}Pu, however, has a half-life of 87.7 years and was only a minor constituent of nuclear weapons fallout. In the 1950/60s, global fallout contained ^{238}Pu/ ^{239,240}Pu with an activity ratio of about 0.025, which increased to about 0.04 after the SNAP satellite burn-up in 1964. ^{241}Pu has a half life of 14.4 years and, as mentioned above, decays to ^{241}Am (Choppin et al., 1995). ^{241}Am has a half life of 432 years. In addition to the production of ^{241}Am from decay of ^{241}Pu from fallout, ^{241}Am has been released to the environment from nuclear fuel reprocessing facilities. Sellafield, for example, had discharged a total inventory of 10^3 TBq of ^{241}Am in liquid effluent up to 1993 (MAFF, 1993).

^{137}Cs and ^{134}Cs are also important anthropogenic radioisotopes that occur as environmental contaminants. ^{137}Cs has a half life of 30.2 years, is a fission product and was deposited on the soil surface in the 1950s and 1960s for several years as a result of atmospheric weapons testing and again in 1986 for a few weeks as a result of the Chernobyl reactor accident.

Also, liquid effluent discharges from nuclear fuel reprocessing plants and nuclear reactors represent a pathway of ^{137}Cs to the environment. ^{134}Cs is a neutron capture product produced in nuclear reactors and was not present in nuclear weapons fallout. ^{134}Cs, as well as ^{137}Cs, was present in the Chernobyl derived fallout with a ^{134}Cs/^{137}Cs activity ratio of about 0.5 (Eisenbud and Gesell, 1987)

Post deposition, Pu and Cs isotopes have different geochemical behaviours and therefore behave differently in the terrestrial environment (see section 1.3.4).

As well as being produced cosmogenically, tritium is also produced in the nuclear energy cycle, with some release to the atmosphere and the hydrosphere. The emissions differ between plants but a typical reprocessing plant could contribute an annual global injection of tritium in the order of 10 PBq (Choppin et al., 1995). This causes local increases in ^{3}H activities in environmental materials.

1.2 Radiation doses and the public

1.2.1 - Sources of exposure

When radiation interacts with matter, it results in an absorbed dose. This is the energy imparted to a small element of material divided by the mass of that element. The unit of absorbed dose is the joule per kilogram, called the gray (Gy). This unit, however does not take into account the type of radiation and the part of the body which has been exposed. The effective dose, which takes these factors into account is quantified by the sievert (Sv) (UNSCEAR, 1993). Among the many contributors to the radiation dose received by the public, the greatest comes from the naturally occurring radionuclides, particularly the uranium and thorium series radionuclides and ^{40}K (NRPB, 1998). NRPB has estimated that the average annual dose to the UK population from all sources, both natural and anthropogenic is 2.6 mSv. The contribution of these different sources to this average annual dose is shown below in Figure 4 (NRPB, 1989).

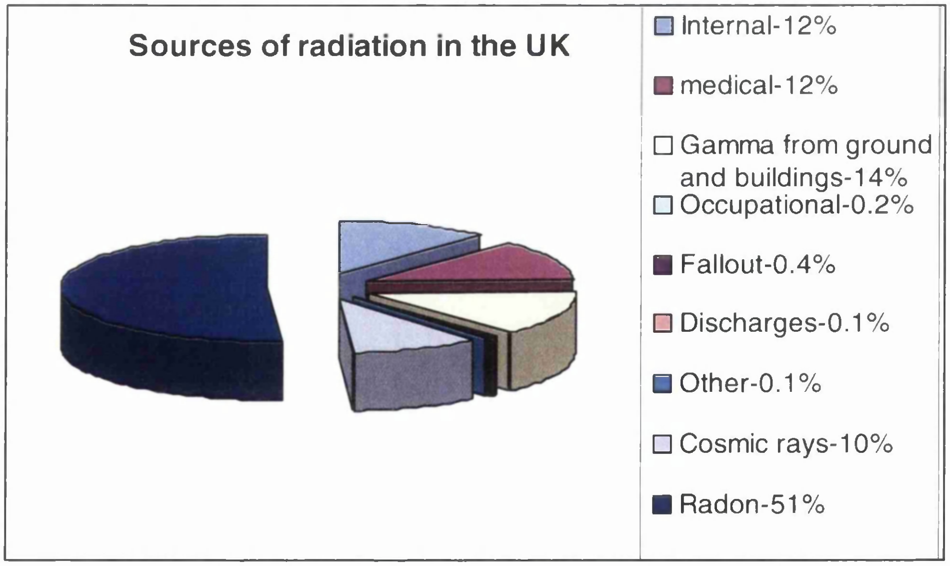


Figure 4 - Sources of radiation exposure in the UK (NRPB, 1989).

Internal exposure arises mainly due to the ^{40}K content of the human body. The average human is approximately a 3.6 x 10^3 Bq source of ^{40}K, because of all potassium, 0.0117% is present as ^{40}K (Brown and Firestone, 1986).

Radiation and radionuclides are used in medicine for a variety of diagnostic and therapeutic purposes. Also, radio-isotopic tracers are a useful tool in numerous areas of biomedical research, including metabolism studies, pharmacokinetics, and the detailed study of biomolecular interactions (WOS, 2001). Due to the large variety of radionuclides and the range of ways in which they are used, it is difficult to pinpoint any specific risks to the public from medical uses of radionuclides. Some hospitals incinerate their waste and also release radionuclides locally to the sewerage system. However, as short half-life radionuclides are most common in most medical procedures and research, they are unlikely to have severe long-term effects on the environment or the population. Examples of short-lived radionuclides commonly used in medicine include ^{99m}Tc which has a half-life of 6.006 hours (Longworth et al., 1998) and ^{131}I which has a half-life of 8.04 days (Germain, 1993). The same is often true of research laboratories and universities, in that wide ranges of radionuclides are used in research, but at low levels. They are tightly controlled and probably present little threat.

Atmospheric fallout constitutes a significant source of manmade radionuclides to the environment and contributes about 0.4% to average radiation exposure in the UK (see section 1.2.1).

1.2.2 – Non-nuclear industries as a source of radionuclides to the urban environment

Naturally occurring radioactive materials (NORM) used in industry, as well as a range of anthropogenic radionuclides used in industry, represent a potential source of radionuclides to the environment and could contribute to the radiation dose received by the population. As outlined in section 1.2.1, the contribution from industry and discharges is a very small percentage of the average annual dose, but it still may be highly significant to individuals living near, or in contact with, areas affected by local enhancement of radionuclides. This technologically enhanced radioactivity may arise from activities such as mining, phosphate-processing, fossil-fuel burning, oil and gas extraction, ore and heavy mineral processing and forestry processes such as pulp production and biofuel use (Baxter, 1996). The metal recycling or ‘scrap metal’ industry also has the potential to distribute natural and man-made radionuclides. This technological enhancement of NORM in the metal recycling industry was demonstrated in this country when ^{210}Po was selectively enriched in a large tin smelter in northeast England, which operated from 1937 to 1992. This selective enrichment of ^{210}Po resulted in a number of factory intermediates and waste gasses having elevated levels above the limit of 1.5 x 10^4 Bq kg^{-1} for classification as a radioactive substance (Baxter et al., 1996). Whilst the specific activities of other natural decay series radionuclides were lower than the limit for classification as a radioactive substance, they were within or close to the range that was estimated by the UK National Radiological Protection Board to be potentially capable of giving rise to an occupational exposure of 5 mSv y^{-1}.

To continue on this theme, a licensed industrial radioactive source which is disposed of improperly is said to be 'orphaned'(USEPA, 2001(a)). These orphaned radioactive sources often find their way to non-nuclear facilities, particularly scrap yards, steel mills, and municipal waste disposal facilities. The consequences of losing track of a radioactive source can be not only costly to the business involved but widespread contamination can ensue, resulting in exposure to radiation of workers and possibly the general public. If these sources are melted down for recycling, the radioactive source can contaminate the whole batch of metal and consequently, the consumer product that is ultimately being manufactured. Also, the act of melting is likely to allow the escape of volatile radionuclides in the gaseous emissions from the plant (Nuclear-Free Local Authorities, 2001).

To give an indication of the scale of the problem, there were an estimated 30,000 orphaned sources in the USA in 2001 and there have been 26 recorded accidental meltings of radioactive material in the USA since 1983 (USEPA, 2001(a)). Closer to home, in May 2000, the Avesta Steel plant in Sheffield melted a small ^{238}Pu source resulting in 16 tonnes of metal slag being contaminated and having to be designated as intermediate or low level waste (Nuclear-Free Local Authorities, 2001). However, there have been no reported meltings of radioactive sources in Scotland.

Over the last 2 or 3 years, many metal recycling companies have installed weighbridge radiation detection equipment. However, generally speaking, operations in Scotland are on a fairly small scale and therefore the cost of installing and running such systems is hard to justify. Detectors would require calibration and maintenance, which would have to be carried out by a qualified person or an approved body. Another important point is that if detection equipment was installed, it would most probably be for the detection of gamma radiation, thus excluding the possibility of detecting any contamination from alpha or beta emitters such as ^{210}Po as in the case of the Capper Pass refinery (Baxter et al., 1996). There are no regulations, at present, requiring detection equipment to be in place and SEPA does not hold records of those recycling facilities in Scotland that have radiation detection systems fitted.

The metal recycling industry is likely to be more vigilant with wastes originating from places where regulation of radioactive substances and wastes are less rigorous than in the UK and hence more likely to contain radionuclides (orphaned sources) but without thorough use of detection equipment, the potential for dispersal of radionuclides still exists. Effective regulation of radioactive substances and wastes, controls on the imports of radioactive wastes, detection systems at points of importation into the UK and the EU, effective communication and intelligence sharing, as well as detection systems within the metal recycling industry itself will all play a part in reducing the risks.

1.2.3 - Generalised Derived Limits

As a means of providing reference levels for public protection, Generalised Derived Limits (GDLs) for selected radionuclides and environmental media have been estimated (NRPB, 2000). These GDLs are intended for use as reference levels and are calculated for uniform conditions over a year, using deliberately cautious assumptions concerning the habits and location of the critical group of exposed individuals in a population. Where appropriate, the calculations take into account the conditions of discharge and the prevailing weather. There are GDLs set for many environmental media and, most importantly for this work, there are limits set for well-mixed soil and grass. There are, however, no GDLs set for street dust. If a guideline value for street dust was to be estimated for the purpose of this work, it would be significantly lower than the soil limit. This is because street dust is more likely to be re-suspended in dry weather than a well-mixed soil and therefore more likely to be inhaled or ingested by members of the population.

The only GDL set for an anthropogenic radionuclide that is of direct significance to this work is that for ^{137}Cs as shown in Table 3. When GDLs are set for natural radionuclides, they only relate to possible increases in specific activities resulting from human activities, but they are useful indicators against which to assess the results from this work (Table 3).

	^{137}Cs (Bq kg^{-1})	^{210}Pb (Bq kg^{-1})	^{235}U (Bq kg^{-1})	^{238}U (Bq kg^{-1})
Well-mixed soil	1000	80	7000	20 000
Grass	3000 (dry weight)	-	-	-

Table 3 – Selected Generalised Derived Limits (NRPB, 2000)

1.2.4 - Effects of radiation on humans

Radiation interacts with, and causes damage to, biological tissue by ionisation. It can induce changes in our DNA, the basic material that controls the structure and function of cells that make up the human body. The amount of damage caused, and the time between exposure and the damage being expressed, depends on the type of radiation and the dose received (see also section 1.2.6) (UNSCEAR, 1993). There are two broad categories into which radiation exposure can be classed. The first is **stochastic** which relates to long-term, low level exposure with no immediately observable effects. In this case it is difficult to link one source of radiation to an illness in an individual. The incidence of lung cancer in smokers can be said to be stochastic, as a particular smoker may never develop the disease whilst someone with a similar smoking history may do so (EB, 2001).

Non-stochastic or a deterministic effect is one which can be directly related to a causative agent. With non-stochastic effects there is a threshold below which no observable effects occur and above which changes are sure to occur. There is also a direct relationship between the size of the dose received and the severity of the effects, once the threshold has been breached (EB, 2001).

α, β and γ radiations have different characteristics and different interactions with matter. This is extremely important when considering dose received by humans. Alpha particles typically have energies in the range of 4 MeV to 9 MeV and have a highly efficient interaction with electrons when passing through matter. They have a high specific ionisation and a high linear energy transfer (LET) (Choppin et al., 1995; Longworth et al., 1998). Ejected electrons will produce further ionisation thus giving rise to secondary ionisation. Alpha particle ranges are very short and are, for example, only a few centimetres in air. As alpha particles transfer all of their energy over short distances, a relatively small volume of tissue receives a high dose. This is the reason that alpha radiation does the most damage to biological tissue.

Beta particles also deposit their energy in absorbers predominantly through coulombic interactions with electrons. These interactions result in ionisation or excitation of electrons in the atoms of the absorbers, with secondary ionisation occurring from ejected electrons. This secondary ionisation accounts for 70-80% of the total ionisation by beta particles. The specific ionisation and LET are lower for beta than for alpha particles but the beta particles have a greater range.

Gamma ray energies are typically in the range of 20 keV to 2 MeV and the decay is characteristic of the nuclide undergoing decay. Gamma radiation interacts with matter primarily through three processes: the photoelectric effect, Compton scattering and pair production (Choppin et al., 1995). Gamma rays are much more penetrating than both alpha and beta particles, but have lower LET and specific ionisation capabilities.

Not only does the type of radiation and the way the dose is delivered influence the severity of the damage caused to the tissue, but the area of exposure is also very important. Localised exposure to the limbs, for example, will result in a less serious outcome than a whole body exposure or exposure to critically important organs (UNSCEAR, 1993).

1.2.5 - Human exposure pathways

The three pathways by which radiation from radionuclides can directly affect humans are: inhalation, ingestion and external exposure.

Inhalation of, for example, the daughter products of ^{222}Rn will result in alpha radiation interacting directly with the lung tissue. Ingestion of radionuclides occurs through food and drink, but could also affect children with a pica habit, who may ingest contaminated soil. External radiation comes from the ground and from building materials as outlined in section 1.2.1.

The numerous and complex environmental pathways by which radionuclides can reach humans are summarised below in Figures 5 and 6:

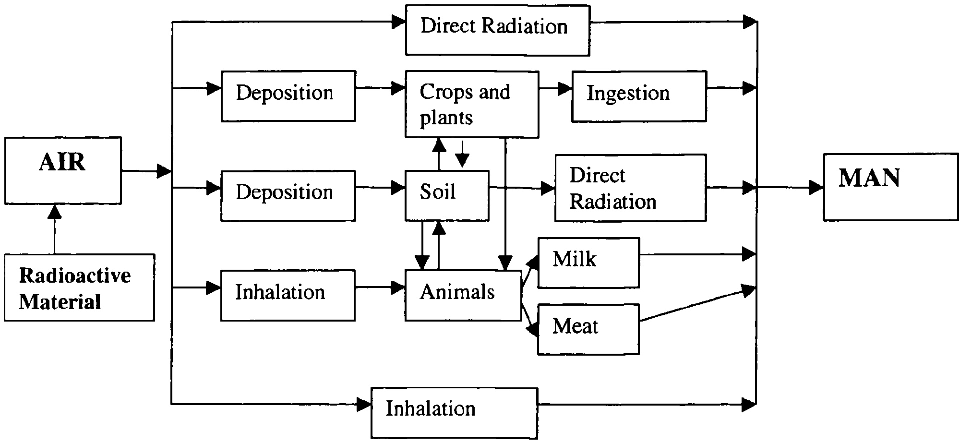


Figure. 5- Pathways between radioactive materials released to the atmosphere and man (adapted from Eisenbud and Gesell, 1987)

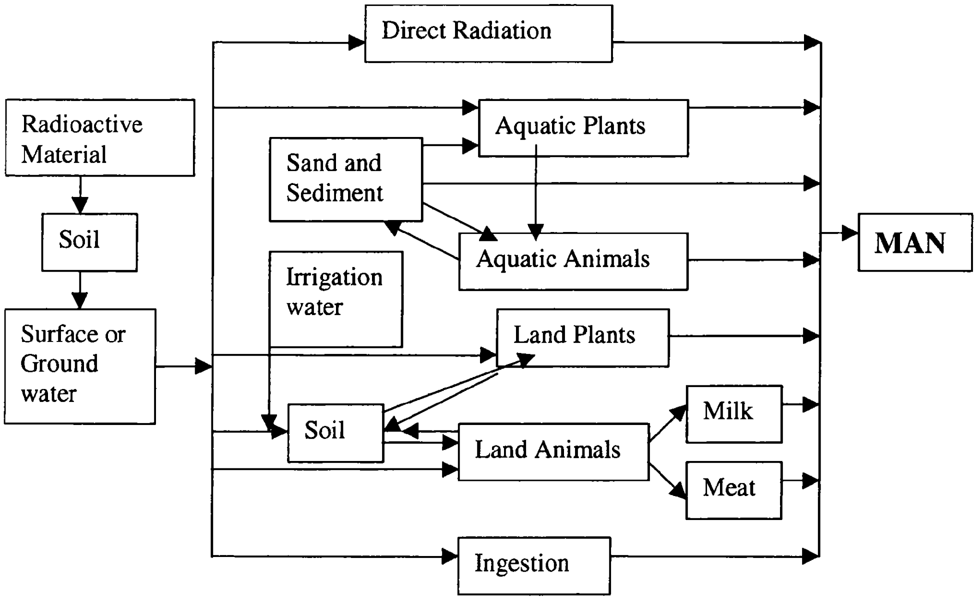


Figure. 6- Pathways between radioactive materials released to ground and surface waters, and man (adapted from Eisenbud and Gesell, 1987)

1.2.6 - Factors controlling dose

Numerous factors related to place and type of dwelling and lifestyle affect the level of radiation dose received. The dose received from cosmic radiation is markedly affected by altitude. The annual cosmic-ray dose is about 0.29 m Sv y^{-1} at sea level but for the first few kilometres above the earth's surface, the cosmic-ray dose rate doubles for each 2000 m increase in altitude. However, for the first 1000m, the *total* dose rate actually decreases with altitude above surface because of attenuation of the gamma rays from terrestrial sources occurs more rapidly than the increase in cosmic radiation (Schaefer, 1971). For example, residents of Leadville, Colorado (altitude 3200 m), receive about 1.25 m Sv y^{-1} from cosmic rays; more than four times the annual dose at sea level (Eisenbud and Gesell, 1987). Cosmic radiation also increases with latitude. Polar dwellers receive a higher dose than that received by people living closer to the equator.

Occupation and travelling habits can also effect radiation doses received. For example, passengers and crew of high-flying aircraft are subject to additional exposure from cosmic rays. A transcontinental flight will result in a dose of about 0.025 mSv. Cabin attendants and flight crew are estimated to receive an incremental dose of about 1.60 m Sv y^{-1} above that received at sea level (Eisenbud and Gesell, 1987).

Foodstuffs vary quite considerably in their level of radioactivity. Brazil nuts have been found to be much more radioactive than other foodstuffs due to the tendency of the Brazil nut tree to concentrate barium, a chemical congener of radium. The radium concentration of Brazil nuts is of the order of 1000 times higher than that of other foodstuffs and this does not depend on the radium or barium content of the soil on which the tree is grown (Eisenbud and Gesell, 1987).

The ground and building materials emit gamma radiation that comes primarily from the decay of natural potassium and uranium and thorium series radionuclides. The dose received from this source varies with the concentration of these elements in the surrounding rocks and building materials but is in the range of 120-1200 μSv y^{-1} in the UK (average 350 μSv y^{-1}) (Eisenbud and Gesell, 1987).

Levels of radon, the biggest single contributor to the radiation dose received in the UK, vary considerably from location to location depending on the geology, drift, soil type and soil moisture content. Indoor radon dose also depends on the type of building and the effectiveness of ventilation. The highest regional levels of radioactivity due to radon are mostly associated with the granites in southwest England, the Cairngorm and Etive granites in Scotland and carboniferous limestone formations, for example in Derbyshire (Appleton and Ball, 1995).

1.3 - Research area: history and environmental setting

1.3.1 - Geological setting of Glasgow

Geology is an important factor in determining background radiation levels as different rocks can have markedly different activities of uranium decay series radionuclides, thorium decay series radionuclides or ^{40}K. The bedrock of the Glasgow area consists of lower, middle and upper coal measures, basic intrusions, the passage group and the upper limestone group (BGS, 1986).

The lower part of the upper limestone group comprises thick, coarse-grained sandstones, with the remainder being fine-grained sandstones with mudstones and a few coal measures. The Passage group consists mainly of pebbly sandstones and interbedded seatclays with thin coals and mudstones (BGS, 1986).

The lower coal measures are also sandy with seatclays. Most of the middle coal measures consist of sandstones and mudstones in approximately equal proportions with numerous seams of coal. The middle and upper coal measures are characterised by the presence of coarse sandstones, with younger mudstones higher up the sequence. The strata have been reddened and many of the coal seams have been oxidised. Intrusive igneous rocks in the area are mostly olivine-dolerite sills of the late Westphalian age (~300 million years old) (BGS, 1986).

Superficial deposits are dominated by the Late Devensian ice sheet's terminal moraine in the east of the city and a submarine outwash fan in the west of the city. Glaciomarine muds occur widely. Numerous drift profiles or sequences have developed as a result of this complicated late quaternary history. However, till, which is the most widespread deposit, often forms the only drift directly resting on bedrock. Much of this till has been moulded into drumlins, for which the city is famous. Clays and silts from the area have been extensively exploited for brick and tile manufacture (BGS, 1986).

The River Clyde, around which Glasgow was built, is tidal through most of the city. It was the presence of coal measures and other mineral wealth, combined with the deep River Clyde that made Glasgow such a viable place for industry.

1.3.2 - City of Glasgow

Glasgow is Scotland's largest city with a current population of approximately 612,000 (Glasgow City Council, 2000). The city was a major industrial centre for over 100 years due to its ideal situation on the West Coast and next to the River Clyde. 1772 saw the first large vessels navigating up the River Clyde and this turned an already considerable tobacco trade into a booming one, along with the sugar trade. By the end of the 18th century, Glasgow was the UK's biggest sugar importer.

This industrial past has left a legacy of widespread and varied industrial contamination, which may provide potential sources of radionuclides to the environment. An example of the use of radionuclides in an old industry is the use of radium in the luminising paint for clock faces (Eisenbud and Gesell, 1987). Watchmakers and repairers in the City of Glasgow would have used this luminising paint. In addition to this, radium needles, which were use in the early days of radiotherapy, were made at Glasgow University by the Nobel Prize winner Fred Soddy (personal communication: Keith McKay). Use of radium in industry occurred out with, but close to the city of Glasgow at the radium factory in Balloch on the banks of the river Leven, and the Smiths Industries factory at Gowkthrapple near Motherwell, South Lanarkshire (personal communication: Keith McKay).

This industrial city had Scotland's first millionaires, who owned large estates and property. One of these estates in Glasgow is Pollok Park, which was used as a study site.

1.3.3 - Pollok Park

Pollok Park has been in existence for in excess of 700 years and covers an area of 361 acres (City of Glasgow District Council, 1985). There are golf courses around the park so, even though the park is within the city, it is remote from many potential sources of radionuclides. There are distinct areas of coniferous and deciduous woodland in the park, which take up 15% of the estate's area. There are also distinct areas of grassland and fertilised hay fields. This combination of contrasting environments in well defined areas in the estate makes it an excellent study area. The estate was gifted to Glasgow City Council in 1967 by the Maxwell family and now houses the Burrell Collection (City of Glasgow District Council, 1985). Soil characteristics of deciduous and coniferous woodland environments are outlined below.

(i) Deciduous forest environment

Deciduous soil is typically well aggregated and porous, is dark brown and grades gradually to a more mineral soil. It is typically rich in soil organisms, the most important of which is the earthworm. This type of soil is characterised by Mull humus. The C: N ratio of the upper horizons is typically between 10 and 15 (see Figure 7) (Fitzpatrick, 1974; The Royal Society of Chemistry, 1999)

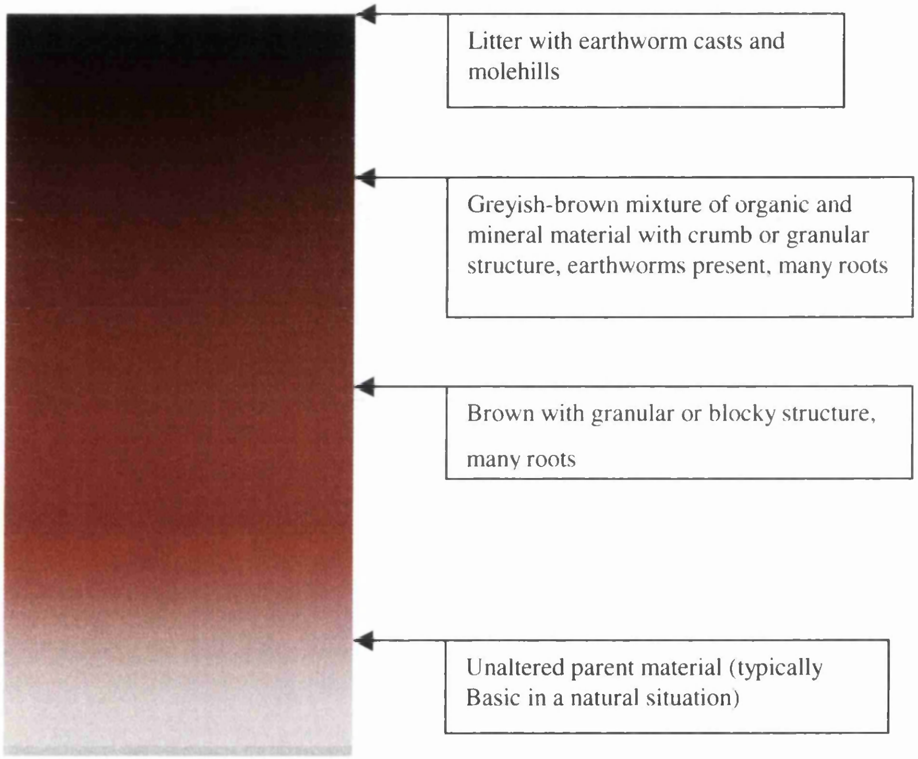


Figure. 7- A typical deciduous soil profile (diagram not to scale) (Fitzpatrick, 1974)

(ii) Coniferous forest environment

The coniferous forest environment typically exhibits a podzol profile, which is sharply differentiated and contains blackish organic layers (mor humus). This soil is most typically poorly drained and devoid of earthworms and has a lower pH than deciduous forest soil (Figure 8). The C: N ratio is typically greater than 30 and the soil is often rich in phenolic groups. The type and steric arrangement of the functional groups facilitates, or otherwise, the complexation of metal cations (Fitzpatrick, 1974; The Royal Society of Chemisty, 1999).

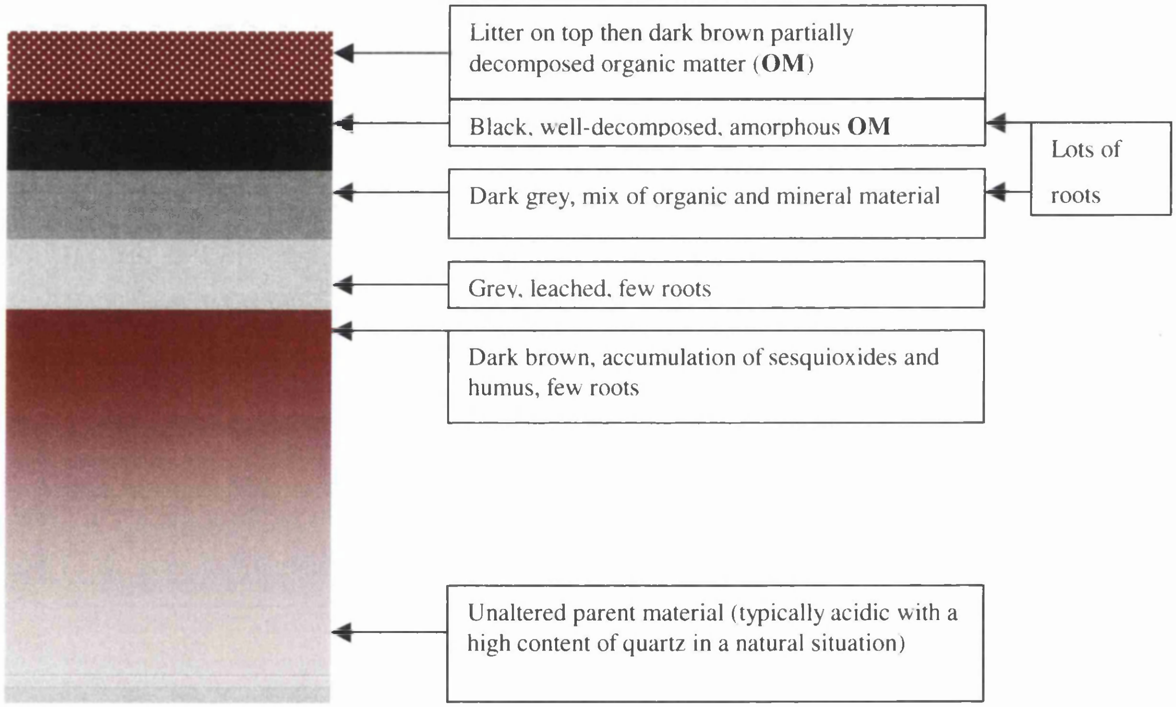


Figure. 8- A typical coniferous soil profile (diagram not to scale) (Fitzpatrick, 1974)

1.3.4 - Behavior of radionuclides in soils

Although some generalisations concerning the relative degree of fixation of many radionuclides in soil are possible, the behaviour of the radionuclides is dependent on site-specific factors such as rates of rainfall, drainage and local physicochemical conditions. However, when a radionuclide (in a soluble form) comes into contact with soil, it can adsorb to reactive coatings on particles, undergo ion exchange, precipitate as an oxide-hydroxide or sulphide, be complexed with organic compounds, or remain in ionic form (Schultz, 1965). The manner in which the radionuclides partition between these fractions (i.e. the physicochemical behaviour) will determine the degree to which soils can act as a sink or a potential diffuse source for radionuclides (Skipperud et al., 2000). When radionuclides are transported by water moving through porous or fractured geologic media, the phenomenon of sorption causes the rate of movement of the radionuclides to be reduced relative to the rate of movement of the water. This "retardation factor" is determined by the rates of sorption and desorption on media surfaces. The ions of some elements are bound so tightly to soil particles that they are nearly immobile. In this respect, cations are *generally* more strongly sorbed than anions because of the dominance of negatively charged particles on soil surfaces.

It was shown by Hardy (1974) that, years after the maximum fallout pulse in 1963, the inventories of ^{90}Sr, ^{137}Cs, and ^{239,240}Pu present in the top 30 cm of a sandy loam were nearly equivalent to the quantities estimated to have been deposited at that latitude. This indicated little removal of these radionuclides from the site. Another study by Alexander (1967) concluded that ^{137}Cs is more tightly bound by soil than ^{90}Sr and that the uptake of ^{137}Cs by plants is limited by its strong retention to clays (Livens and Baxter, 1988; Eisenbud and Gesell, 1987; Szerbin et al., 1999). Radiocaesium, however, does have a high degree of mobility in organic soil, but this movement can be restricted by the presence of small amounts of mineral matter. Interactions with illitic clays hold the ^{137}Cs in a relatively available form, with more permanent fixation occuring where interlayer collapse results in trapping of the ^{137}Cs within the mineral lattice (Livens and Loveland, 1988; Hird et al., 1996; Askbrant et al., 1996). This mobility of radiocaesium in organic soils may render it of limited value in providing chronological information, but it has been suggested that the presence of even small amounts of mineral matter may restrict this mobility (Szerbin et al., 1999). A study by MacKenzie et al., (1997) established that downward migration of radiocaesium happened at a faster rate immediately post-deposition (on a 5-year time scale) than longer term migration (30-year timescale) being characterised by apparent diffusion coefficients of the order of 10^{-7} cm^{2} s^{-1} and 10^{-8} cm^{2} s^{-1}, respectively (MacKenzie et al., 1997). The vertical distribution of ^{137}Cs activity, the percentage of exchangeable radioceasium in each horizon and the root distribution over the soil profile are all factors in governing the availability of ^{137}Cs transfer to plants (Fesenko et al., 2001).

In research carried out by Hogbom and Nohrstedt (2001), wood-ash from areas of forest contaminated by Chernobyl fallout was applied to previously relatively uncontaminated forest soil. Contrary to expectations, there was a decrease in the ^{137}Cs radioactivity in some of the soils. This decrease in radioactivity was partly due to the fact that one of the main constituents of wood-ash is K, which is antagonistic to ^{137}Cs (Hogbom and Nohrstedt, 2001). A study which investigated the associations of radionuclides with different molecular size fractions in soil solution concluded that ^{137}Cs was present in low molecular weight forms and, as such, was mobile in soil and potentially available for plant uptake (Nisbet et al., 1993). This finding was in contrast to the large proportion (61-87%) of the ^{239}Pu and ^{241}Am that was associated with colloidal and high molecular weight material and is therefore less available for plant uptake (Nisbet et al., 1993; Agapkina et al., 1995). The study by Nisbet et al. (1993) was carried out on a loam, a peat and a sandy soil and so some of the conclusions may not be directly transferable to the contrasting forest soils used in this current study. A novel method of investigating radionuclide associations involving gel chromatographic and gel electrophoretic separation, (Graham et al., 2000) showed that the association of U in soils was skewed towards larger humic molecules. These humic materials have strong complexing and redox reaction potential with metal ions (Choppin, 1988).

Plutonium generally forms insoluble fluorides, hydroxides and oxides in soil, and solubility in water is dependent on the redox conditions, pH and organic ligands present (Eisenbud and Gesell, 1987). A study by Skipperud et al. (2000), concluded that after only one hour of

contact time, 33% of the Pu (III, IV)-organic, 35 % of the Pu (V,VI) and up to 40% of Pu (III, IV) were already in the strongly bound fraction. This limited mobility may be related to the previously mentioned association with high molecular weight material. The conclusions of a study by Muramatsu et al. (2001) suggested that the mobility of Pu in the soil column is less than that of ^{137}Cs. Sequential extraction experiments (Livens and Baxter, 1988) identified major associations of plutonium with organic matter and found plutonium phase dominance to decrease in the order:
Organic > Oxide > Residual > Absorbed > Exchangeable (Livens and Baxter, 1988).

There has been a considerable amount of research on lead in soils and the use of ^{210}Pb-dated ombrotrophic peat cores to reconstruct historical trends in atmospheric deposition had become fairly well established (Shotyk et al., 1997; MacKenzie et al., 1998; Testa et al., 1999). There is strong evidence that lead is immobile in unsaturated ombrotrophic systems but in saturated peat systems, lead may be subject to diagenetic remobilization and thus may have limited use when interpreting the historical record (Urban et al., 1990; MacKenzie et al., 1998). Post depositional mobility of lead was also observed in a study by Johnson et al., (1995) in which it was reported 30 % of the total atmospheric input of Pb was not retained in the organic soils and pond sediments. Friedland et al. (1993) similarly reported mobility of lead in organic rich forest soils in the USA, suggesting that Pb mobility was induced by formation of a soluble organic complex. The processes behind this immobility or mobility are controlled by a number of physicochemical factors. A study by Wang and Benoit (1996) showed that soils with low pH exhibited reduced Pb complexation with both organic and inorganic dissolved ligands, thus decreasing the chance for Pb transport (Wang and Benoit, 1996). This has implications for the present study as the forest soils under investigation have relatively low pH values. Sequential extraction studies observed lead phase dominance in soil in the order: reducible> residual > oxidizable > absorbed, exchangeable and carbonate (AEC) phase (Sutherland and Tack, 2000).

Particle size distribution of radionuclides in soils is another important factor to consider. In general, it is the finer fractions of soil which have the higher concentrations of radionuclides. For example, in a study of ^{232}Th and ^{238}U in soils, the highest activity was observed in the 125-250 µ fraction (Narayana et al., 1995) and ^{137}Cs concentrations were several times higher in the finer fractions (< 8 µm) than those in coarser fractions (He and Walling, 1996).

1.4 - Background to research

There has been little geochemical research done in urban areas to date. As 70% of the population in developed countries live in urban areas (Warner and Harrison, 1993), it is an extremely important, and often overlooked, area of study. The findings from rural settings are often non-transferable to the urban context, as there are many complexities that come with a managed environment.

In a three-month pilot study of radionuclides in the urban environment in July 2000, 14 sites around Glasgow were investigated (Peurou, 2000). Sites were selected close to universities, hospitals and chemistry laboratories and the work focussed on the distribution of ^{137}Cs, ^{7}Be and ^{40}K. The activities of these radionuclides were found to vary considerably from site to site and this formed the basis for the sampling protocol adopted for this current study.

1.5 – Detailed objectives

This thesis presents the results of a study undertaken to investigate natural and anthropogenic radionuclides within the city of Glasgow. The research incorporates a city-wide investigation of radionuclide activities in a range of environmental materials and a more detailed study of radionuclide distributions and inventories in soil cores from a park within the city. The aims of this study were:

- To undertake a preliminary investigation of the distribution, environmental concentrations and variability of radionuclides in the urban environment of Glasgow
- To investigate potential enhancement of radionuclide concentrations in the environment around medical and research establishments
- To investigate the vertical distributions, inventories and geochemical associations of radionuclides in coniferous forest soil, deciduous forest soil and grassland soil in a managed parkland environment

Chapter 2. Sample collection, experimental methods and analysis

This initial investigation of the behaviour and distribution of radionuclides in the urban environment of Glasgow, entailed selection of sites both close to, and remote from possible point sources of radionuclides. Although the selection of the majority of sample sites focused on their location relative to possible point sources the sampling strategy also attempted to achieve a comprehensive geographical spread of sample sites over the city. The sampling strategy included two main sampling programmes in October- November 2000 and April 2001, in which the same sites were re-sampled. Sample locations are given in Table 4. The results from these two sample sets were used in conjunction with those from a pilot study (Peurou, 2000) in order to assess variability of radionuclide activities over both space and time. Small scale spatial variation was also investigated at two sites.

The amalgamation of these three sample sets allows the creation of a tentative baseline for comparison with any possible future enhancement from either a point source or a larger scale release of radionuclides to the environment. This data set may also be used to attempt to establish causes for any variations in background radiation levels.

As an extension to this urban sampling programme, a more detailed investigation of radionuclide distribution and inventories in contrasting soils in a managed parkland environment was carried out. In addition to acting as a reference site for the city-wide sampling programme, Pollok Park provided an ideal setting for this more detailed component of the research which had the specific aims:

- To act as a control site remote from possible point sources of radionuclides.
- To define inventories of radionuclides (Bq m^{-2}) for the urban environment of Glasgow.
- To attempt to derive a more detailed understanding of radionuclide behaviour in three contrasting environments within the park.

Pollok Park is an ideal site for the investigation of radionuclide behaviour in contrasting soil types as there are well established, clearly defined areas of grassland, coniferous woodland and deciduous woodland very close together. The proximity of all the sites is advantageous, as it is reasonable to assume that the amount and type of atmospherically deposited radionuclides are the same at all the sample sites in the park. Thus any differences in the inventories or behaviour of radionuclides can be directly attributed to canopy or vegetation interception, soil type or an environmental factor specific to the area in question.

2.1 – Sample site locations

Site ID	Site locations:	Grid Ref:	Sample type	Analysis laboratory
1	Glasgow University/ Western Infirmary	NS 568 664	3 x grass 3 x bulk soil (0-15 cm) 3 x street dust.	[d]GCSS
2	Gartnavel Hospital	NS 555 684	3 x grass 3 x bulk soil (0-15 cm) 3 x street dust.	GCSS
3	Strathclyde University/ Royal Infirmary	NS 604 658	3 x grass 3 x bulk soil (0-15 cm) 3 x street dust.	GCSS
4	Victoria Infirmary	NS 582 619	[a]5 x grass 5 x bulk soil (0-15 cm) 5 x street dust.	GCSS
5	Southern General Hospital	NS 539 656	3 x grass 3 x bulk soil (0-15 cm) 3 x street dust.	GCSS
6a	Pollok Park Unfertilised grassland	NS 554 621	[b]1 x Grass 1 x bulk soil (0-15 cm) 1 x street dust 1 x 1 cm increment core (0-17cm) 1 x bulk soil (0-2 cm) 1 x 1 cm increment short core (0-7cm)	GCSS [e]SUERC
6b	Pollok Park Fertilised hayfield	NS 559 623	[b]1 x Grass 1 x bulk soil (0-15 cm) 1 x street dust	GCSS

6c	Pollok Park Coniferous forest	NS 556 627	[b]1 x 1 cm increment core (0-20cm) 1 x 1 cm increment short core (0-7cm) 1 x bulk soil (0-2 cm) 1 x bulk soil (2-4 cm) 1 x bulk leaf sample	SUERC
6d	Pollok Park Deciduous forest	NS 556 628	[b]1x 1 cm increment core (0-20cm) 1x 1 cm increment short core (0-7cm) 1 x bulk soil (0-2 cm) 1 x bulk leaf sample 1 x bulk litter sample	SUERC
7	Colston Laboratory, Everard Drive	NS 603 692	[a]6 x grass 6 x bulk soil (0-15 cm) 6 x street dust.	GCSS
8	Summerston, Staffin Drive	NS 568 704	[c]2 x grass 2 x bulk soil (0-15 cm) 2 x street dust.	GCSS
9	Robroyston roundabout	NS 623 684	[c]2 x grass 2 x bulk soil (0-15 cm) 2 x street dust.	GCSS
	River Clyde sample sites:			
10	Dalmarnock Bridge	NS 616 627	3 x 1litre estuarine water	GCSS
11	Kings Bridge, Kings Drive	NS 600 638	3 x 1litre estuarine water	GCSS
12	Scottish Exhibition and Conference Centre	NS 572 652	3 x 1litre estuarine water	GCSS
13	Balmoral Street	NS 527 673	3 x 1litre estuarine water	GCSS
14	Renfrew Ferry	NS 512 686	3 x 1litre estuarine water 2 x 5 litre estuarine water	GCSS

Table 4 – Sample site locations, grid references, sample details and analysis laboratory

[a] Ideally, several samples would have been taken at each site and at each sampling time to assess the local spatial variation in radionuclide concentrations. This, however, was outwith the scope of a one year project, so at each sampling time, one of the sites was chosen for more thorough investigation. In November 2000, four of each sample type were collected, instead of simply one, at the Colston Laboratory site and in April 2001, three of each were collected at the Victoria Infirmary. An average specific activity for each radionuclide from each site was calculated from these replicate samples and used in the main body of the results. The individual values were not used in calculation of the summary statistics as used in the discussion, to avoid the results being biased towards the values at these sites.

[b] Access to the reference site in Pollok Park was not permitted during April due to the foot and mouth outbreak in the UK.

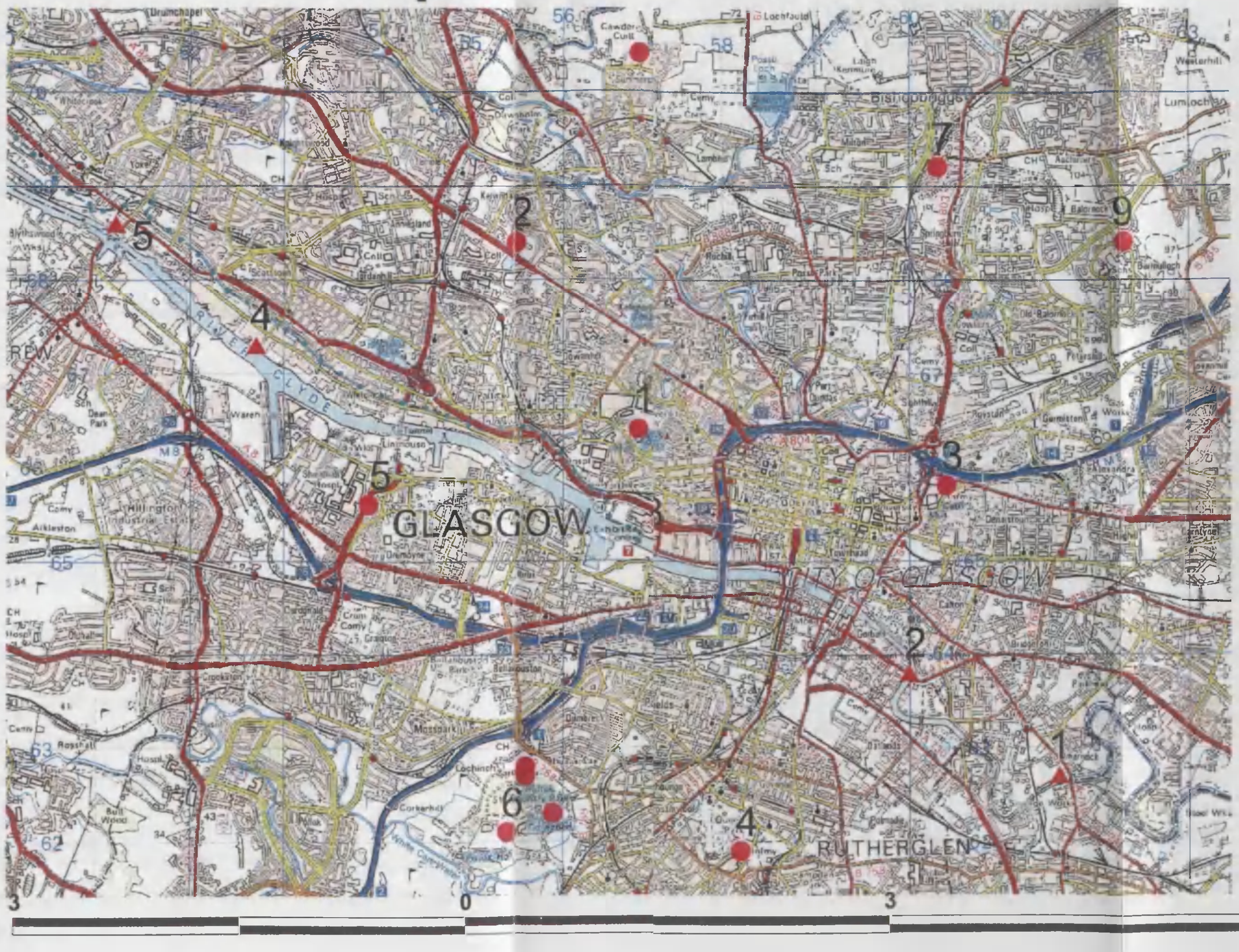
[c] The sites in Summerston and Robroyston were not sampled during the pilot study in July 2000 (Peurou, 2000).

[d] Glasgow City Scientific Services

[e] Scottish Universities Environmental Research Centre

Also see the fold out map (Figure 9) for sample site locations.

Sample site locations



Soil, Street dust and grass sample locations (pink dot)
1 - Glasgow University/ Western Infirmary
2 - Gartnavel Hospital
3 - Strathclyde University / Royal Infirmary
4 - Victoria Infirmary
5- Southern General Hospital
6 - Pollok Park
7 - Colston Laboratory
8 - Summerston
9 - Robroyston

Clyde Estuary water sampling locations (red triangle)
1 - Dalmarnock Bridge
2 - Kings Bridge
3 - SECC
4 - Balmoral Street
5 - Renfrew Ferry

2.2 - Sample collection and initial preparation

2.2.1 - Grass

The sample was collected by cutting the grass close to the ground with shears (avoiding including any soil or roots in the sample) over an area of approximately 2 m^2. Upon return to the laboratory, the fresh weight of the sample was recorded, after which the sample was shredded mechanically, mixed, and a 500 g sub-sample transferred to a 1 litre plastic tub for autoclaving. This sub-sample was autoclaved at 121^0C for 15 minutes at 15 psi. A portion of the autoclaved sample was packed into a pre-weighed 500 ml container, weighed and sealed with tape. The container was labelled and stored pending gamma-ray spectrometry analysis at the Glasgow City Scientific Services Colston Laboratory.

Once the sample had been analysed it was spread out on an oven tray and dried for 24 hours at105 ^{0}C. The weight of the dried sample was recorded and used in calculation of the results in Bq kg^{-1}.

2.2.2 - Soil

(i) Bulk soil samples

The bulk soil samples were collected to a depth of 15 cm using a metal coring implement. The sample was taken from beneath the previously cut grass sampling area and roots were removed from the top of the sample. Enough cores were taken close together and combined at each site to make up a fresh sample weight of approximately 500 g.

Once back in the laboratory, the fresh weight of the sample was recorded using a top pan balance. The sample was spread out on an oven tray lined with foil and roughly mixed, after which it was dried at 105 ^{0}C for 24 hours and the weight of the dried sample was recorded. The dried sample was milled through a 2 mm sieve in a hammer mill. The soil was then mixed thoroughly and packed into a pre-weighed plastic counting container. The weight of the sample used was recorded and the container was sealed pending gamma-ray spectrometry analysis.

(ii) Soil cores and leaves/ leaf litter

Three soil cores were taken in October 2000:

1) A 0-20 cm deciduous (Beech) forest soil core taken in 1 cm increments.
2) A 0-20 cm coniferous (Corsican pine) forest soil core taken in 1 cm increments.
3) A 0-17 cm grassland soil core taken in 1 cm increments.

A hole was dug at each site to expose a soil profile and the 1cm increments were removed individually using a flat metal trowel. The area from which the sample was taken was measured and recorded.

Once back in the laboratory, the soil was dried at 105^{0}C for 24 hours, ground using a mortar and pestle and sieved to < 2 mm. 10g samples of dried, sieved soil were pelleted using an Enerpac hydraulic press (made by Runice Bros). These pellets were then placed in polystyrene weighing pots with lids, weighed accurately and sealed using Araldite glue. This seal provided an impenetrable barrier to ^{222}Rn and thus allowed radioactive equilibrium between ^{226}Ra and ^{222}Rn to be reached. Once this equilibrium had been established (after approximately 21 days) the samples were ready to be analysed by gamma spectrometry.
As access to the park was unavailable in April 2001, due to the foot and mouth outbreak in the UK, additional samples were collected in June 2001 to provide material for pH measurement and for a study of radionuclide geochemical associations using the BCR sequential extraction technique (Rauret et al., 2000). The samples collected in June 2001 were as follows:

1) A grassland soil core (0-7 cm) and a bulk grassland soil sample (0-2 cm)
2) A deciduous soil core (0-7 cm), bulk soil sample (0-2 cm), a leaf litter sample and a sample of deciduous leaves.
3) A coniferous soil core (0-7 cm), bulk soil samples from 0-2 cm and 2-4 cm and a sample of coniferous leaves.
4)

The short cores were dug out intact and taken to the laboratory to be split into 1 cm increments. The leaves were collected from directly above the area where the soil samples were taken. The deciduous litter was scraped off the top of the soil, whilst *in situ*, after the removal of the leaves.

Once back in the laboratory, pH measurements were carried out on the field moist short cores using a multifunction 'Jenway' pH and conductivity meter. The pH in soil is determined by several factors: the amount and type of organic matter, the type of cations present and the clay content of the soil (Fitzpatrick, 1974). Large amounts of organic matter can induce acidity except when counterbalanced by high concentrations of basic cations. The clay content of a soil is of major importance in this context, as a strong neutralising or buffering capacity is provided by the reaction represented below:

$$\text{Clay-}\ O - H \rightarrow \text{Clay-}\ O^- + H^+$$

As these factors will also affect the behaviour of radionuclides, it is valuable to have pH measurements. Redox conditions are also an important variable within soil but measurement of the Eh was out with the scope of this project.

Once the pH measurements were completed on the field moist soil, the samples were oven dried at 105 ^{0}C. Once dried, the soil was ground using a mortar and pestle and passed through a 2 mm sieve. The portion to pass through the sieve was stored in sealed plastic bags pending further analysis.

To gain a greater understanding of the soil characteristics, loss on ignition measurements were made on the 0-20 cm deciduous and coniferous cores and the 0-17 cm grassland core. Furnacing dry soil and determining the loss in weight provides an approximate measure of the soil organic matter content. This was useful in defining an important difference between the soil types. Sub-samples of the dried soil samples were weighed accurately into furnace-proof, ceramic crucibles and heated at 500 ^{0}C for 24 hours. Once cooled, they were re-weighed and the % loss in weight was calculated. In the case of the more organic samples, the temperature of the furnace was increased incrementally in 100 ^{0}C increments to avoid sudden, violent ignition of the sample.

2.2.3 – Street dust

Street dust was collected using a dustpan and brush. The samples were taken from as close to the grass and soil sampling sites as was possible. Upon return to the laboratory the fresh sample was weighed, mixed roughly, spread out on an oven tray and dried at 105 ^{0}C for 24 hours. The dried sample was weighed, ground using a mortar and pestle then sieved (2 mm). The portion that passed through the sieve was weighed and a sub-sample was removed using a 50 ml scoop. This was transferred to a pre-weighed 50 ml plastic counting tub, weighed and stored pending gamma spectrometry analysis.

2.2.4 - Clyde Estuary water

Clyde Estuary water was sampled at 5 locations within the city, twice throughout the year, using a bucket attached to a rope. These one-litre samples were stored in clean, labelled plastic bottles prior to processing and analysis for tritium.

An additional five-litre sample was taken twice at the Renfrew Ferry sampling site (Grid Ref: NS 512 686) for analysis by gamma spectrometry. Once back in the laboratory, these five litre samples were filtered through whatman No: 40 filter papers then filtered again through 0.45 μm cellulose nitrate membrane filters. Once filtered, the sample was placed in a large glass beaker, 100 ml of concentrated nitric acid was added and the sample was mixed. It was then reduced down to a volume of one-litre on a medium temperature hot-plate. This one litre sample was transferred to a pre-weighed one-litre counting geometry and stored pending analysis by gamma spectrometry.

2.3 Analysis techniques

2.3.1 - Gamma Spectrometry analysis

Gamma spectrometry analysis was carried out both at the Glasgow City Scientific Services (GCSS) laboratory in Colston and at the Scottish Universities Environmental Research Centre (SUERC). Both laboratories had slightly different approaches to the technique but in both cases, the samples were analysed using shielded high-resolution germanium semiconductor detectors. Energy calibration is an essential requirement for gamma spectroscopy as it is the exact energy that identifies radionuclides on the basis of the photopeaks in the spectrum. This energy calibration is achieved by measuring standards of mixed sources of known radionuclides with well-defined energies in the range of interest. These standards are also used to determine the counting efficiency as a function of energy. This is dependent on the energy of the gamma-emitter, the source geometry and on self-absorption in the source (Debertin and Helmer, 1998).

Three different detectors at GCSS were used in this research:

1) Ortec Gem series HPGe coaxial detector (model GEM-25185-p)
2) Ortec GMX series HPGe coaxial detector (model GMX-18190)
3) Canberra germanium detector (model GC3518-7500SL)

Each detector is shielded by a graded Pb-Cd-Cu shield and all have been calibrated using standards of a range of sample types spiked with known activities of a mixed radionuclide standard solution supplied by the National Physics Laboratory. Background counts were recorded for each detector and were subtracted from sample spectra as appropriate. To achieve good counting statistics, the number of counts in the time interval chosen must be significantly larger than the number of background counts in that same time interval (Debertin and Helmer, 1998). To this effect, the samples were counted for 60 000 seconds each, with the exception of the 1 litre estuarine samples, which were counted for 200 000 seconds . Spectral analysis was performed using FitzPeaks 32 gamma analysis and calibration software version 3.36.

The gamma spectroscopy analysis at SUERC was carried out using a low background Canberra low energy germanium (LEGe) detector with a graded Pb-Cd-Cu shield. Gamma spectrometry analysis of the samples was carried out using a three and a half day count time for each sample. The sample pots were placed directly on the end cap of the detector, which itself was covered by a cling-film cover. The peak analysis programme used was the EG&G Ortec package, Gamma Vision.

Significant variations in self absorption of gamma radiation (in particular at low energy) occur in response to variations in the organic content of the samples. As the samples to be analysed at SUERC had variable and often high organic contents, it was necessary to prepare a set of standards that matched the chemical composition of the samples. In order to do this, standards of mass 10g, spiked with appropriate activities of ^{241}Am, ^{226}Ra, ^{210}Pb and ^{137}Cs, were made with 7% organic matter, 20% organic matter and 55% organic matter. Gamma spectra of the standards were recorded and the % detection efficiency calculated from:

$$\%\ \text{detection efficiency} = \frac{\text{counts per second from standard}}{\text{decay corrected activity (Bq) of standard}} \quad \text{x} \quad \frac{100}{1}\ \%$$

The ^{226}Ra specific activity was calculated on the basis of the ^{214}Pb spectra peaks at 295 keV and 352 keV and the total ^{210}Pb specific activity was calculated from the 46.5 keV peak. The unsupported ^{210}Pb activity was then calculated by subtracting the ^{226}Ra specific activity from the total ^{210}Pb specific activity. The detectors at GCSS were calibrated between 50 and 2000 keV and therefore, as the principal ^{210}Pb gamma peak occurs at 46.5 keV, reliable ^{210}Pb results were not achievable.

In both laboratories, corrections were made for the background count rate by subtracting an experimentally established background rate. The detection limit will depend on the sample composition, the energy of the radiation, the source-detector distance, the detector efficiency, the background and the available time for measurement. Potential sources of error include improper spectral analysis, changes in background, errors in calibration and geometry and lack of homogeneity in samples.

2.3.2 Plutonium analysis

The oven-dried soil was weighed into heatproof, ceramic crucibles and furnaced overnight at 500°C to destroy the organic matter. Once cooled, the sample was washed into a large beaker using reverse osmosis water (ROW) and the samples were dampened with a few drops of 9 mol l^{-1} HCl. Once effervescence had ceased, the samples were spiked with 1 ml of ^{242}Pu with an activity of 0.0073 Bq ml^{-1} using an accurate pipette. At this stage, a few drops of 10 mg ml^{-1} Fe^{3+} carrier solution were added. Approximately 300 ml of 9 mol l^{-1} HCl were then added, the beakers were placed on a hot plate at medium temperature (100-200^{0}C) and taken to near dryness.

Up to 50 ml of hydrogen peroxide were carefully added, as required, to break down any residual organic material and, once any vigorous reaction had died down, the sample was again taken to near dryness. 100 ml of aqua regia were added and, again, the solution was taken almost to dryness. Approximately 200 ml of 9 mol l^{-1} HCl were mixed well with the sample, heated gently then left to stand overnight.

Each sample was transferred to a centrifuge bottle, was washed in with 9 mol l^{-1} HCl, and centrifuged for 10 minutes at 3500 rpm. The solution was decanted and the supernatant saved. 9 mol l^{-1} HCl was added to the residual solid in the centrifuge tubes after which the tubes were shaken and then centrifuged once again. This was repeated, ensuring the supernatant was clear then repeated again with ROW. This volume of supernatant was reduced to approximately 500 ml (if required) and ammonia was added until a brown $Fe(OH)_3$ precipitate formed. The sample was mixed well and left to settle out, after which it was again centrifuged as above, but this time the supernatant was discarded. The precipitate in the centrifuge bottles was washed with ROW until all the ammonia was removed. The precipitate was then re-dissolved in 9 mol l^{-1} HCl. As Fe^{3+} behaves in a similar way to plutonium, it was necessary to remove this using a Di-isopropyl Ether (DIPE) solvent extraction. This extraction was carried out in solvent extraction flasks until the iron had been visibly removed (until the solution was colourless). The ether layer was discarded and the aqueous layer was saved. This solution was taken almost to dryness and up to 10 ml of 15 mol l^{-1} HNO_3 were added then taken to near dryness again. This step was repeated three times to ensure oxidation of the plutonium from the +3 to the +4 state. The sample was then taken up in concentrated HCl and reduced to near dryness prior to dissolution in 9 mol l^{-1} HCl for ion exchange separation of plutonium using a method based upon that of Wong (1971).

The next stage involved separation using a 6 cm x 1 cm^2 column of Biorad AG1-x8 anion exchange resin preconditioned with 1.2 mol l^{-1} HCl and 9 mol l^{-1} HCl (approximately 50 ml of each). The sample was passed through the column in 9M HCl and rinsed with approximately 100 ml of 9 mol l^{-1} HCl. This rinsing was to ensure that the entire sample was washed off the sides of the glassware. In 9 mol l^{-1} HCl all the plutonium is held on the resin in the forms $(PuCl_5)^-$ and $(PuCl_6)^{2-}$. Iron and uranium are also held on the column under these conditions but other interfering substances such as radium, thorium and americium pass through the column. The plutonium was eluted, into a clean beaker, with 6 batches of 20 ml 9 mol l^{-1} HCl mixed with 5ml 1 mol l^{-1} NH_4I. This elution involves the reduction of the plutonium by I^- to the 3^+ oxidation state, which cannot form anionic complexes with the HCl and is therefore not held by the resin. Once through the column, the eluting solution was taken to dryness on the hotplate and concentrated HNO_3 was added to remove the iodine by oxidation. This was repeated until all traces of iodine had been removed and no brown gas was given off. This is important as any traces of I^- left behind can result in a low plutonium yield. The solution was then transferred by pipette into a clean beaker, taking care to wash only the base of beaker and not the sides where there may have been residual iodine. A few ml of concentrated HNO_3 were added, the sample was taken to dryness then this was repeated once. The sample was dissolved in 8 mol l^{-1} HNO_3 prior to the second stage of the anion exchange separation. The second column was preconditioned with 50 ml 1.2 mol l^{-1} HCl then with approximately 50 ml of 8 mol l^{-1} HNO_3. The sample was passed through in 8 mol l^{-1} HNO_3 and the column sides were rinsed, using a clean pipette each time, with up to 100 ml of 8 mol l^{-1} HNO_3, then with a few millilitres of 9 mol

l^{-1} HCl. This time the plutonium is held on the column but the iron and uranium pass through. The plutonium was eluted into a clean beaker using 6 batches of 20 ml 9 mol l^{-1} HCl mixed with 5ml 1 mol l^{-1} NH_4I. Iodine was removed by evaporation and addition of concentrated HNO_3 (Wong, 1971).

The samples were transferred by pipette to clean beakers, washing in with HNO_3. Once this solution was taken to near dryness, the plutonium isotopes were re-dissolved in approximately 4 drops of concentrated HCl. To prepare a thin source of plutonium on a 2.5 cm diameter stainless steel planchette, the solution was transferred to an electrodeposition cell in approximately 40 ml of 3.75% (w/V) ammonium chloride plating solution. The cell consists of a Perspex cylindrical body with a platinum wire anode in the centre. The perspex cell is screwed onto a copper base and a stainless steel planchette, on which the plutonium is deposited, acts as the cathode. This planchette is firmly fixed in the centre of the base and sealed from the plating solution by a Teflon coated rubber-o-ring fitted between the planchette and the cell body. A locating pin is fitted in the copper base for electrical contact to the power supply. The copper base of the cell acts as a sink for the heat produced during the plating. The samples were electrodeposited onto the stainless steel planchette for one hour at a current of 3 Amps and a potential of 15 Volts (Hallberg and Brise, 1960).

Immediately before the end of the plating, 2 ml of ammonia were added to raise the pH of the solution and prevent re-dissolution of the electrodeposited plutonium once the voltage was switched off. The planchette was then removed, rinsed using ROW and air dried prior to alpha spectroscopy analysis.

2.3.3 - Alpha spectroscopy

Alpha spectrometry was performed using Canberra passivated ion implanted planar Si detectors in a Canberra Quad Alpha spectroscopy system. The planchettes were counted for between one week and three weeks in order to obtain suitable counting statistics.

Alpha spectroscopy must be performed under vacuum and usually has a low background. However, the background count was measured periodically through the counting period and subtracted from the sample count rate.

The activity of the nuclide of interest in the sample is calculated from:

$$\frac{\text{Asample}}{\text{Aspike}} = \frac{\text{Csample}}{\text{Cspike}}$$

Where **A**sample and **A**spike are the respective activities of the isotope of interest in the sample and the spike, and **C**sample and **C**spike are the corresponding count rates. The specific activity of the isotope of interest is then calculated by dividing its activity by the mass of the sample.

Error calculations

Net counts = **counts**(sample)- **counts**(background)

1σ net counts = $[(\text{counts sample or spike}) + (\text{counts background})]^{1/2}$

The certified uncertainty on the spike is 1%, and so the total % error (S) involved in the measurement is:

$$S_{\text{sample}} = \sqrt{[(S_{\text{counts sample}})^2 + (S_{\text{counts spike}})^2 + (S_{\text{certified uncertainty of spike}})^2]}$$

2.3.4 - BCR sequential extraction

The BCR sequential extraction scheme was developed to act as a standard method when aiming to determine the geochemical association pattern of metals within a soil or sediment. There are three extraction steps, with a different phase of the soil being targeted at each stage (Rauret et al., 2000). These phases along with the reagents used are listed below:

(i) Reagent	**Target phase**
Acetic acid	Exchangeable
Hydroxylammonium Chloride	Reducible
Hydrogen peroxide/ Ammonium acetate	Oxidizeable

Formerly the reducible phase was often designated as the Fe/ Mn oxyhydroxides and the oxidizeable phase designated as organic. In practice however, extractants do not specifically and selectively dissolve these phases, so the terminology was broadened to the more general terms of reducible and oxidizeable.

The extracting solutions were prepared as follows:

- 0.11 mol l^{-1} Acetic acid

6.25 ml of redistilled glacial acetic acid was accurately measured using a pipette and transferred to a 1 litre volumetric flask. The solution was made up to volume with deionised water.

- 0.1 mol l^{-1} Hydroxylammonium Chloride (~ pH 2)

13.893g of hydroxylammonium chloride were weighed and dissolved in approximately 1800 ml of deionised water. Nitric acid was added as required to bring the pH to approximately 2. The solution was then made up to exactly 2 litres with deionised water.

- 8.8 mol l^{-1} Hydrogen Peroxide

This was available as 30% stock solution. The solution, which is stored in a refrigerator, was allowed to come up to room temperature before it was used.

- 1 mol l^{-1} Ammonium Acetate

77.08 g of ammonium acetate were weighed and dissolved in approximately 900 ml deionised water. The solution was adjusted to approximately pH 2 with nitric acid. The solution was made up to volume with deionised water in a 1 litre volumetric flask.

Note: The solutions were made up immediately prior to use.

(ii) Extraction procedure

Step one – The exchangeable fraction

Approximately 1g of each sample was accurately weighed into 100 ml polypropylene centrifuge bottles. It was decided that this was the best geometry to use for consistency in the experiment but in order to provide sufficient weight of samples for plutonium analysis, each sample had eight 1g sub-samples weighed into separate centrifuge bottles. Eight blank centrifuge bottles were included in the experiment. 40 ml acetic acid were added to each bottle and the bottles were shaken for 8 hours on an end-over-end mechanical shaker. The extracts were then separated from the residue by centrifugation at 4000 rpm for 10 minutes. The extracts were then filtered into bottles for extraction of plutonium. At this stage, all of the 8 sub-samples from each sample were filtered into the same bottle for plutonium extraction as a single sample.

Every effort was made not to lose any of the residues when separating the liquid extract but, particularly with the lighter materials like deciduous leaves, it was very difficult not to lose some. Different centrifuge speeds and times were tested in an attempt to reduce the amount of solid residue left floating on the liquid after centrifugation. Whilst improvements were noted, this feature would have to be addressed if the experiment was repeated.

Step two – Reducible fraction

40 ml hydroxylammonium chloride (pH 2) were added to the residue from step one in the

centrifuge bottles. The same shaking and separation of the extractant from the sample procedure was carried out, as in step one.

Step three – Oxidizeable

10 ml hydrogen peroxide were added to the residue from step 2 and the samples were allowed to stand for an hour at room temperature with an occasional manual agitation. The centrifuge bottles were then gradually heated in a water bath to 85°C, inside a fume cupboard, until the contents had reduced to approximately 1-2 ml. A second aliquot of hydrogen peroxide was added and allowed to evaporate in the same way. 50 ml ammonium acetate were added to the moist residue and shaken for 2 hours. The bottles were centrifuged and the extracts separated as described above (Arunachalam et al., 1996; Rauret et al., 2000).

(iii) Plutonium isolation from the sequential extracts

The extraction was carried out in approximately the same way as described above (section 2.3.2) with the only differences occurring towards the beginning of the procedure. No furnacing or centrifuging of the sample was required and many of the stages were carried out simply as a precaution and smaller volumes of reagent were used e.g. the hydrogen peroxide stage. A blank from each stage of the sequential extraction was included in the plutonium analysis. The samples were plated after the extraction and analysed using alpha spectrometry.

2.3.5 - Determination of Tritium (^3H) in estuarine water samples by liquid scintillation counting

In liquid scintillation, the water sample was added to a scintillation 'cocktail' containing primary and secondary fluors (sometimes called scintillants), aromatic solvent and adjuvants (mainly surfactants). The emitted β^- particles from the sample transfer energy to the solvent present in the cocktail. The solvent then transfers energy to the scintillator causing promotion of electrons to excited levels followed by instantaneous de-excitation, which results in emission of light. This process is proportional, so the quantity of light emitted is proportional to the radiation energy (Packard, 1990).

The procedure in the GCSS laboratory is designed to quantify any ^3H contamination above the natural levels in fresh water in central Scotland. To achieve this, correction for the ambient ^3H level was achieved by including a deionised water 'reference' sample in the same preparation and analysis procedure as with the estuarine samples.

In order to prepare the samples for liquid scintillation counting, 100 ml of each water sample were measured into a labelled 250 ml round bottomed flask and several anti-bumping chips were added. 1 g of potassium permanganate was added to each flask to oxidise organic matter. The clean and dry distillation apparatus was assembled with another labelled 250 ml flask at the end of the system. The sample was distilled until near dryness under vacuum. The distillation unit heater was then switched off, the vacuum released and the system was left to cool.

The second flask, previously from the end of the system was then placed on the heating mantle with several anti-bumping chips added. The cleaned distillation apparatus was then reassembled but with a 150 ml round bottomed flask to end the system. The sample was then redistilled until near dryness. The second distillate was, in preparation for counting, mixed in a 1:1 ratio with the scintillation cocktail: 10 ml of sample were pipetted into a polythene vial with 10 ml of Pico-flour low-level tritium scintillation cocktail also being added. The lid of the vial was securely fastened and the vial was shaken vigorously.

The vials were then transferred to a Packard 1900TR liquid scintillation analyser and left to cool and dark adapt overnight. The next morning, once the samples were chilled, the analysis was started.

Efficiency

The efficiency of counting of samples by liquid scintillation may be reduced by a number of factors:

- Photon Quenching – incomplete transfer of beta particle energy to solvent molecules.
- Chemical Quenching – impurities in the prepared sample can cause energy loses in the transfer from solute to solvent.
- Optical Quenching – causes the attenuation of photons produced in the solution.

The results of the energy losses are collectively referred to as Quenching. Because of these energy losses, quenching causes an apparent shift in the energy spectrum to the lower energy region, with a corresponding reduction in the counting efficiency. This effect can be corrected for by the preparation of a quench curve by plotting a Quench Indication Parameter (QIP) against the efficiency of detection of the selected radionuclide, in this case tritium. For the method used in the determination of tritium, the Transformed Spectral Index (tSIE) of the external standard spectrum, is calculated from the Compton spectrum produced in the sample by the external standard source (Ba-133), and is used as the QIP (Packard, 1990).

A quench curve was prepared by adding small amounts of nitromethane as quenching agent to known activities of tritiated water and scintillation cocktail. Each of these samples were counted for a known time and the instrument software calculated the efficiency of counting for each sample and prepared a quench curve under a pre-defined protocol which is then stored in the instruments memory (Appendix 2). Samples which are counted using this protocol are automatically corrected for any quench effects using this quench correction curve from the instruments memory.

Calculations

The quench corrected results are given in disintegrations per minute with a 95% confidence counting error. This result can be converted to Bq l^{-1} by: -

$$\text{Bq } l^{-1} = (\text{dpm}/60) \times (1000/\text{volume used})$$

Chapter 3 - Results

The soil pH and loss on ignition results for the Pollok Park soils can be found in Tables **5-10**.

The gamma spectrometry results from the Pollok Park cores, including total ^{210}Pb, unsupported ^{210}Pb, estimated ^{210}Pb inventories, ^{137}Cs and ^{226}Ra can be found in Tables **11-16**.

The Pollok park unfertilised grassland and fertilised hay field sites have been treated separately for comparison of ^{40}K specific activities only, as for every other nuclide they had very similar specific activities and therefore there was limited value in treating them separately. For every nuclide other than ^{40}K, the average of the two sites has been taken. The gamma spectrometry results obtained at the GCSS laboratory for bulk soil, street dust and grass are presented below in Tables **17-20**. Wherever possible, 1 σ errors have been included in the results Tables. Extended results from the analysis carried out at GCSS are in **Appendix 1**. This includes data for other nuclides not presented in the main body of the results. The individual values for the replicated samples taken at the Colston Laboratory in November 2000 and the Victoria Infirmary in April 2001 are shown below in Tables **21-23**. The results from the gamma spectrometry analysis carried out on the estuarine water are shown in Table **24**.

The alpha spectrometry plutonium analysis results are given in Tables **25-27** and the plutonium analysis results from the Pollok Park bulk soil samples are shown in Table **28**. The results from the BCR sequential extraction experiment are shown in Table **29** and the tritium results from analysis of the Clyde estuarine samples are shown in Table **30**.

3.1 - Loss on Ignition and pH

Depth (cm)	% Loss on Ignition
0-1	38.57
1-2	24.88
2-3	17.28
3-4	14.61
4-5	11.67
5-6	10.95
6-7	9.59
7-8	7.83
8-9	8.06
9-10	7.28
10-11	7.16
11-12	7.23
12-13	6.93
13-14	6.11
14-15	5.92
15-16	5.92
16-17	5.79
17-18	5.29
18-19	5.78
19-20	5.55

Table 5 – Loss on ignition data for Pollok Park deciduous core (October 2000)

Depth (cm)	% Loss on Ignition
0-1	ND
1-2	57.47
2-3	51.92
3-4	53.87
4-5	57.85
5-7	35.35
7-8	25.94
8-9	27.26
9-10	21.41
10-11	18.77
11-12	22.65
12-13	15.65
13-14	14.24
14-15	12.26
15-16	11.62
16-17	10.68
17-18	10.12
18-19	9.98
19-20	9.42

ND = No data available, in this case due to lack of available sample.

Table 6 - Loss on ignition data for Pollok Park coniferous core (October 2000)

Note: The 1 cm increments from 5-6 cm and 6-7 cm from the coniferous soil core were accidentally mixed at an early stage in the sample preparation and so are treated as one 2 cm increment from 5-7 cm.

Depth (cm)	% Loss on Ignition
0-1	ND
1-2	ND
2-3	18.54
3-4	17.32
4-5	14.27
5-6	13.87
6-7	14.78
7-8	12.22
8-9	11.43
9-10	11.48
10-11	10.34
11-12	10.8
12-13	14.06
13-14	12.93
14-15	12.79
15-17	9.55

Table 7 - Loss on ignition data for Pollok Park Grassland core (October 2000)

Note: The grassland core was only taken to a depth of 17 cm to minimise damage to the sample area. The lack of data available for increments 0-1cm and 1-2 cm is due to there being a lack of available sample after the removal of the roots from the surface.

pH

Depth (cm)	pH
0-1	3.55
1-2	3.67
2-3	3.64
3-4	3.62
4-5	3.60
5-6	3.51
6-7	3.68

Table 8 – pH data for Pollok Park deciduous short core (June 2001)

Deciduous Litter = pH 3.52
Deciduous bulk 0-2 cm = pH 3.77
Deciduous leaves = pH 4.2

Depth (cm)	pH
0-1	3.57
1-2	3.42
2-3	3.36
3-4	3.30
4-5	3.42
5-6	3.29
6-7	3.30
7-8	3.32

Table 9 - pH data for Pollok Park coniferous short core (June 2001)

Coniferous Bulk 0-2 cm = pH 3.4
Coniferous bulk 2-4 cm = pH 3.31

Coniferous leaves = pH 4.55

Depth (cm)	pH
0-1	6.00
1-2	5.46
2-3	5.71
3-4	5.43
4-5	5.50
5-6	5.52
6-7	5.57

Table 10 - pH data for Pollok Park grassland short core (June 2001)

Grass bulk 0-2cm = pH 5.83

3.2 - Gamma spectrometry results

Sample depth (cm)	Total ^{210}Pb	^{137}Cs	^{241}Am	^{226}Ra
0-1	120 ± 4	387 ± 6	1.4 ± 0.4	26 ± 3
1-2	91 ± 4	281 ± 4	2.4 ± 0.4	26 ± 3
2-3	53 ± 6	125 ± 4	1.9 ± 0.0	27 ± 4
3-4	39± 5	70 ± 4	BDL	23 ± 4
4-5	34 ± 6	36 ± 3	BDL	23 ± 4
5-6	27 ± 4	14 ± 2	BDL	22 ± 3
6-7	27 ± 3	11 ± 1	BDL	23 ± 2
7-8	29 ± 3	BDL	BDL	24 ± 2
8-9	33 ± 4	BDL	BDL	27 ± 3
9-10	31 ± 4	BDL	BDL	26 ± 3
10-11	26 ± 3	BDL	BDL	26 ± 2
11-12	25 ± 3	BDL	BDL	29 ± 3
12-13	32 ± 4	BDL	BDL	33 ± 3
13-14	30± 4	BDL	BDL	32 ± 3
14-15	22 ± 4	BDL	BDL	32 ± 4
15-16	34 ± 7	BDL	BDL	31 ± 6
16-17	21 ± 3	BDL	BDL	27 ± 5
17-18	BDL	BDL	BDL	BDL
18-19	BDL	BDL	BDL	BDL
19-20	BDL	BDL	BDL	BDL

Table 11 – Radionuclide specific activities (Bq kg^{-1}) for the Pollok Park deciduous forest soil core (October 2000)

Sample depth (cm)	Unsupported ^{210}Pb (Bq kg^{-1})	ln unsupported ^{210}Pb	Total dry weight of section (g)
0-1	95 ± 5	4.6 ± 0.6	48.50
1-2	65 ± 5	4.2 ± 0.1	53.08
2-3	26 ± 7	3.3 ± 0.3	58.80
3-4	16 ± 6	2.8 ± 0.5	59.95
4-5	10 ± 6	2.3 ± 0.7	62.18
5-6	BDL	BDL	63.62
6-7	BDL	BDL	63.65
7-8	BDL	BDL	64.32
8-9	BDL	BDL	65.35
9-10	BDL	BDL	65.41
10-11	BDL	BDL	65.51
11-12	BDL	BDL	66.53
12-13	BDL	BDL	65.69
13-14	BDL	BDL	65.95
14-15	BDL	BDL	67.24
15-16	BDL	BDL	66.04
16-17	BDL	BDL	66.58
17-18	BDL	BDL	67.08
18-19	BDL	BDL	67.15
19-20	BDL	BDL	66.15

Table 12 - Unsupported ^{210}Pb (Bq kg^{-1}) for the Pollok Park deciduous forest soil core (October 2000)

The inventory (in Bq cm^{-2}) is the sum of the activities for each section divided by the area of the core in cm^{2} (10 x 10 cm^{2}). This is then multiplied by 10 000 to give the result in Bq m^{-2}.

Inventory = 1116.75 Bq m^{-2} **Deciduous ^{210}Pb Inventory = 1117 Bq m^{-2}**

Flux = (ln 2/half life ^{210}Pb) x Inventory $\therefore$ Flux = 0.0301 x 1117 = 33.6

Deciduous ^{210}Pb Flux = 34 Bq m^{-2} y^{-1}

Sample depth (cm)	Total ^{210}Pb	^{137}Cs	^{241}Am	^{226}Ra
0-1	232 ± 5	111 ± 3	BDL	6 ± 2
1-2	133 ± 5	347 ± 6	2 ± 0.5	22 ± 4
2-3	182 ± 7	367 ± 6	BDL	17 ± 5
3-4	138 ± 4	243 ± 4	3 ± 0.4	21 ± 3
4-5	131 ± 5	62 ± 1	2 ± 0.5	18 ± 3
5-7	65 ± 4	108 ± 3	BDL	24 ± 4
7-8	52 ± 4	70 ± 3	BDL	28 ± 5
8-9	56 ± 4	BDL	BDL	26 ± 4
9-10	47 ± 3	49 ± 2	BDL	27 ± 4
10-11	30 ± 6	36 ± 0.5	BDL	26 ± 5
11-12	33 ± 4	53 ± 2	BDL	24 ± 3
12-13	36 ± 5	31 ± 3	BDL	26 ± 6
13-14	35 ± 5	33 ± 3	BDL	28 ± 5
14-15	29 ± 7	21 ± 3	BDL	27 ± 5
15-16	38 ± 7	BDL	BDL	51 ± 11
16-17	34 ± 7	BDL	BDL	38 ± 7
17-18	BDL	BDL	BDL	BDL
18-19	BDL	BDL	BDL	BDL
19-20	BDL	BDL	BDL	BDL

Table 13 – Radionuclide specific activities (Bq kg^{-1}) for the Pollok Park coniferous forest soil core (October 2000)

Sample depth (cm)	Unsupported ^{210}Pb (Bqkg^{-1})	ln excess ^{210}Pb (Bqkg^{-1})	Total dry weight of section (g)
0-1	220 ± 6	5.4 ± 0.03	39.08
1-2	107 ± 6	4.7 ± 0.05	44.14
2-3	163 ± 8	5.1 ± 0.06	46.90
3-4	114 ± 5	4.7 ± 0.05	46.09
4-5	116 ± 6	4.8 ± 0.08	44.76
5-7	40 ± 6	3.7 ± 0.15	101.9
7-8	24± 7	3.2 ± 0.30	53.77
8-9	30 ± 6	3.4 ± 0.21	55.18
9-10	21 ± 5	3.0 ± 0.26	58.31
10-11	BDL	BDL	58.40
11-12	BDL	BDL	56.45
12-13	BDL	BDL	59.53
13-14	BDL	BDL	61.28
14-15	BDL	BDL	61.24
15-16	BDL	BDL	62.03
16-17	BDL	BDL	62.08
17-18	BDL	BDL	61.63
18-19	BDL	BDL	63.01
19-20	BDL	BDL	64.09

Table 14 – Unsupported ^{210}Pb (Bq kg^{-1}) for the Pollok Park coniferous forest soil core (October 2000)

(Area of core = 10 x 10 cm)

Coniferous ^{210}Pb Inventory = 3966 Bq m^{-2}
Flux = (ln 2/halflife ^{210}Pb) x Inventory
Flux = 0.0301 x 3966 = 119.4

Coniferous ^{210}Pb Flux = 119 Bq m^{-2} y^{-1}

Sample depth (cm)	Total ^{210}Pb	^{137}Cs	^{241}Am	^{226}Ra
0-1	92 ± 11	28 ± 5	BDL	ND
1-2	83 ± 4	81 ± 2	BDL	39 ± 3
2-3	56 ± 4	84 ± 3	BDL	37 ± 3
3-4	54 ± 3	85 ± 3	BDL	36 ± 3
4-5	53 ± 4	86 ± 3	BDL	35 ± 3
5-6	51 ± 4	99 ± 3	BDL	42 ± 3
6-7	62 ± 5	83 ± 3	BDL	37 ± 4
7-8	52 ± 5	72 ± 3	BDL	36 ± 3
8-9	48 ± 4	53 ± 3	BDL	37 ± 3
9-10	40 ± 6	43 ± 2	BDL	41± 3
10-11	48 ± 6	29 ± 2	BDL	43 ± 3
11-12	45 ± 5	25 ± 3	BDL	41 ± 4
12-13	47 ± 4	18 ± 2	BDL	39 ± 3
13-14	43 ± 5	18 ± 2	BDL	44 ± 4
14-15	37 ± 7	14 ± 3	BDL	44 ± 6
15-17	30 ± 5	BDL	BDL	47 ± 5

Table 15 – Radionuclide specific activities (Bq kg^{-1}) for the Pollok Park grassland soil core (October 2000)

Sample depth (cm)	Unsupported ^{210}Pb (Bqkg^{-1})	ln excess ^{210}Pb (Bqkg^{-1})	Total dry weight of section (g)
0-1	92 ± 11	4.5 ± 0.13	67.69
1-2	44 ± 5	3.8 ± 0.11	42.75
2-3	19 ± 5	3.0 ± 0.27	91.00
3-4	18 ± 4	2.9 ± 0.26	53.83
4-5	18 ± 5	2.9 ± 0.29	54.59
5-6	9 ± 5	2.2 ± 0.68	54.89
6-7	25 ± 7	3.2 ± 0.28	59.06
7-8	17 ± 6	2.8 ± 0.38	68.88
8-9	12 ± 5	2.4 ± 0.50	67.00
9-10	BDL	BDL	65.33
10-11	BDL	BDL	93.80
11-12	BDL	BDL	70.02
12-13	BDL	BDL	117.98
13-14	BDL	BDL	117.08
14-15	BDL	BDL	71.99
15-17	BDL	BDL	88.72

Table 16 – Unsupported ^{210}Pb (Bq kg^{-1}) for the Pollok Park grassland soil core (October 2000)

(Area of core = 10 x 10 cm)

Grassland ^{210}Pb Inventory = 1573 Bq m^{-2}

Flux = (ln 2/halflife ^{210}Pb) x Inventory
Flux = 0.0301 x 1573 = 47.4

Grassland ^{210}Pb Flux = 47 Bq m^{-2} y^{-1}

Sample site	Street dust July 2000	Street dust November 2000	Street dust April 2001	Grass July 2000	Grass November 2000	Grass April 2001	Soil July 2000	Soil November 2000	Soil April 2001
1 (Glasgow Uni/ Western)	27.3 ± 3.2	14.0 ± 1.9	46.7 ± 3.1	5.3 ± 2.3	22.5 ± 2.5	17.4 ± 1.7	58.8 ± 3.5	38.5 ± 2.4	30.3 ± 1.3
2 (Gartnavel Hosp)	25.7 ± 2.7	3.9 ± 1.7	7.5 ± 1.9	<4.1	1.2 ± 0.4	2.6 ± 0.6	26.3 ± 1.7	34.3 ± 2.1	46.3 ± 1.9
3 (Strathclyde/ Royal)	11.4 ± 1.7	BDL	2.1 ± 0.9	<4.05	6.6 ± 2.4	1.7 ± 0.7	19.1 ± 1.3	30.7 ± 1.9	22.1 ± 0.9
4 (Victoria Inf)	63.4 ± 6.0	48.5 ± 3.9	16.7 ± 2.9	<2.7	11.7 ± 2.5	3.9 ± 0.8	56.9 ± 3.4	64.9 ± 4.3	65.4 ± 2.5
5 (Southern General Hosp)	205 ± 13	6.9 ± 2.0	31.7 ± 2.4	<3.2	15.8 ± 2.2	<1.4	17.9 ± 1.2	32.3 ± 2.0	85.1 ± 3.1
6 (Pollok Park)	ND	24.8 ± 2.7	ND	ND	BDL	ND	ND	38.2 ± 2.4	ND
7 (Colston Laboratory)	23.4 ± 2.6	211 ± 12	29.6 ± 2.6	5.0 ± 0.9	3.5 ± 1.4	7.4 ± 1.3	22.4 ± 1.2	34.8 ± 2.2	2.4 ± 0.5
8 (Summerston)	ND	5.7 ± 1.3	4.2 ± 1.1	ND	BDL	5.1 ± 1.4	ND	32.1 ± 2.0	38.4 ± 1.5
9 (Robroyston)	ND	5.7 ± 2.3	2.3 ± 0.47	ND	BDL	2.6 ± 1.2	ND	4.3 ± 0.4	5.8 ± 0.5

Table 17 - ^{137}Cs (Bq kg^{-1}) measured in street dust, grass (dry weight) and bulk soil in July 2000, November 2000 and April 2001. ND = No data available, BDL = below detection limit

Sample site	Street dust July 2000	Street dust November 2000	Street dust April 2001	Grass July 2000	Grass November 2000	Grass April 2001	Soil July 2000	Soil November 2000	Soil April 2001
1 (Glasgow Uni/ Western)	668 ± 60	933 ± 69	337 ± 40	1605 ± 124	1060 ± 76	574 ± 46	365 ± 24	364 ± 25	295 ± 14
2 (Gartnavel Hosp)	1000 ± 75	575 ± 55	189 ± 32	1130 ± 110	197 ± 17	313 ± 24	369 ± 24	286 ± 19	262 ± 16
3 (Strathclyde/ Royal)	891 ± 66	552 ± 52	295 ± 31	2115 ± 155	674 ± 83	336 ± 30	335 ± 22	343 ± 22	374 ± 18
4 (Victoria Inf)	760 ± 83	526 ± 48	649 ± 72	1370 ± 98	1480 ± 120	470 ± 34	355 ± 23	225 ± 18	253 ± 13
5 (Southern General Hosp)	820 ± 60	363 ± 57	369 ± 39	853 ± 68	1160 ± 86	252 ± 29	384 ± 25	345 ± 24	327 ± 16
6a (Pollok- Unfertilised)	ND	279 ± 47	ND	ND	BDL	ND	ND	257 ± 19	ND
6b (Pollok- fertilised)	ND	652 ± 54	ND	ND	1910 ± 150	ND	ND	255 ± 18	ND
7 (Colston Laboratory)	600 ± 55	364 ± 53	299 ± 39	507 ± 31	616 ± 42	553 ± 52	267 ± 18	275 ±19	238 ± 14
8 (Summerston)	ND	494 ± 43	261 ± 34	ND	710 ± 40	710 ± 54	ND	318 ± 21	300 ± 15
9 (Robroyston)	ND	286 ± 66	786 ± 45	ND	664 ± 61	497 ± 46	ND	413 ± 27	479 ± 23

Table 18 - ^{40}K (Bq kg^{-1}) measured in street dust, grass (dry weight) and bulk soil in July 2000, November 2000 and April 2001.

Sample site	Street dust July 2000	Street dust November 2000	Street dust April 2001	Grass July 2000	Grass November 2000	Grass April 2001	Soil July 2000	Soil November 2000	Soil April 2001
1 (Glasgow Uni/ Western)	211 ± 29	129 ± 18	83 ± 19	132 ± 30	183 ± 37	195 ± 23	<3.7	BDL	BDL
2 (Gartnavel Hosp)	127 ± 25	142 ± 37	BDL	61 ± 36	46 ± 12	151 ± 14	<5.7	BDL	BDL
3 (Strathclyde/ Royal)	164 ± 21	70 ± 24	47 ± 12	163 ± 51	173 ± 69	83 ± 11	< 5.6	BDL	BDL
4 (Victoria Inf)	53 ± 28	60 ± 40	57 ± 16	58 ± 21	213 ± 54	144 ± 16	<5.7	BDL	BDL
5 (Southern General Hosp)	44 ± 20	132 ± 56	360 ± 26	89 ± 25	265 ± 48	190 ± 18	<4.6	BDL	BDL
6 (Pollok Park)	ND	236 ± 26	ND	ND	240 ± 49	ND	ND	BDL	ND
7 (Colston Laboratory)	595 ± 49	191 ± 29	199 ± 21	54 ± 9	314 ± 32	530 ± 41	<3.1	BDL	BDL
8 (Summerston)	ND	135 ± 15	45 ± 9	ND	148 ± 13	362 ± 30	ND	BDL	BDL
9 (Robroyston)	ND	124 ± 21	135 ± 17	ND	332 ± 34	496 ± 36	ND	BDL	BDL

Table 19 - ^{7}Be (Bq kg^{-1}) measured in street dust, grass (dry weight) and bulk soil in July 2000, November 2000 and April 2001.

Sample site	Street dust November 2000	Street dust April 2001	Grass November 2000	Grass April 2001	Soil November 2000	Soil April 2001
1 (Glasgow Uni/ Western)	15.5 ± 2.8	4.6 ± 1.5	17.4 ± 2.9	BDL	29.6 ± 4.4	20.6 ± 1.3
2 (Gartnavel Hosp)	18.4 ± 3.6	15.1 ± 3.3	2.0 ± 0.7	BDL	23.9 ± 1.8	19.5 ± 1.4
3 (Strathclyde/ Royal)	16.8 ± 3.3	3.8 ± 1.1	BDL	BDL	22.3 ± 1.6	17.4 ± 2.3
4 (Victoria Inf)	22.2 ± 3.3	3.8 ± 0.7	17.5 ± 4.0	1.5 ± 0.1	30.5 ± 2.4	28.8 ± 1.7
5 (Southern General Hosp)	21.8 ± 3.6	11.6 ± 3.1	17.1 ± 3.0	BDL	33.8 ± 2.5	44.4 ± 1.8
6 (Pollok Park)	14.3 ± 3.3	ND	BDL	ND	27.4 ± 2.1	ND
7 (Colston Laboratory)	11.4 ± 2.8	7.8 ± 2.9	5.8 ± 2.6	BDL	17.6 ± 1.3	11.9 ± 1.0
8 (Summerston)	19.8 ± 2.8	5.0 ±1.1	5.2 ± 1.4	BDL	21.5 ± 1.6	20.9 ± 1.3
9 (Robroyston)	5.7 ± 2.2	8.3 ± 1.3	9.4 ± 3.2	BDL	23.5 ± 1.8	37.3 ± 2.2

Table 20 – ^{226}Ra (Bq kg^{-1}) in street dust, grass and bulk soil measured in November 2000 and April 2001

Sample site & replicate number	Date of collection	^{137}Cs	^{40}K	^{7}Be	^{226}Ra
Colston –1	17/11/2000	144 ± 6.8	631 ± 50	150 ± 20	24 ± 3.6
Colston – 2	17/11/2000	255 ± 16	238 ± 47	BDL	7 ± 1.7
Colston – 3	17/11/2000	271 ± 17	295 ± 65	223 ± 37	6 ± 1.9
Colston – 4	17/11/2000	176 ± 12	294 ± 49	213 ± 34	10 ± 4.2
Victoria Inf. – 1	10/04/2001	19 ± 2.4	135 ± 32	109 ± 21	BDL
Victoria Inf. – 2	10/04/2001	18 ± 3.5	1300 ± 110	62 ± 28	BDL
Victoria Inf. – 3	10/04/2001	14 ± 2.8	514 ± 74	BDL	11 ± 4.0

Table 21 – Radionuclide specific activities (Bq kg^{-1}) in replicate street dust samples from Colston, November 2000 and the Victoria Infirmary, April 2001.

Sample site & replicate number	Date of collection	^{137}Cs	^{40}K	^{7}Be	^{226}Ra
Colston –1	17/11/2000	BDL	1610 ± 14	361 ± 44	10.4 ± 5
Colston – 2	17/11/2000	3.3 ± 0.5	169 ± 15	40 ± 6	2.1 ± 0.8
Colston – 3	17/11/2000	2.2 ± 0.8	578 ± 43	335 ± 27	5.8 ± 1.4
Colston – 4	17/11/2000	8.5 ± 2.8	106 ± 97	523 ± 49	5.2 ± 2.5
Victoria Inf. – 1	10/04/2001	4.3 ± 0.5	313 ± 19	111 ± 9.2	BDL
Victoria Inf. – 2	10/04/2001	5.5 ± 1.5	435 ± 47	223 ± 26	BDL
Victoria Inf. – 3	10/04/2001	1.8 ± 0.5	663 ± 35	99 ± 12	4.2 ± 0.9

Table 22 – Radionuclide specific activities (Bq kg^{-1}) in replicate grass samples from Colston, November 2000 and the Victoria Infirmary, April 2001.

Sample site & replicate number	Date of collection	^{137}Cs	^{40}K	^{7}Be	^{226}Ra
Colston –1	17/11/2000	49 ± 3.0	312 ± 22	BDL	23 ± 1.8
Colston –2	17/11/2000	29 ± 1.8	259 ± 17	BDL	16 ± 1.3
Colston –3	17/11/2000	26 ± 1.7	253 ± 18	BDL	13 ± 1.1
Victoria Inf. – 1	10/04/2001	73 ± 2.7	256 ± 13	BDL	29 ± 1.6
Victoria Inf. – 2	10/04/2001	57 ± 2.2	250 ± 13	BDL	29 ± 1.7

Table 23 – Radionuclide specific activities (Bq kg^{-1}) in replicate soil samples from Colston, November 2000 and the Victoria Infirmary, April 2001.

Data was unavailable for Colston replicate number 4 and Victoria Infirmary replicate numbers 3 and 4.

Gamma spectrometry results for Clyde Estuary water

Date sample collected	^{40}K	^{131}I
March 2001	5.36 ± 0.40	BDL
May 2001	10.0 ± 0.66	0.041 ± 0.033

Table 24 - Specific activity of ^{40}K and ^{131}I (Bq l^{-1}) in Clyde estuary water

All other radionuclides were below the detection limit.

3.3 - Alpha spectrometry- plutonium results

Sample depth (cm)	$^{239,240}Pu$	^{238}Pu	^{238}Pu / $^{239,240}Pu$ activity ratio
0-1	3.63 ± 0.19	0.083 ± 0.011	0.023 ± 0.0033
1-2	2.51 ± 0.13	0.088 ± 0.011	0.035 ± 0.0047
2-3	1.42 ± 0.06	0.023 ± 0.003	0.016 ± 0.0022
3-4	0.77 ± 0.04	0.025 ± 0.004	0.033 ± 0.0056
4-5	0.034 ± 0.02	0.005 ± 0.003	0.014 ± 0.0118
5-6	0.23 ± 0.01	0.010 ± 0.001	0.042 ± 0.0046
6-7	0.24 ± 0.02	0.01 ± 0.003	0.040 ± 0.0124
7-8	0.08 ± 0.02	BDL	-
8-9	Not tested	Not tested	-
9-10	0.11 ± 0.01	BDL	-

Table 25 – $^{239,240}Pu$ and ^{238}Pu (Bq kg^{-1}) for the Pollok Park deciduous forest soil core (October 2000)

Sample depth (cm)	^{239,240}Pu	^{238}Pu	^{238}Pu / ^{239,240}Pu activity ratio
0-1	0.42 ± 0.05	0.016 ± 0.014	0.039 ± 0.0344
1-2	1.71 ± 0.11	0.057 ± 0.012	0.033 ± 0.0073
2-3	3.64 ± 0.21	0.11 ± 0.014	0.030 ± 0.0042
3-4	3.44 ± 0.20	0.138 ± 0.018	0.040 ± 0.0057
4-5	3.83 ± 0.19	0.142 ± 0.017	0.037 ± 0.0048
5-7	2.40 ± 0.13	0.045 ± 0.007	0.019 ± 0.0031
7-8	1.04 ± 0.06	0.036 ± 0.007	0.035 ± 0.0071

Table 26 – ^{239,240}Pu and ^{238}Pu (Bq kg^{-1}) for the Pollok Park coniferous forest soil core (October 2000)

Sample depth (cm)	^{239,240}Pu	^{238}Pu	^{238}Pu / ^{239,240}Pu activity ratio
0-2	Not tested	Not tested	-
2-3	0.40 ± 0.03	0.019 ± 0.005	0.049 ± 0.0389
3-4	0.39 ± 0.02	0.010 ± 0.004	0.027 ± 0.0109
4-5	0.42 ± 0.04	0.024 ± 0.008	0.059 ± 0.0205
5-6	0.88 ± 0.12	0.082 ± 0.025	0.092 ± 0.0307
6-7	0.41 ± 0.02	0.010 ± 0.004	0.025 ± 0.0100
7-8	0.044 ± 0.03	0.013 ± 0.004	0.030 ± 0.0224

Table 27 – ^{239,240}Pu and ^{238}Pu (Bq kg^{-1}) for the Pollok Park grassland soil core (October 2000)

Sample depth (cm)	^{239,240}Pu	^{238}Pu	^{238}Pu / ^{239,240}Pu activity ratio
Dec 0-2 cm	2.14 ± 0.12	0.128 ± 0.018	0.060 ± 0.0091
Con 0-2 cm	5.03 ± 0.34	0.290 ± 0.039	0.058 ± 0.0087
Con 2-4 cm	3.39 ± 0.19	0.068 ± 0.012	0.020 ± 0.0037
Grass 0-2 cm	BDL	BDL	-

Table 28 - ^{239,240}Pu and ^{238}Pu (Bq kg^{-1}) for the Pollok Park bulk soil samples (June 2001)

3.4 - BCR sequential extraction results

No plutonium was found in the extracts from stages one and two or in the blanks. The coniferous bulk samples were the only samples to have values greater than the detection limit as shown in Table 29.

Sample depth (cm)	^{239,240}Pu	^{238}Pu	^{238}Pu / ^{239,240}Pu activity ratio
Con 0-2 stage 3	0.046 ± 0.002	0.0019 ± 0.0004	0.040 ± 0.0086
Con 2-4 stage 3	0.020 ± 0.002	0.0011 ± 0.0006	0.054 ± 0.0299

Table 29 - ^{239,240}Pu and ^{238}Pu (Bq kg^{-1}) for the Pollok Park bulk soil sample sequential extracts (June 2001)

3.5 - Liquid scintillation counting results

Each sample and each deionised water 'reference sample' was counted 10 times in 100-minute cycles. Disintegrations per minute were reported for each cycle and an average was calculated for each sample. The estuarine samples were corrected by subtracting the average disintegrations per minute value from the reference sample and then the results were converted to Bq l^{-1}. The limit of detection above the ambient ^{3}H level found in the deionised water is defined as the 3σ error value on the background counts (the reference sample counts per minute value). 1σ error of the background is determined by:

$1\sigma = \sqrt{}$ Total counts for 1000 minute count

(This is 10 x 100 minute count)

This 1σ value is multiplied by 3, to gain a 3σ value. This is then converted back to counts per minute by dividing the 3σ value by 1000. This value is then converted to Bq, using the experimentally pre-determined counting efficiency of 23.1% and, as this was for a 10 ml sample, the value is then converted to Bq l^{-1}.

	^{3}H (Bq l^{-1})		
	October 2000	December 2000	March 2001
Dalmarnock Bridge	<3.4	<3.4	<3.4
Kings Bridge	<3.4	<3.4	<3.4
SECC	<3.4	<3.4	<3.4
Balmoral Street	<3.4	<3.4	<3.4
Renfrew Ferry	<3.4	<3.4	<3.4

Table 30 - ^{3}H values in Clyde estuarine water

As can be seen from table 30, after background correction, all results were below the limit of detection of 3.4 Bq l^{-1}. There are techniques, such as electrolytic enrichment, in which the sensitivity of ^{3}H detection could be increased (Neary, 1997). This however was out-with the scope of this one-year project.

Chapter 4- Discussion

In the following discussion of the results, rather than exhaustively apply every interpretative approach to every nuclide, different techniques have been applied selectively to individual nuclides to illustrate the type of interpretation that is possible.

Gamma spectrometry analysis

^{40}K

A convenient way to define and display the spatial variability in a data set of environmental measurements is to represent them on a map. To interpolate data accurately onto a grid would, however, require a much higher spatial resolution of sampling points than was possible in the present work. By not extrapolating the results to other areas but by simply putting the findings into a geographical context, the results can still be depicted usefully as illustrated in Figures 10-12. These maps allow an instantly appreciable and accessible presentation of the results and the differences in specific activities between soil, street dust and grass can be seen, as well as short-term temporal variations for a given sample site. It can be seen from these illustrations that the variability of ^{40}K activities in the three matrix types generally decreases in the order grass > street dust > soil. ^{40}K was chosen to exemplify this as it gives a good indication of the variability of a natural radionuclide, which is a major contributor to the background radiation exposure of humans. As can be seen from the maps (Figures 10-12), there is considerable variation both spatially and over time in the three environmental media. Not only would this create considerable variation in the dose received from ^{40}K, it highlights the difficulty in defining an ‘average’ dose.

^{40}K activities in the environment vary not only with bedrock, and therefore soil type, but also with environmental management practices such as the application of fertiliser.

The ^{40}K specific activity in grass exhibits a variability that could be attributed to a number

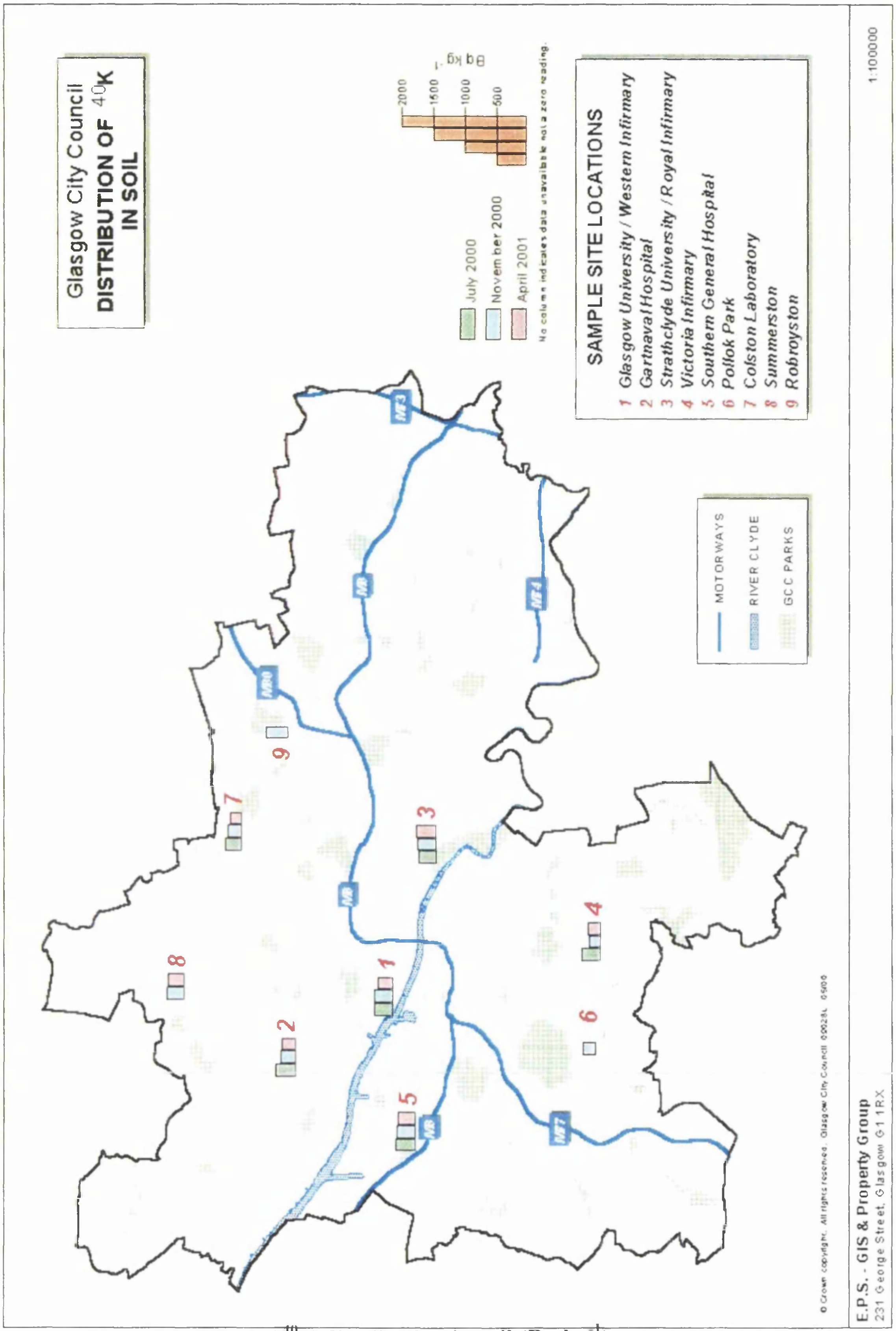


Figure 10. Map of ^{40}K distribution in soil (Bq kg^{-1})

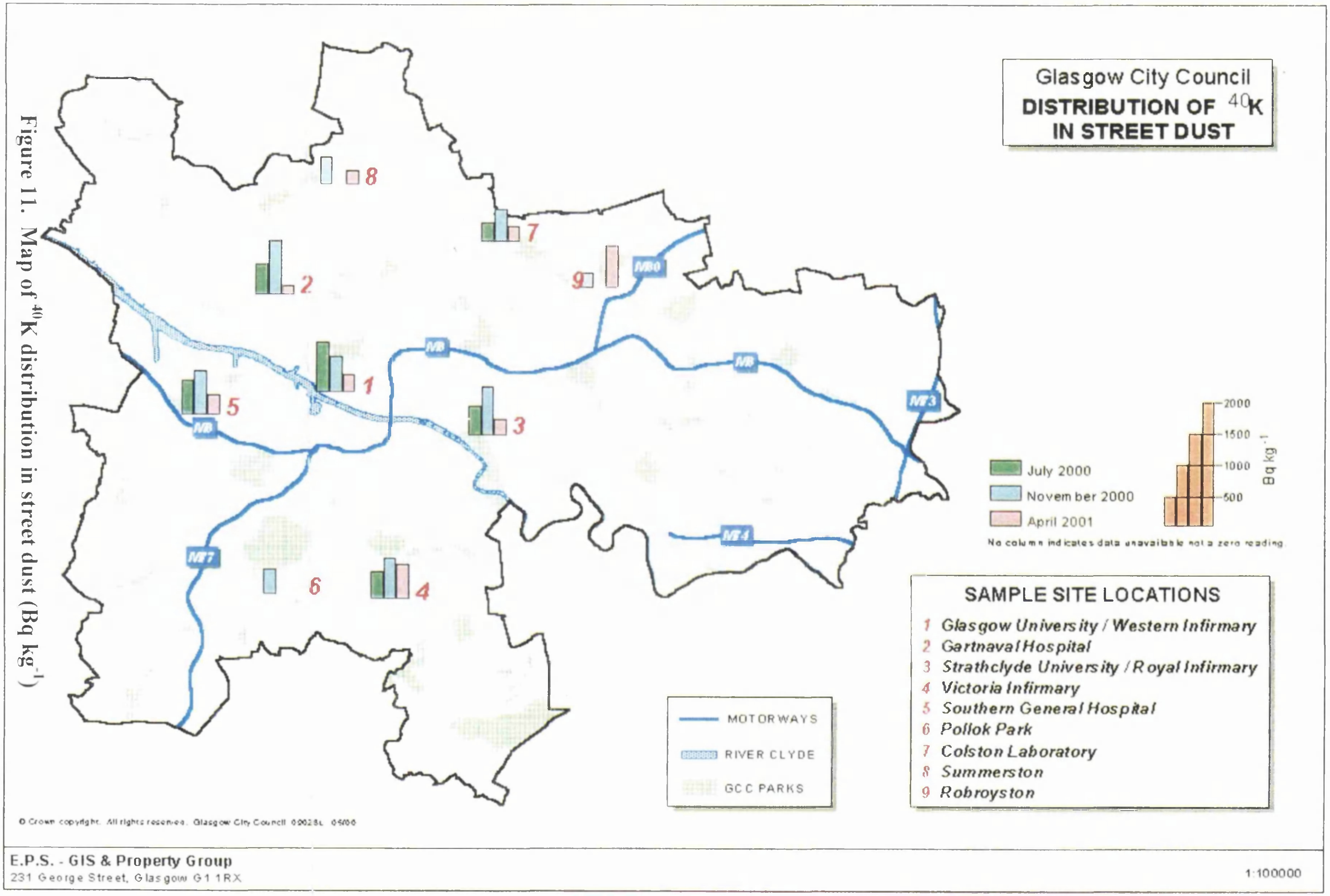


Figure 11. Map of ^{40}K distribution in street dust (Bq kg^{-1})

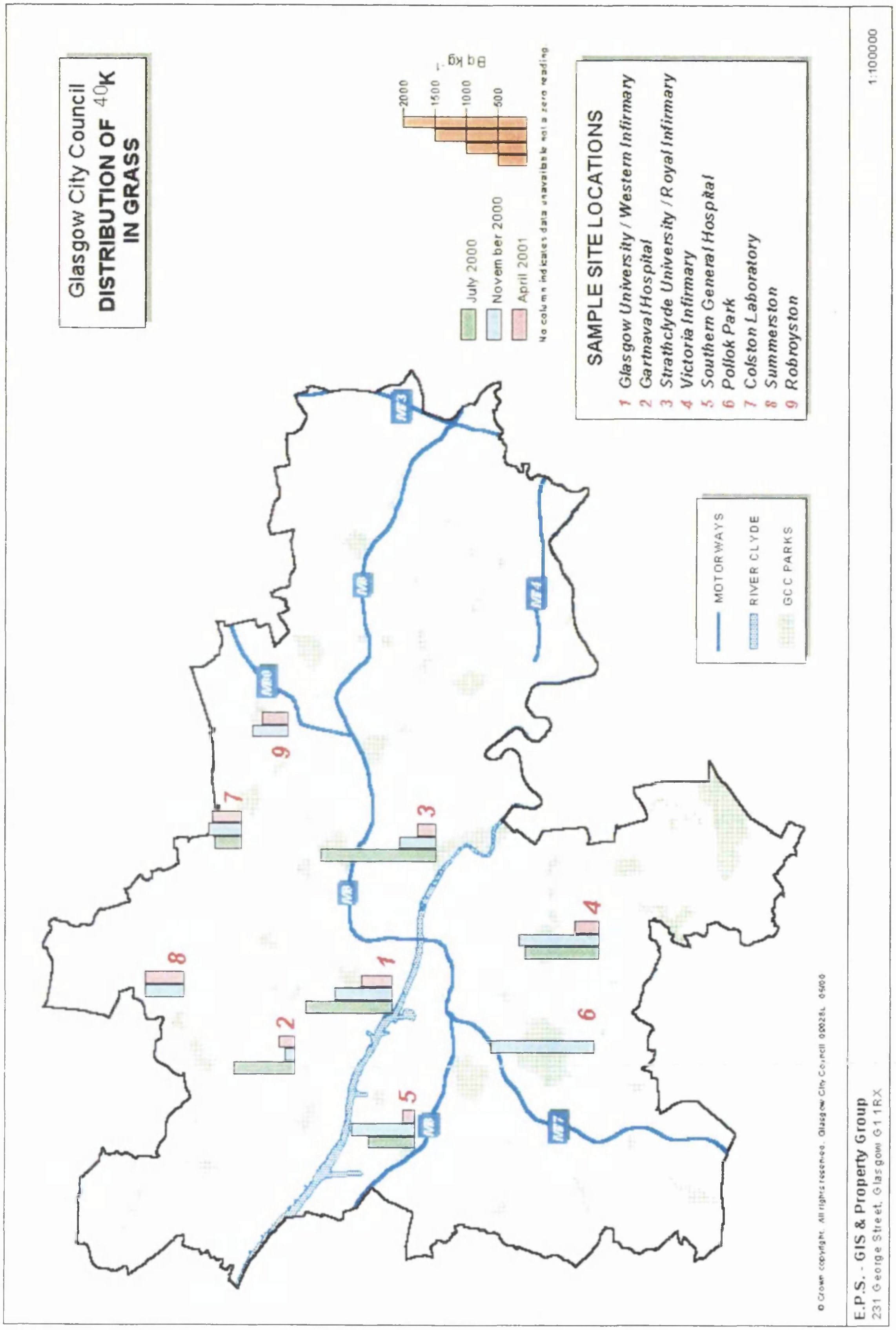


Figure 12. Map of ^{40}K distribution in grass (Bq kg^{-1})

of factors (Figure 13). For Glasgow University, Gartnaval Hospital and Strathclyde University, the highest results were obtained in July, which may be after the application of fertiliser and is during a high growth rate period. The degree of management at each site will vary, thus introducing further variation into the ^{40}K results. Potassium is a major element found in plants and is required for many of their vital functions. If potassium is present in soil at high enough levels, grass will continue to take up potassium, beyond the requirements of the plant (De Boer, 1999). This is known as "luxury uptake" and may be responsible for some of the variability in the K content of the grass. The measured ^{40}K specific activities of the grass were generally lowest in April. This may reflect a slight decrease in soil ^{40}K over the winter due to leaching of the available potassium (Figure 14), or be caused by slower growth rates, and therefore slower potassium uptake than in July. It may also be possible that, at sites that receive environmental management, fertiliser may not yet have been applied for the year. The measurements taken at the Colston Laboratory, Summerston and Robroyston are all very similar, both spatially and over the three (or two) sampling times. Therefore, although some of the grass results display considerable variation, this is not the case for all the sites.

The ^{40}K measured in street dust is generally highest in July 2000 (Figure 15) with the exceptions of Glasgow University and Robroyston sampling sites, where maximum values were observed in November 2000 and April 2001 respectively. This could also be possibly caused by the application of fertiliser to land prior to the sampling in July 2000. The specific activities of ^{40}K in street dust are generally slightly higher than in soil. A study by Narayana et al (1995) found that the highest ^{40}K activities were in the 125-250 μm fraction of soil (see section 1.3.4) and it is feasible that the street dust may be made up of wind blow, or otherwise, fine grained soil.

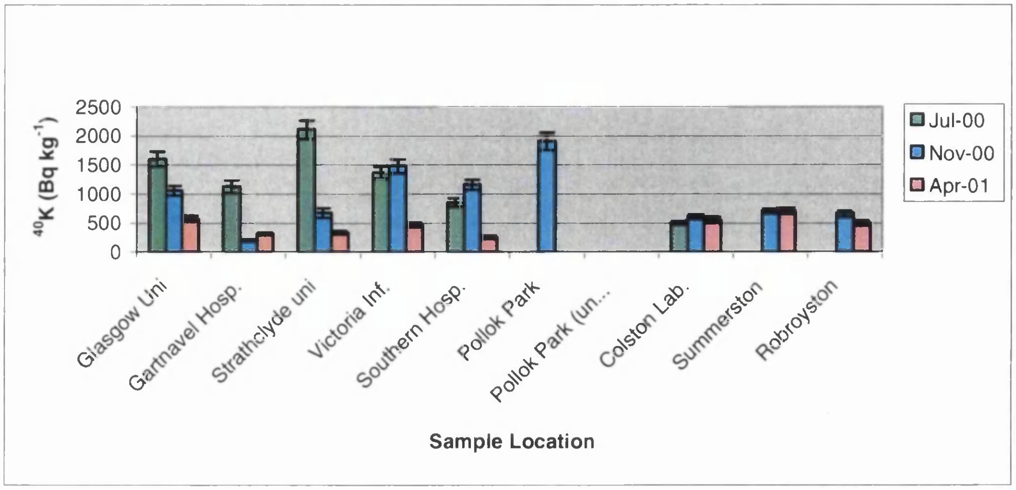


Figure - 13 – ^{40}K in measured in grass (Bq kg^{-1}) in July 2000, November 2000 and April 2001

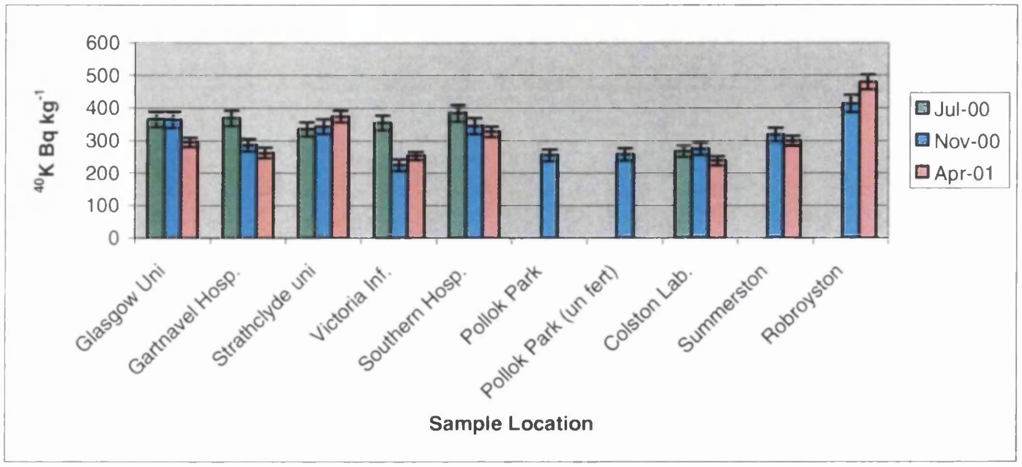


Figure - 14 – ^{40}K in measured in soil (Bq kg^{-1}) in July 2000, November 2000 and April 2001

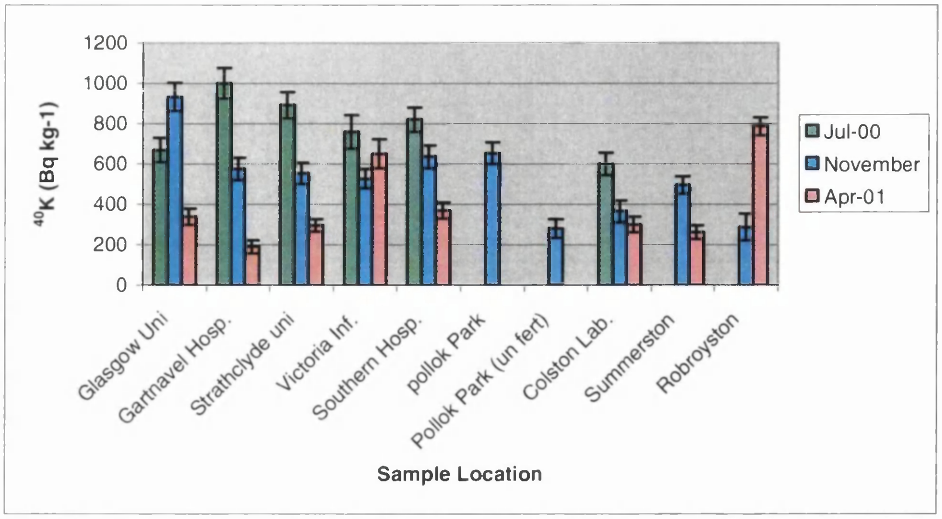


Figure - 15 – ^{40}K in measured in street dust (Bq kg^{-1}) in July 2000, November 2000 and April 2001

As can be observed in Figure 14, the variability in the soil is less than that of the other two matrices indicating it is a more homogeneous medium. The comparison of the fertilised grassland and un-fertilised grassland from Pollok Park shows that the two soil samples have very similar ^{40}K specific activities. The fertilised grass sample however, has a ^{40}K specific activity of 1910 Bq kg^{-1} whilst the unfertilised grass has a ^{40}K specific activity below the limit of detection. This pattern is repeated to some extent in the street dust samples, with a decrease from 652 Bq kg^{-1} to 279 Bq kg^{-1} in the fertilised and unfertilised samples, respectively. This highlights a pattern which can almost certainly be attributed to environmental management.

The relationship between the ^{40}K specific activities of the sample types can be explored further by looking at transfer factors (Green et al., 1995). The transfer factor of ^{40}K from soil to grass can give an indication of the variation in grass ^{40}K uptake from site to site and quantify the level of influence the soil ^{40}K has on the grass ^{40}K activities. The transfer factors were calculated as follows:

$$\text{Transfer factor} = \frac{^{40}\text{K specific activity in grass}}{^{40}\text{K specific activity in soil}}$$

Sample location	Soil to grass transfer factor		
	July 2000	Nov 2000	April 2001
1 (Glasgow University / Western Infirmary)	4.4	2.9	1.9
2 (Gartnavel Hospital)	3.1	0.7	1.2
3 (Strathclyde University/ Royal Infirmary	6.3	2.0	0.9
4 (Victoria Infirmary)	3.9	6.6	1.9
5 (Southern General Hospital)	2.2	3.4	0.8
6a (Pollok Park, un-fertilised)	-	-	-
6b (Pollok Park, fertilised)	-	7.5	-
7 (Colston Laboratory)	1.9	2.2	2.3
8 (Summerston)	-	2.2	2.4
9 (Robroyston)	-	1.6	1.0

Table 31 - Soil to grass ^{40}K transfer factors

If the ratio is greater than one, for example as at the Strathclyde University/ Western Infirmary site in July and November 2000, it indicates preferential uptake of potassium by the grass relative to the soil. If the ratio is less than one, for example as at the Southern General Hospital in April 2001, it indicates preferential retention of the potassium in the soil. Although there is considerable variation between the transfer factors observed at different sampling times at some of the sites, tentative inferences about the influence of environmental management may be drawn. From communication with park rangers at Pollok Park, is known that surface application of fertilisers is carried out in some areas of the park. As can be seen from Table 31, the soil to grass transfer factor is highest for the Pollok Park fertilised site indicating that the grass receives, or takes up, higher levels of potassium relative to the soil. This would suggest that the fertiliser is applied in a soluble form and is removed from the soil in solution with no significant long term soil retention occurring. The two lowest ratios occur at Gartnaval Hospital and the Southern General Hospital where samples were taken in areas unlikely to receive any application of fertilisers. Therefore fixed potassium in the soil (rather than potassium in a soluble, plant available form) is likely to have had a more dominant effect over the soil to grass transfer ratio. However, there is considerable variation in the transfer factors and more samples would be required to assess if the variability seen in these factors is related to the size of the data set, or if this parameter is inherently subject to large variation.

Another approach that could be taken when considering transfer factors is to calculate the aggregated transfer factor (TAG), which uses the inventory of a radionuclide in the soil rather than simply using the specific activity (Amundsen et al., 1996). Using the inventory like this avoids complications introduced by variations in dry bulk density between different soil types. The TAG is calculated as follows:

$$\text{TAG} = \frac{\text{specific activity in vegetation}}{\text{Inventory in soil}}$$

This may be a useful approach to take in any future sampling to eliminate the variability caused by sampling of different soil types.

Up to this point, this discussion has focussed on interpreting the experimental results gained for the chosen sample sites. If inferences drawn from these results were going to be applied to other areas of the city, an understanding of the data population would help define the likely range of results that would be expected. A useful method for gaining more understanding of the data population and a good way for presenting environmental data, is to plot a histogram of the results. These data are presented below in Figures 16 and 17.

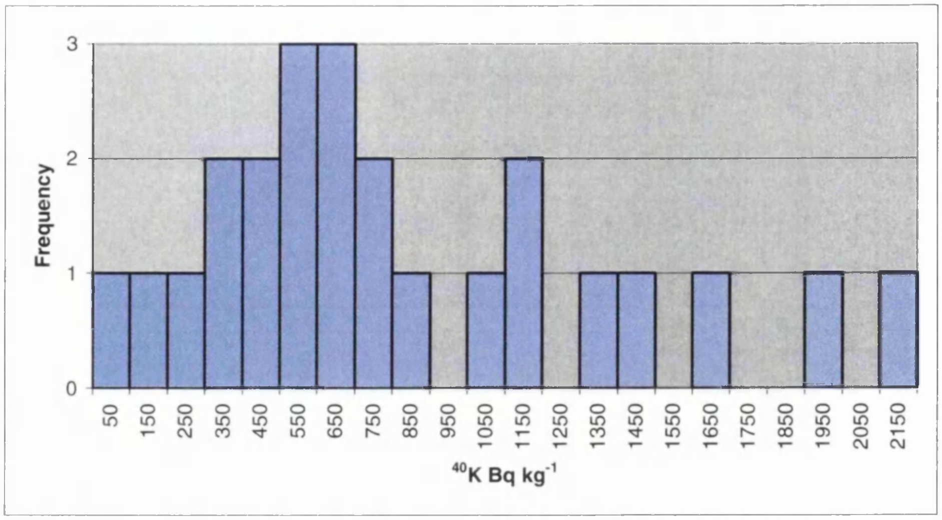


Figure 16 – ^{40}K (Bq kg^{-1}) in grass- Distribution of results

The data for grass suggest a spread of results around a modal value in the range 600- 700 Bq kg^{-1} of ^{40}K. This could possibly be taken to represent the 'natural' variation in ^{40}K plus a 'tail' at the higher end of the scale, which could reasonably be taken to represent the effects of fertiliser application. The five samples which make up this 'tail' are: Glasgow University/ Western Infirmary in July 2000; Strathclyde University/ Royal Infirmary in July 2000; the Victoria Infirmary in both July 2000 and November 2000 and Pollok park (fertilised site) in November 2000. These are all sites that receive some degree of environmental management.

This observed variability may to some degree, however, reflect the grass sampling times with the higher values representing periods of maximum growth, whilst the lower values may represent periods of plant senescence.

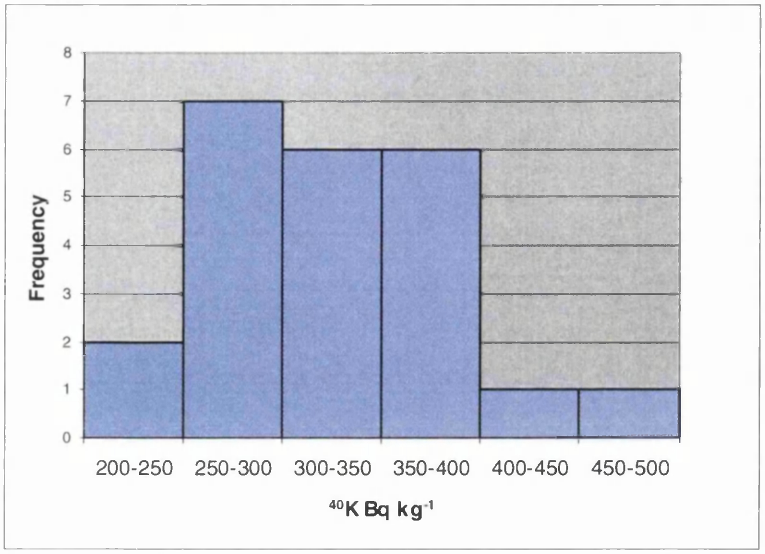


Figure 17 - ^{40}K (Bq kg^{-1}) in soil- Distribution of results

The data set for soil, displayed in figure 17, is close to a normal distribution. The range in results for the soil samples is much smaller than for the grass and the street dust, reflecting a higher level of homogeneity of the soil than in the two other sample types.

With the limited data set available here, a rigorous statistical interpretation would be of limited value. However, summary statistics were derived using Microsoft Excel to illustrate the type of information that can be derived from such data. The ^{40}K summary statistics are shown in Table 32.

	^{40}K Grass	^{40}K Soil	^{40}K Street Dust
Mean	859 (Bq kg^{-1})	323 (Bq kg^{-1})	551 (Bq kg^{-1})
Standard deviation	532 (Bq kg^{-1})	62 (Bq kg^{-1})	236 (Bq kg^{-1})
Minimum	197 (Bq kg^{-1})	225 (Bq kg^{-1})	189 (Bq kg^{-1})
Maximum	2115 (Bq kg^{-1})	479 (Bq kg^{-1})	1000 (Bq kg^{-1})
No: of observations	23	23	23

Table 32 – ^{40}K data population summary statistics

Firstly, the mean gives an indication of the ^{40}K specific activities that were found in each matrix. This shows that the specific activities in the three data populations decrease from grass, the highest, to street dust and soil, respectively. The standard deviation describes the variation in the measurement. The grass population exhibited the largest standard deviation unlike the soil, which had the smallest standard deviation.

The ^{40}K specific activity range found for street dust is larger than that for soil. This could be attributed to the large variation in grain size of the street dust samples and its origin from a number of different sources. For example, the street dust from sites close to roads with a heavy traffic load, such as the Victoria Infirmary samples, will be largely made of abraded road making materials and dust from the traffic. In contrast, at sites like Pollok Park, the street dust will be largely made up of soil.

^{137}Cs

The variability in specific activities and distribution of ^{137}Cs, an anthropogenic radionuclide, provided an opportunity to analyse the distribution of this environmental contaminant. As previously stated, the sources of ^{137}Cs to the environment include fallout from atmospheric nuclear weapons tests and the Chernobyl reactor accident.

The collection of such a data set, whilst not extensive enough to provide a comprehensive baseline, can define a range with which any future enhancement could be compared.
The ^{137}Cs specific activities in grass were generally very low (Figure 18) and show a broadly exponential distribution (Figure 19). Uptake, and subsequent release, of ^{137}Cs by soil could represent a fairly long-term source to the grass if the ^{137}Cs in the soil was not too tightly held by clay minerals (see section 1.3.4).

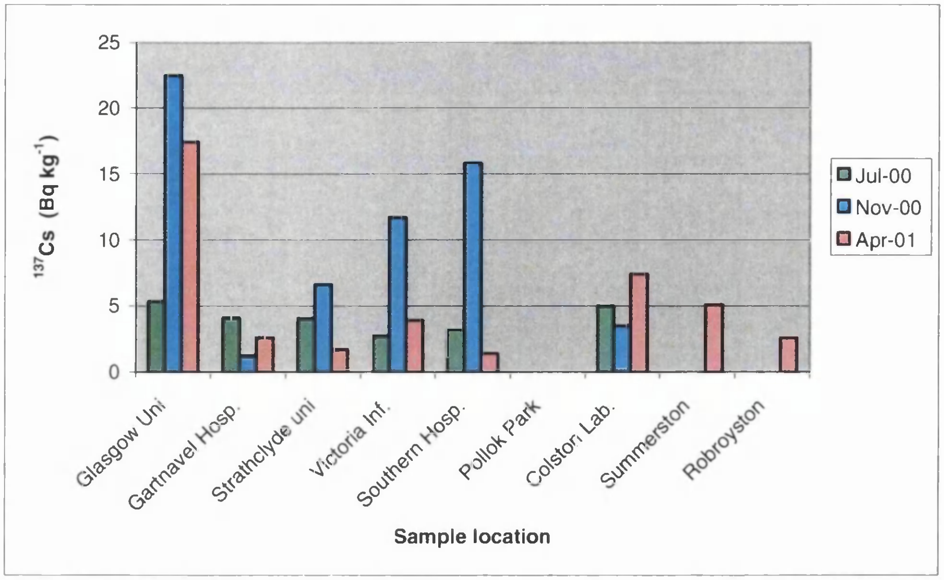


Figure 18 - ^{137}Cs (Bq kg^{-1}) in grass measured in July 2000, November 2000 and April 2001.

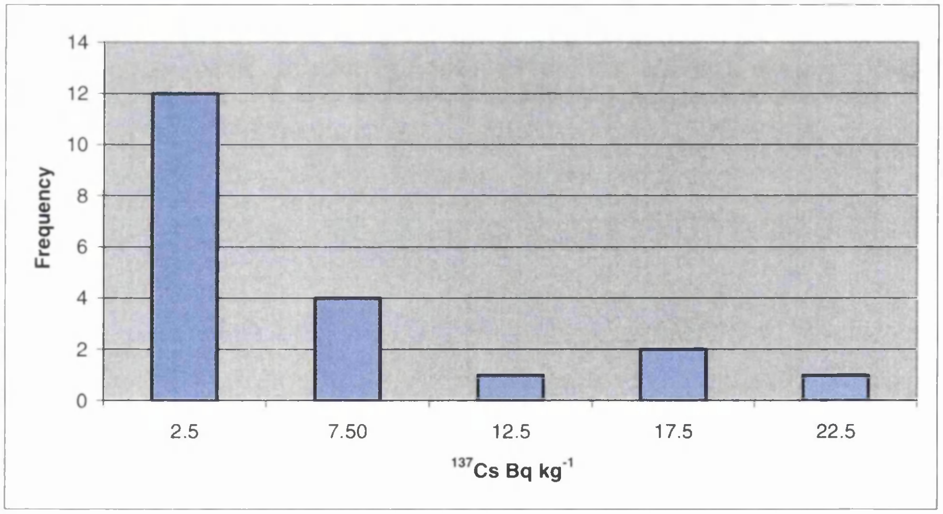


Figure 19 - ^{137}Cs (Bq kg^{-1}) in grass – distribution of results.

The specific activity of ^{137}Cs in soil varies between approximately 2 and 85 Bq kg^{-1} and the data appear to have a normal distribution, as was seen with the soil ^{40}K data.

As mentioned before, variability in the street dust results is to be expected, as it is a very heterogeneous matrix. The street dust results for ^{137}Cs, however, show two outlying results (Figure 20), which whilst not of radiological concern when compared to the GDLs set for mixed soil (no GDL available for street dust), are definitely outliers from the main body of results. The two outliers do not occur at the same site and in both cases were observed only at one sampling time at each site. They are both derived from sites which potentially could be sources of ^{137}Cs, so a point source release from the laboratories may have been responsible for the elevated levels. There are, however, a number of other possible explanations: the grain size of these particular samples could be smaller than the other samples and therefore could have a higher concentration of ^{137}Cs (see section 1.3.4); or the two samples could have had a higher illitic clay content than the other samples and thus have a better binding capacity for ^{137}Cs.

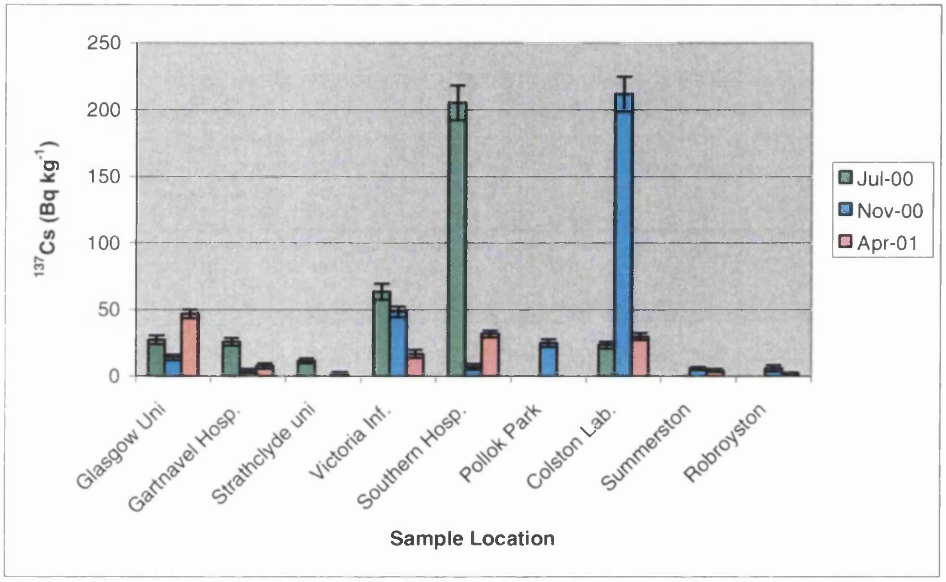


Figure 20 - ^{137}Cs (Bq kg^{-1}) in Street dust measured in July 2000, November 2000 and April 2001.

The ^{137}Cs data populations for grass, soil and street dust were analysed using the same descriptive statistics as used for ^{40}K. These results are presented below in Table 33. In order to assess the influence of the two outlying results, the ^{137}Cs data population for street dust was analysed once including the outliers and once excluding the outliers.

	^{137}Cs Grass	^{137}Cs Soil	^{137}Cs Street dust (with outliers)	^{137}Cs Street dust (without outliers)
Mean	6.9 (Bq kg^{-1})	35.1 (Bq kg^{-1})	35.6 (Bq kg^{-1})	19.1 (Bq kg^{-1})
Standard deviation	5.9 (Bq kg^{-1})	20.7 (Bq kg^{-1})	57.0 (Bq kg^{-1})	17.5 (Bq kg^{-1})
No: observations	23	23	23	21
Minimum	1.2 (Bq kg^{-1})	2.4 (Bq kg^{-1})	0	0
Maximum	22.5 (Bq kg^{-1})	85.1 (Bq kg^{-1})	211.5 (Bq kg^{-1})	63.4 (Bq kg^{-1})

Table 33 – ^{137}Cs data population summary statistics

In this case, the mean for the grass is the lowest. The standard deviation and the range of the street dust population is lowered significantly by removing the outliers. More sampling and analysis would be required to assess if these ‘outliers’ really do represent a significant increase from the average expected value. These results, whilst highlighting the variability of environmental ^{137}Cs specific activities, characterise the range of activities for comparison with future monitoring. These ranges could be used to estimate an average dose, but the uncertainty related to this estimate would be large.

^{7}Be

As ^{7}Be is a cosmic ray-produced radionuclide, it provides an interesting example of recent atmospheric deposition and, given its uniform atmospheric production rate and relatively uniform rainfall over the study area, it might be reasonable to expect a fairly uniform distribution. Therefore, variations observed must be largely due to differences in sample composition or the management of the site. Certainly, in this case it would be expected that temporal variations would be larger than spatial variations. Calculating the coefficient of variation can quantitatively demonstrate the amount of variation observed and may be useful when comparing spatial and temporal variation. It is calculated as follows:

$$\text{Coefficient of variation} = \frac{\text{standard deviation of data population}}{\text{mean of population}} \times 100\ \%$$

However, with only three sampling times, considerable variation was observed. This is the type of approach that could be applied if a more regular monitoring program was adopted. Basic summary statistics were applied and can be found in Table 34.

	^{7}Be in grass	^{7}Be in street dust
Mean	201 (Bq kg^{-1})	153 (Bq kg^{-1})
Standard deviation	132 (Bq kg^{-1})	124 (Bq kg^{-1})
No: of observations	23	23
Minimum	46 (Bq kg^{-1})	44 (Bq kg^{-1})
Maximum	530 (Bq kg^{-1})	595 (Bq kg^{-1})

Table 34 – ^{7}Be data population summary statistics

It was not surprising to find the highest specific activities in the grass samples as ^{7}Be will be deposited on the surface of the grass both by wet and dry deposition. The street dust had the next highest values after the grass samples and this may be explained by the fact that the street dust samples were gathered from exposed street surfaces, which will also receive direct input of ^{7}Be. Also, all of the street dust samples will have been at or near the surface at the time of sampling unlike the soil samples, a large proportion of which will have been at depth (0-15 cm) for some time. The bulk soil samples had no detectable ^{7}Be (see section 3.2).

^{226}Ra

The extended results from the gamma spectrometry analyses carried out at GCSS are presented in Appendix one. The measured ^{214}Pb and ^{214}Bi activities were in generally good agreement and the activity of ^{226}Ra was calculated from their average.

^{226}Ra is a naturally occurring radionuclide, which is a member of the ^{238}U decay chain. The distribution and variation in the specific activities of ^{40}K, which is also a naturally occurring radionuclide, showed possible influences from anthropogenic input. Anthropogenic influence is less likely with a radionuclide such as ^{226}Ra, but is not impossible. There was considerable variation between the ^{226}Ra results obtained for the two sampling times in November 2000 and April 2001 and, certainly in the case of the grass and the street dust samples, the November samples are almost consistently higher. The soil sample results are the most consistent between the two sampling times at the majority of the sites. As the soil formed from the geological parent material, the parent nuclides for ^{226}Ra (most importantly ^{238}U) will be present, supplying ^{226}Ra to the soil through their own decay. The soil data population approximates to a normal distribution and, again, is fairly similar to the ^{40}K soil data population. This is consistent with the nature of the ^{226}Ra origin in soil. These results suggest a natural range of specific activities from approximately 12 to 40 Bq kg^{-1} as shown in Figure 21.

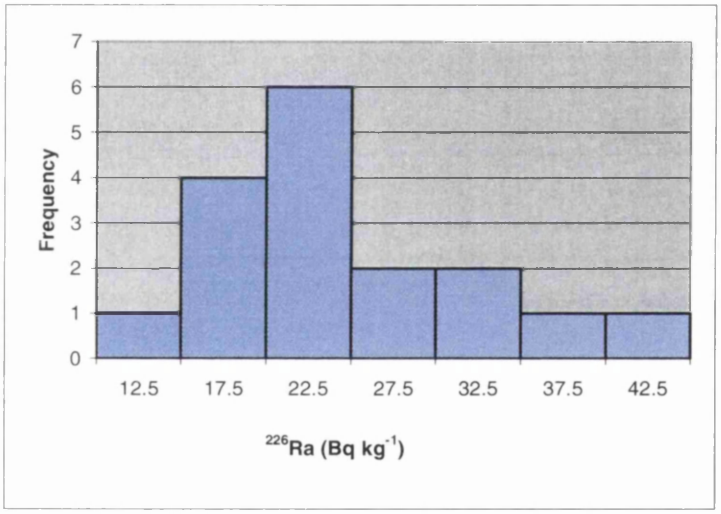


Figure 21 – ^{226}Ra (Bq kg^{-1}) in soil – distribution of results.

The grass population, however, shows a distribution which is close to exponential (Figure 22) as was observed for ^{137}Cs in grass. This differs from the ^{40}K grass data population which resembled a normal distribution with a 'tail'. ^{226}Ra is not a macronutrient required by plants, as ^{40}K is, and so it will not be as readily taken up by the grass.

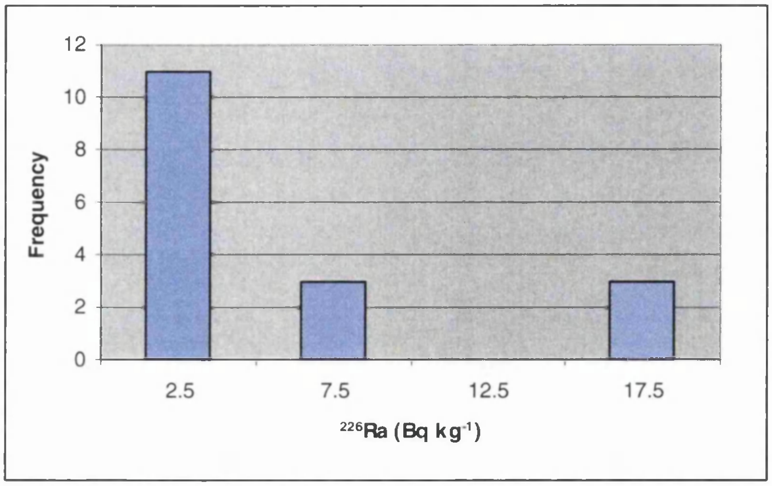


Figure 22 – ^{226}Ra (Bq kg^{-1}) in grass – distribution of results.

	^{226}Ra Grass	^{226}Ra Soil	^{226}Ra Street dust
Mean	9.5	25.3	12.1
Standard deviation	6.9	8.1	6.4
No: of observations	8	17	17
Minimum	1.45	11.9	3.8
Maximum	17.5	44.4	22.2

Table 35 – ^{226}Ra data population summary statistics

The ^{226}Ra data population summary statistics are shown in Table 35. The standard deviation was largest in the case of the soil. The grass, soil and street dust results show mean ^{226}Ra specific activities of 9.5 Bq kg^{-1}, 25.3 Bq kg^{-1} and 12.1 Bq kg^{-1}, respectively.

Clyde estuary water

^{40}K was detectable in both estuarine samples at 5.36 Bq l^{-1} and 10 Bq l^{-1} in March 2001 and May 2001, respectively. These results are comparable to the specific activity of 11.16 Bq l^{-1} of ^{40}K in open sea water (Riley and Skirrow, 1975), with the slightly lower values here being attributed to the lower salinity in estuarine waters. On one occasion (May 2001), ^{131}I was detected at low levels. ^{131}I is used in hospitals as a tool in thyroid function diagnosis and thyroid imaging (Germain, 1993). The Renfrew Ferry sampling location is down stream of the sewage works which handles the waste from hospitals, including the Southern General Hospital. This hospital has a nuclear medicine department, is licensed to discharge15 GBq of ^{131}I monthly to sewers, and so is a feasible source of the ^{131}I (Personal communication: J Gemmill, SEPA). The detection of this radionuclide illustrates that such a survey is probably sensitive enough to detect radionuclides that can be linked to a point source. With more regular monitoring, the extent to which ^{131}I is present in the Clyde could be assessed and the probable source of this nuclide could be identified with more certainty.

Small scale spatial variation

In studies of soil systems it is difficult to obtain truly representative samples because of spatial variability and in order to characterise such variations, replicate samples were taken at two of the sites, one at each sample time. Clearly, any spatial variation will also be incorporated into the 'temporal' variation observed by this sampling regime.

The ^{137}Cs street dust samples showed good reproducibility (Tables 21-23) at both the sites under investigation. ^{137}Cs specific activities ranged from 14-19 Bq kg^{-1} at the Victoria Infirmary site as opposed to 144-271 Bq kg^{-1} at the Colston Laboratory. The ^{137}Cs specific activities in grass however, do not show the same degree of between-site variation and are all within the range 1.8-8.5 Bq kg^{-1}. ^{137}Cs variability is greatest in street dust as opposed to ^{40}K, which had a high variability in both street dust and grass. This variation is up to an order of magnitude in street dust and grass at both sites and at both sampling times. Sample composition is almost certainly the cause of this variation.

^{226}Ra in each matrix, and ^{7}Be in street dust and grass (soil results were below the limit of detection) show a considerable degree of variation but it is difficult to define the variation within such a small data set. If the data set was larger, the coefficient of variation could be used to quantify the variation.

Pollok Park Cores

An important point to note before embarking on the discussion of these cores is that only one core of each soil type was analysed. Therefore, findings are specific to the cores analysed and may not necessarily be representative of the soil type in general. Additional cores would have to analysed to assess the local spatial variation and to determine if the trends observed in this study are consistent for the area.

Soil characteristics

Figure 23 illustrates a significant difference in organic content between the three soil types. In the top 5 cm of the coniferous profile, there is close to 60% organic material at the surface, representing an area of little organic breakdown. Below 5 cm in the coniferous soil, the percentage of organic material decreases relatively regularly down to approximately 10% at the base of the core. This represents an increase in breakdown of organic material relative to the top 5 cm of the core. The organic input to this system would be continuous throughout the year and would largely be made up of waxy, resistant leaves (Behera et al., 2002; Dursun et al., 1993).

The deciduous soil shows a smooth decline from close to 40 % organic material at the surface down to approximately 5% at the base of the core. This smooth decline in organic material is consistent with field observations of the well-graded soil profile. Organic input here would differ from the coniferous forest soil input in that it would be concentrated in the autumn months and consist with less resistant, deciduous leaves (Reith et al., 2002). There is also the possibility of loss of leaves (and any associated metals) by either wind action or human activity.

The grassland soil exhibited less of a decline in % organic material than the two other soils, with a variation from approximately 20% at the surface to 10% at the base of the core. Organic input in the grassland system would be relatively small compared to the other two systems and would consist of roots and decaying grass.

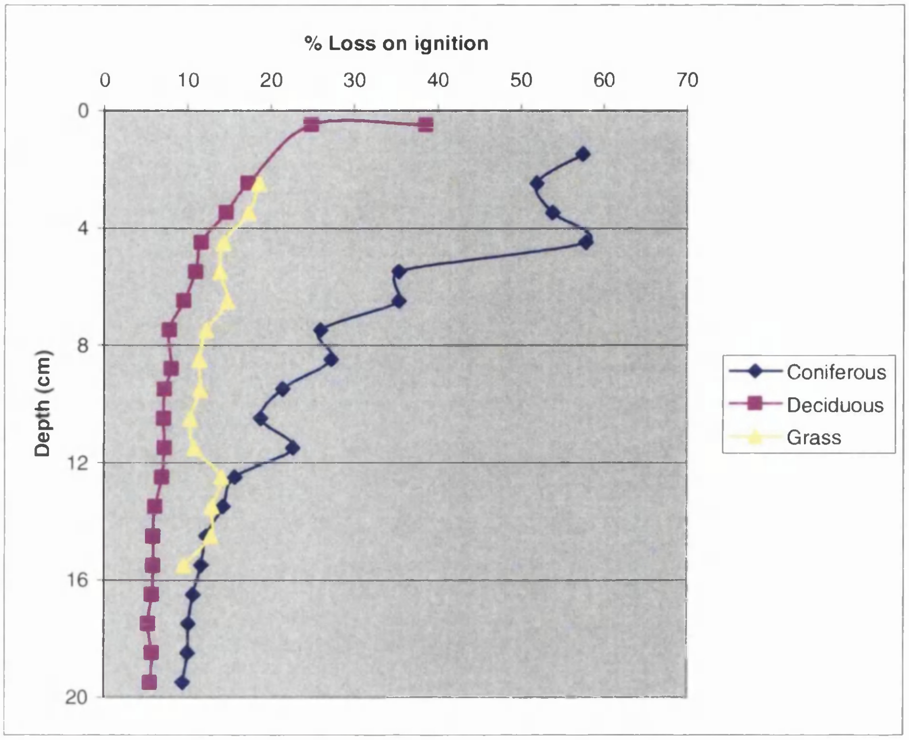


Figure 23 - % Loss on ignition for the coniferous, deciduous and grassland cores collected in Pollok Park, October 2000.

The pH of the deciduous and coniferous cores are very similar at the surface (Figure 24). The deciduous pH, however, then rises slightly and stays fairly constant down to 7 cm, with an average of 3.61. The pH of the coniferous soil decreases slightly down the profile, with an average of 3.38.

The pH of 6.0 at the surface of the grassland sample is much higher than corresponding values for both forest soils. The pH of the grassland soil decreases slightly down the profile, but stays consistently above the pH of both the coniferous and deciduous soils. This is consistent with the large input of organic material to the two forest soils. This organic input provides a large supply of humic acids which will lower the pH and the relatively lower mineral content of these soils will reduce the buffering capacity by clays (VanLoon and Duffy, 2000).

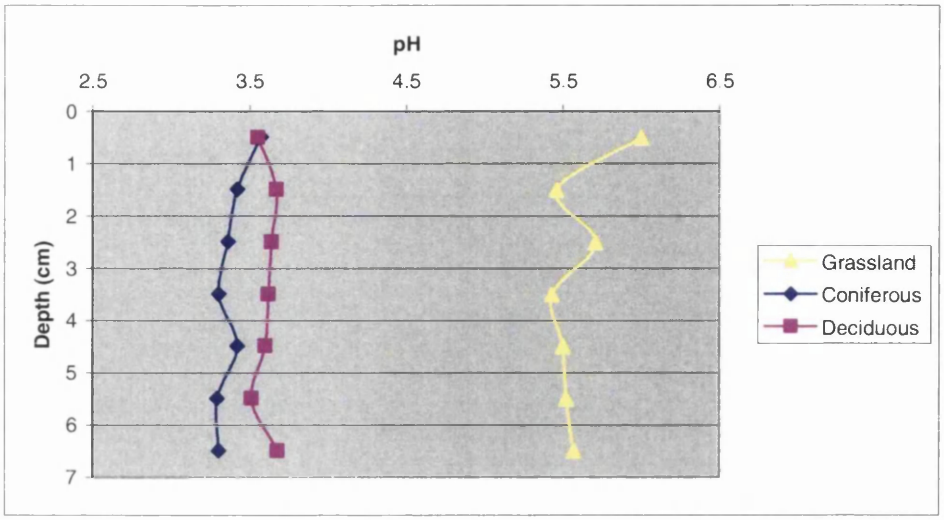


Figure 24 - pH in short cores of deciduous, coniferous and grassland soil

Radionuclide distributions and inventories

^{210}Pb

Input of ^{210}Pb to the study area is dominated by natural atmospheric input from decay of ^{222}Rn. Enhancement of ^{210}Pb in the soils from anthropogenic sources is possible and sources include atmospheric deposition of lead from the combustion of coal and industrial activities such as smelting. Lead in leaded petrol is of 'geological' age and is therefore unlikely to be a major source of ^{210}Pb (MacKenzie, 2000).

All three soils have their highest specific activities at the surface (Figure 25). The coniferous soil shows a general decrease in ^{210}Pb activity with depth, but an irregularity in the profile, with an anomalously low value at approximately 2 cm, indicating heterogeneity in the soil composition.

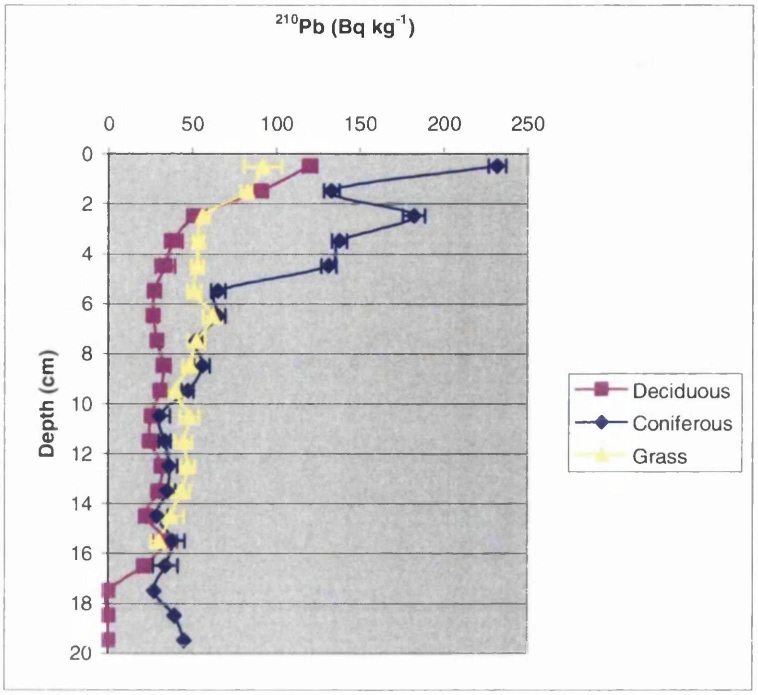


Figure 25 - Total ^{210}Pb specific activity (Bq kg^{-1}) in deciduous, coniferous and grassland cores, October 2000.

Unsupported ^{210}Pb (Figure 26) is detected down to a depth of between 9 and 10 cm in the coniferous soil. The deciduous profile decreases smoothly and always stays lower than the specific activity in the coniferous soil. In the case of the deciduous core, unsupported ^{210}Pb is detected only to a depth of between 4 and 5 cm. Unsupported ^{210}Pb was detected in the grassland soil down to between 8 and 9 cm.

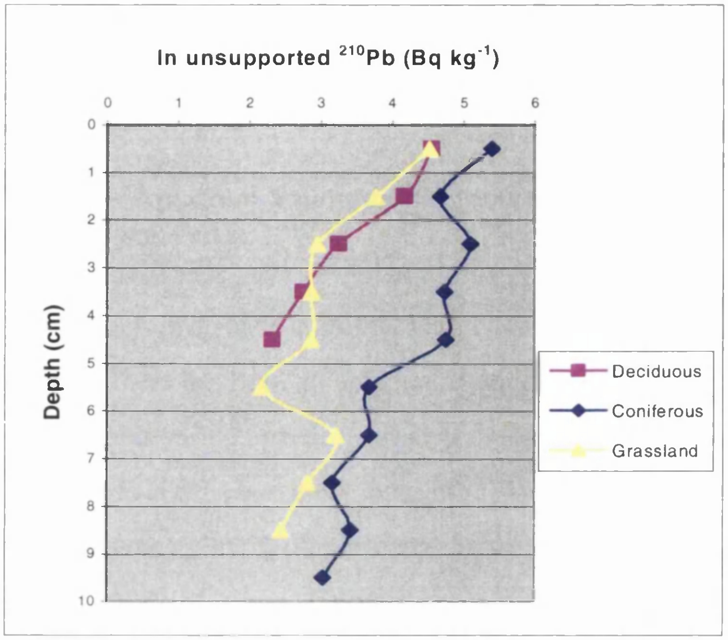


Figure 26 – ln unsupported ^{210}Pb in the coniferous, deciduous and grassland cores, October 2000

The empirical results show that the unsupported ^{210}Pb decreases exponentially with increasing depth. There are two possible explanations for this exponential decrease.

i) The soil could be accumulating as is observed for ombrotrophic peat deposits (MacKenzie et al., 1997). Or,

ii) Downward migration of ^{210}Pb may be occurring.

These possible processes are considered below.

Accumulation:

Apparent accumulation rates were calculated as follows. Assuming the rate of deposition of ^{210}Pb and the rate of accumulation are constant, gives S/λ as a constant (S= accumulation rate) and the graph of ln ^{210}Pb (x-axis) against depth (y-axis) is of the form $y = mx + c$, i.e. a straight line graph with the gradient $m = -S/\lambda$ (Appleby and Oldfield, 1992).

Apparent accumulation rates were calculated for the three soils and are shown in Table 36.

Soil Type	Accumulation rate (cm y^{-1})
Deciduous	0.052
Coniferous	0.099
Grassland	0.133

Table 36 – Apparent accumulation rates (cm y^{-1}) for the deciduous, coniferous and grassland soil cores.

The buildings and roads in Pollok Park have been at the same level for several hundred years and there has been no apparent change in the level of the soil surface relative to these features. This would appear to make it unlikely that the soils are accumulating. The soils, however, do receive a regular input of organic matter and could possibly be accumulating and undergoing compaction, leaving the surface level relatively unaltered. This requires further investigation and is further discussed later.

Accumulation rates in the literature for peats in Central Scotland are of the order of 0.07-0.08 cm $year^{-1}$ (MacKenzie et al., 1997, 1998) and compare fairly well with the apparent accumulation rates calculated for the deciduous and coniferous soils. From the calculations, the grassland soil seems to have a slightly higher apparent accumulation rate.

Migration:

If migration is the process operating then, since the ^{210}Pb profile follows an exponential trend, it can be modelled as a steady state diffusion process. This allows the calculation of an effective diffusion coefficient as follows:

Diffusive mixing generates a profile of the form:

$$A_z = A_s e^{-kz}$$

Where $k = (\lambda / D)^{1/2}$
(D = Diffusion coefficient)

Again, assuming steady state conditions this is a straight line graph with the gradient $-1/k$ (Huh and Kadko, 1992). The gradient from the plot of ln unsupported ^{210}Pb verses depth was again used to calculate effective diffusion coefficients for the three soils as shown in Table 37.

Soil Type	Diffusion Coefficient (cm^2 yr^{-1})
Deciduous	0.09
Coniferous	0.33
Grassland	0.59

Table 37 –Effective diffusion rates (cm^2 yr^{-1}) for the deciduous, coniferous and grassland soil cores.

The effective diffusion coefficients imply that the fastest diffusion occurs in the grassland soil, which could suggest that lead exists in more mobile forms in this soil. It is likely in the case of the grassland soil that mixing will occur at the top of the core due to root action and soil faunal activity.

It is not possible, on the basis of the ^{210}Pb profile alone, to distinguish between accumulation of the soils or migration of ^{210}Pb. However, comparison with the distribution of other radionuclides can be used in this context as discussed later.

The ^{210}Pb inventories for each soil type were calculated as outlined in section 3.2, and are summarised in Table 38.

Soil Type	^{210}Pb Inventory (Bq m^{-2})	Average ^{210}Pb Flux (Bq m^{-2} y^{-1})
Deciduous	1117	34
Coniferous	3966	119
Grassland	1573	47

Table 38- Summary of soil ^{210}Pb inventories and flux from the deciduous, coniferous and grassland cores

The inventory for the coniferous soil core is comparable to those for ombrotrophic peat cores from central Scotland, which are regarded as providing good estimates of the inventory derived from atmospheric deposition of ^{210}Pb (MacKenzie *et al.*, 1998).

e.g. Flanders moss - Inventory: 3531 Bq m^{-2}
Flux: 110 Bq m^{-2} y^{-1}

Fenwick moor -Inventory: 3380 Bq m^{-2}
Flux: 106 Bq m^{-2} y^{-1}

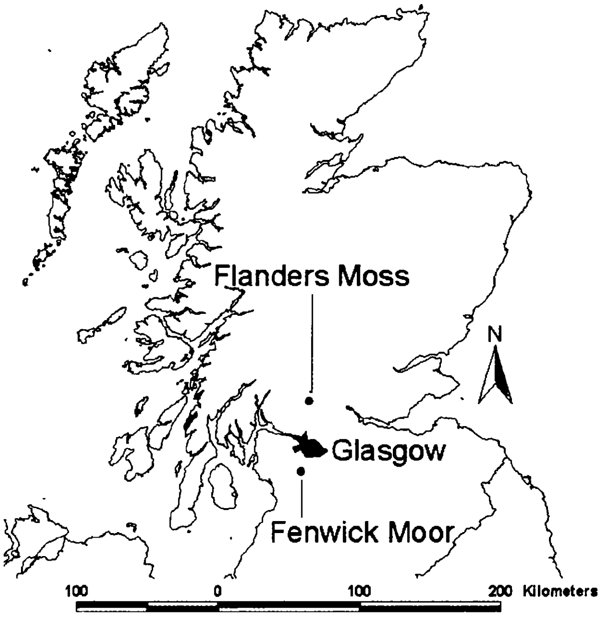


Figure 27 – Map of Scotland showing the locations of Flanders Moss and Fenwick Moor in relation to Glasgow

The deciduous and grassland ^{210}Pb inventories, however, are significantly lower than the inventory for the coniferous site. This suggests that the deciduous and grassland systems are losing lead by a factor of approximately three. There are a number of factors that could be causing this loss. Firstly, the deciduous forest canopy is very different from the coniferous canopy due to the seasonality of the deciduous trees and the difference in surface area of the leaves. The initial interception of atmospheric particles by the canopy may have an effect on the overall behaviour of lead in the two systems. The coniferous soil has the same degree of protection from the canopy all year round as opposed to the deciduous soil, which is exposed over autumn and winter. This could allow the falling rain to have more of an influence on the deciduous soil and promote faster leaching of lead. This increased exposure relative to the coniferous site would affect the grassland site all year round thus potentially contributing to the higher diffusion coefficient and lower lead inventory.

Pb is complexed by organic matter and the breakdown of organic matter may be an important mechanism controlling Pb transport in soils via organic colloids (Wang and Benoit, 1996). Although the % organic matter in the grassland soil is fairly high, it is *relatively* lower than the other two soil types. This lower % organic matter in the grassland soil *may* result in a lower Pb binding capacity and the higher rate of organic matter breakdown in the deciduous soil may promote Pb movement via the organic colloid transport mechanism. The lead which is leached through the profiles may then be removed at depth by groundwater or may be immobilised in the sub soil, beneath 20 cm, and thus would not have been detected by this research. In addition, the type of organic matter between the three sites is likely to differ as each site receives input from different types of organic material. For example the coniferous site will receive input of resistant coniferous leaves which are hard to breakdown, whilst the grassland site will receive roots and grass which are relatively less tough and easier to break down. These differences in organic matter type may influence Pb retention.

Although the grass on the grassland site is cut, it is not removed from the site, and therefore this would not represent a significant pathway for loss of lead from the system.

The greater exposure in the deciduous system and the greater surface area of the leaves may result in the wind blowing more of the deciduous leaves away and therefore removing the lead. Differing rates and types of microbial activity may also have a part to play in the different ^{210}Pb inventories. Soil microbes and fauna, in particular earthworms, in the deciduous system greatly assist with the breakdown of organic matter and may promote movement of radionuclides in the system. This, in turn, may make the radionuclides more readily lost by leaching.

^{137}Cs

The ^{137}Cs profile in the deciduous soil exhibits a smooth exponential decrease with increasing depth, strongly suggestive of downwards migration in a diffusion like process (Figure 28). As noted above, there is considerable uncertainty over the validity of the apparent accumulation rates derived from the ^{210}Pb data. However, these accumulation rates can be used to estimate ages of specific horizons in the soil that would apply if accumulation is occurring. Such ages can then be compared with known dates of input of other species such as ^{137}Cs. On this basis, the deepest point at which ^{137}Cs is detectable (6.5 cm) would equate to 1876 using the apparent ^{210}Pb deciduous accumulation rate of 0.052 cm y^{-1}. This date is obviously much earlier than the introduction of ^{137}Cs to the environment, so it points to the fact that the ^{137}Cs has undergone significant migration even if the apparent accumulation rate for the deciduous soil is correct.

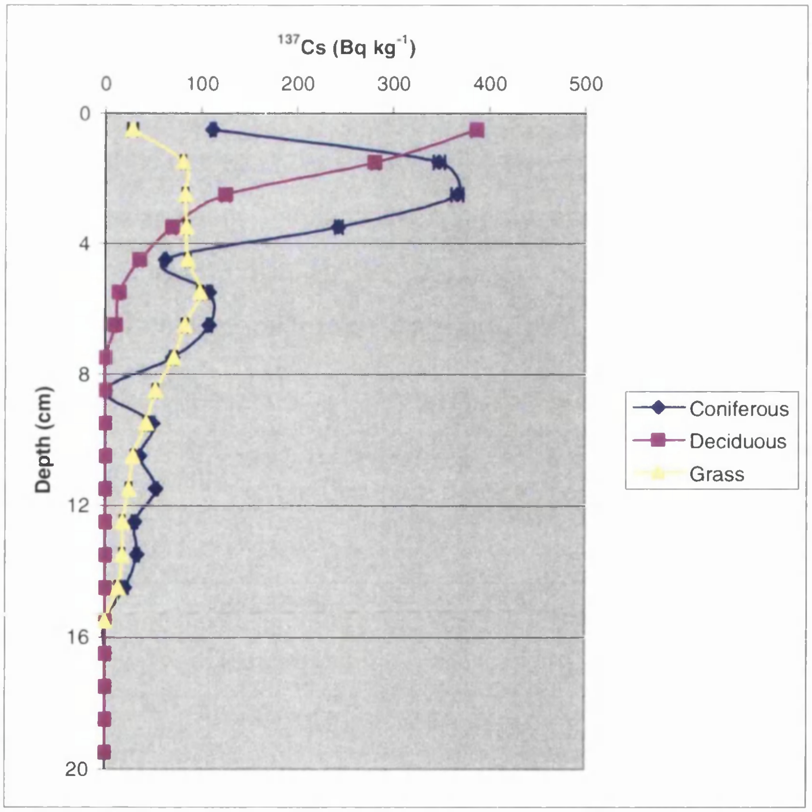


Figure 28 – ^{137}Cs (Bq kg^{-1}) in deciduous, coniferous and grassland cores.

The highest specific activity found in the coniferous profile is at a depth of between 2 and 3 cm with another small sub-surface peak at a depth of approximately 6 cm. This shape of profile could be attributed to two things. The first is heterogeneity in the physicochemical features of the coniferous soil. Different layers in this heterogeneous soil may create a geochemical barrier and thus could increase accumulation of radionuclides in selected soil layers, as was found in a study by Fesenko et al. (2001). The second possible interpretation of this profile shape is that it may reflect the maximum inputs of ^{137}Cs resulting in two peaks representing the 1986 Chernobyl reactor accident, and 1962-3, the peak of atmospheric weapons testing fallout. To investigate this further, the apparent ^{210}Pb accumulation rate of 0.099 cm^2 y^{-1} was used. This calculation implied the peak at 2 cm would equate to approximately 1980 whilst the peak at 6 cm would equate to approximately 1940. If the deepest point where ^{137}Cs is observed in the profile (14.5 cm) is treated in the same way, this would equate approximately to 1854. This date, along with the estimated date of 1940 for the lower peak, is well before the introduction of ^{137}Cs to the environment and again indicates downward migration of ^{137}Cs.

The grassland ^{137}Cs profile is consistent with the ^{210}Pb profile in that the profile is probably influenced by mixing at the surface. This may explain the low value at the surface of the core. If the apparent ^{210}Pb accumulation rate of 0.133 for the grassland soil was used to calculate an approximate age for the deepest point where ^{137}Cs is detected in the core (14.5 cm). This would approximately equate to 1892, which again is indicative of downward leaching of ^{137}Cs.

Inventories of ^{137}Cs in the three soil types were calculated and are shown in Table 39.

Soil Type	**^{137}Cs inventory (Bq m^{-2})**
Deciduous	4906
Coniferous	6561
Grassland	5364

Table 39 – ^{137}Cs inventory (Bq m^{-2}) in deciduous, coniferous and grassland cores.

Interestingly, the coniferous soil shows a higher ^{137}Cs inventory than the deciduous soil, which corresponds to the trend observed for ^{210}Pb inventories. All three ^{137}Cs inventories were broadly comparable to the value of 3700 Bq m^{-2} given by Cawse and Horrill (1986) for 0-30 cm ^{137}Cs inventory measurements in the UK. The inventories measured in 2001 in the coniferous, deciduous and grassland soils would have received input of ^{137}Cs additional to this from the Chernobyl accident in April 1986 and are therefore slightly higher than the value of 3700 Bq m^{-2} published by Cawse and Horrill.

^{241}Am

^{241}Am was not generated in significant quantities during weapons tests and its presence in the environment can be attributed to decay of fallout-derived ^{241}Pu. In both the coniferous and deciduous soils, the results are so close to the limit of detection that it is difficult to draw firm conclusions from the ^{241}Am profiles (Figure 29). However, the presence of the ^{241}Am in the coniferous sample at 1-3 cm indicates that the ^{137}Cs peak observed at this depth is not solely a Chernobyl peak. ^{241}Am is detected further down the profile in the case of the coniferous soil, which is consistent with other nuclides. If the ^{241}Am peak is taken to be at 3.5 cm and is dated using the apparent ^{210}Pb coniferous accumulation rate, the peak equates to 1965, which is close to the maximum input of ^{241}Am to the environment due to atmospheric weapons testing.

If the apparent ^{210}Pb accumulation rate is used to investigate the lowest detectable point of ^{241}Am in the deciduous core (2.5 cm), this equates to 1954 which is a viable estimate for the start of weapons testing and therefore the introduction of ^{241}Pu into the environment. The ^{241}Am data could therefore be taken to support the hypothesis that the forest soils are accumulating.

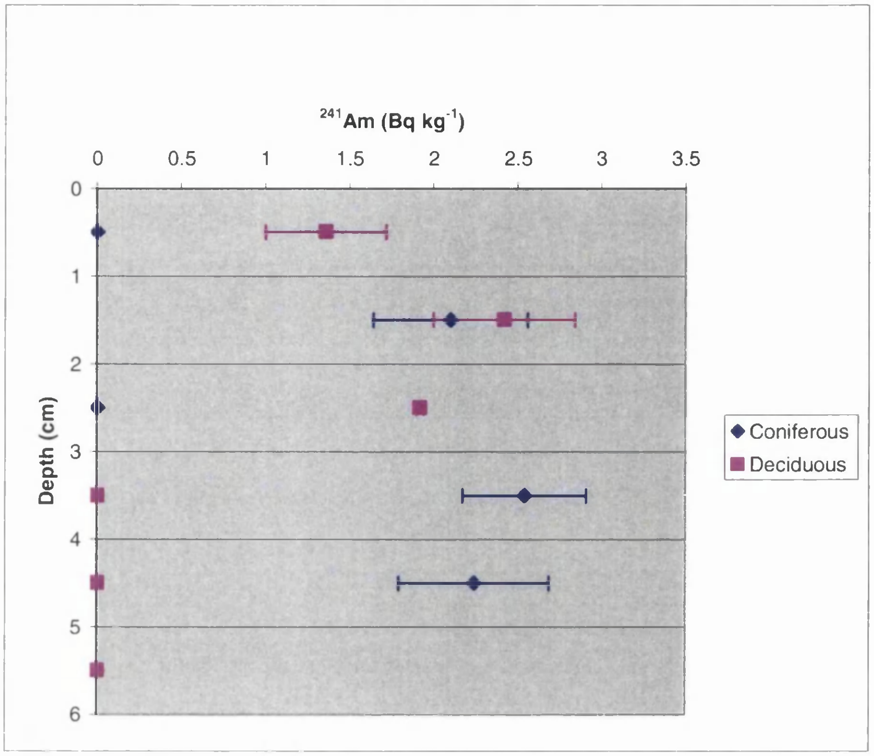


Figure 29 – ^{241}Am (Bq kg^{-1}) in deciduous and coniferous cores.

Pu isotopes

The ^{239,240}Pu in the coniferous soil shows a sub-surface peak from approximately 2.5 cm – 4.5 cm, corresponding to the most organic subsurface part of the profile. This was consistent with other work in that most of the plutonium held in soil is associated with the organic phase (Skipperud et al., 2000; Bunzl et al., 1992; Livens and Baxter, 1988). The ^{238}Pu/ ^{239,240}Pu ratio for the 5-6 cm grassland core section indicated contamination of the sample in the laboratory and the increase in Pu concentration at this depth probably does not represent a true peak in the core. Therefore, excluding this sample, grassland plutonium profiles show consistently low values. Due to this contamination and the lack of Pu results for 0-2 cm section of the grassland core, an inventory was not calculated for this soil type. ^{239,240}Pu inventories (Table 40) were 47 Bq m^{-2} and 89 Bq m^{-2} for the deciduous and coniferous cores, respectively. This follows the same pattern as the % organic content which may explain the difference in inventories. The ^{239,240}Pu inventories were however both comparable to the ^{239,240}Pu inventory of 67 Bq m^{-2} given by Cawse and Horrill (1986). ^{238}Pu inventories (Table 41) showed a similar pattern as the ^{239,240}Pu inventories with the deciduous inventory being lower than that of the coniferous. Cawse and Horrill (1986) gave a UK ^{238}Pu inventory value of 4.8 Bq m^{-2}, measured in 1977. Both the coniferous and deciduous inventories are slightly lower than this value, owing to a degree of decay of the ^{238}Pu. When this value is decay corrected to 2001, the ^{238}Pu inventory value is 3.9 Bq m^{-2}, which is of a similar magnitude to the values obtained for the coniferous and deciduous cores in Pollok Park.

Soil Type	**^{239,240}Pu inventory (Bq m^{-2})**
Deciduous	47
Coniferous	89

Table 40 - ^{239,240}Pu inventory (Bq m^{-2}) in deciduous and coniferous cores.

Soil Type	**^{238}Pu inventory (Bq m^{-2})**
Deciduous	1.3
Coniferous	2.8

Table 41 – ^{238}Pu inventory (Bq m^{-2}) in deciduous and coniferous cores.

Using the coniferous apparent ^{210}Pb accumulation rate, the ^{239,240}Pu peak in the coniferous profile at 3.5 cm would approximately equate to 1965 and the ^{238}Pu peak at 3.9 cm would equate to approximately 1962. As this was a time of significant release of Pu to the environment, it would appear as if the coniferous ^{210}Pb accumulation rate is feasible.

However, the lowest detectable ^{238}Pu and ^{239,240}Pu at 7.5 cm would equate to approximately 1925, which is earlier than the introduction of plutonium into the environment and therefore refutes the apparent agreement between the ^{210}Pb and Pu profiles. If the deciduous ^{210}Pb accumulation rate was applied to the lowest detectable point in the deciduous ^{239,240}Pu profile (9.5 cm), this would equate to approximately 1819 and the lowest point of ^{238}Pu (6.5 cm) would approximately equate to 1876. This indicates that either the deciduous ^{210}Pb accumulation rate is not correct (i.e. the soil is not accumulating), or the Pu is migrating down the profile.

The lowest detectable ^{239,240}Pu and ^{238}Pu in the grassland soil (7.5 cm) would equate to 1944 when the ^{210}Pb grassland accumulation rate of 0.133 $cm^2\ yr^{-1}$ is applied. Again, this date is too early suggesting that the grassland core is not accumulating.
This evidence indicates that either the apparent ^{210}Pb accumulation rates for the three soils types are not correct (i.e. the soils are not accumulating), or the plutonium is migrating down the profiles. The latter is more probable, but this does not necessarily indicate that plutonium is migrating in solution, since bioturbation could be contributing to this movement.

The expected ^{238}Pu/ ^{239,240}Pu fallout activity ratio for 2001 is approximately 0.029. The ^{238}Pu/ ^{239,240}Pu activity ratio varies slightly down each profile but, in the case of the deciduous soil type, the average of the measured values is 0.029 and so agrees very well with the expected value. The coniferous soil ^{238}Pu/ ^{239,240}Pu activity ratio is slightly higher than the deciduous, but stays approximately within error of the expected fallout ^{238}Pu/ ^{239,240}Pu activity ratio of 0.029. As previously mentioned, the 5-6 cm grassland sample ^{238}Pu/ ^{239,240}Pu activity ratio indicated the presence of contamination and so must be discounted from any discussion of the ratio. The other grassland samples did vary but, like the coniferous samples, did stay approximately within error of the expected value.

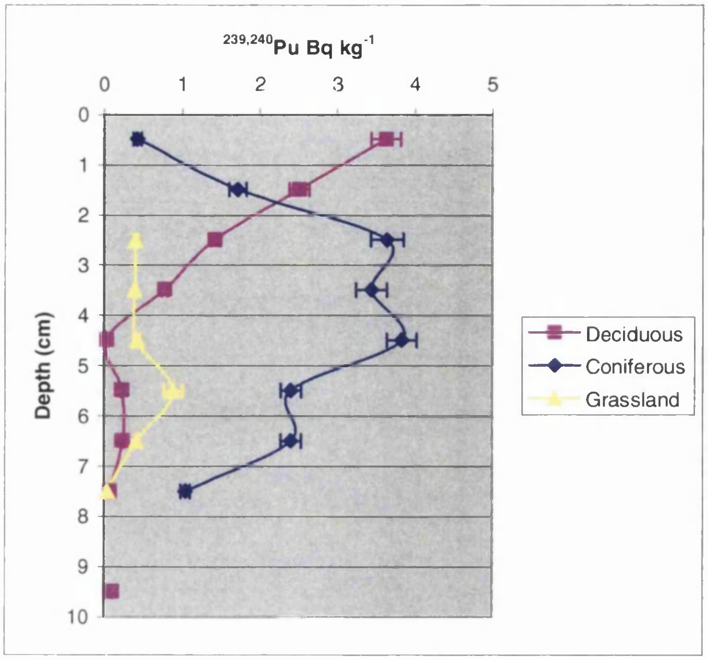


Figure 30 – ^{239,240}Pu (Bq kg^{-1}) in deciduous, coniferous and grassland soil

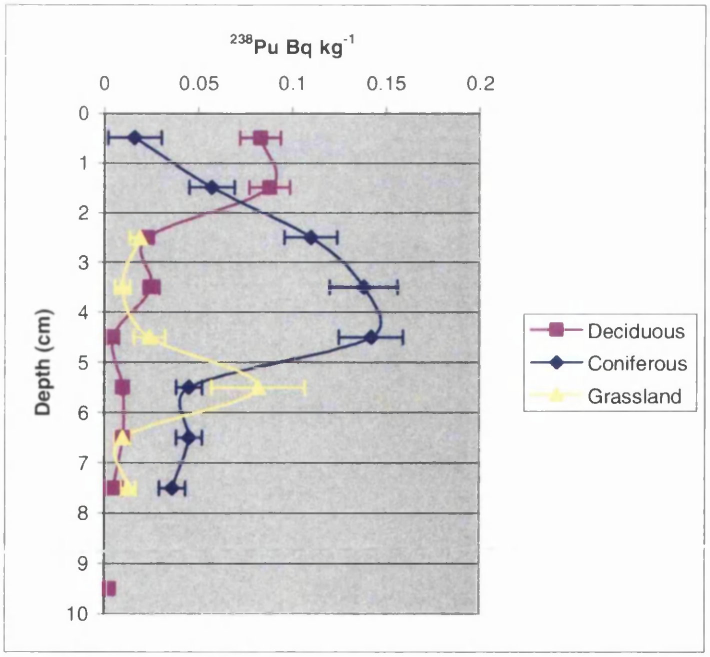


Figure 31- ^{238}Pu (Bq kg^{-1}) in deciduous, coniferous and grassland soil

To bring these results together, all three cores suggested the downward movement of ^{137}Cs. ^{241}Am in the coniferous core at depths of 1-3 cm indicates that the ^{137}Cs peak at this depth is not solely a Chernobyl peak. The apparent ^{210}Pb accumulation rates calculated for the coniferous and deciduous soils are not consistent with the times of plutonium release to the environment. It can therefore be concluded that the coniferous and deciduous soils are not accumulating. It is more likely that the exponential decrease observed in the unsupported ^{210}Pb is caused by diffusion of lead.

The grassland soil exhibited a profile indicative of mixing, probably due to disturbance from roots and soil fauna. No ^{241}Am was detectable in the grassland core. The ^{210}Pb accumulation rates calculated for the grassland core did not correspond to the ^{238}Pu and ^{239,240}Pu detected in the core and therefore it could be concluded that the grassland core is probably not accumulating.

Plutonium sequential extraction

As previously expressed, most of the Pu held in soil was expected to be associated with the organic phase. The sequential extraction (although subject to experimental errors) did highlight this association of Pu with the organic phase.

The sequential extraction results also highlighted that there were problems with the technique. It was expected to achieve both higher results and a positive reading from more of the samples. This may be due to the fact that the sequential extraction introduced many more steps into an already lengthy Pu extraction experiment. These steps gave more opportunity for the plutonium to be lost along the way. Also, as mentioned in section 2.3.4, this method was developed for more mineral soils and a small percentage of the sample was probably lost at each stage due to the highly organic nature of the samples. It can therefore be concluded that this method would need considerable revision to achieve better results on such organic soil.

Chapter 5. Conclusions

The main findings from this research are:

- ^{40}K specific activities generally decreased in the order grass> street dust > soil. The ranges observed in ^{40}K specific activities were 197-2115 (Bq kg^{-1}), 189-1000 (Bq kg^{-1}) 225-479 (Bq kg^{-1}), in grass, street dust and soil, respectively. The variability in the ^{40}K specific activities also decreased in this order. Some of this variation has been attributed to environmental management of selected sites. For example, the Pollok Park fertilised grass sample had a ^{40}K specific activity of 1910 ± 150, and the Pollok Park unfertilised grass sample had a ^{40}K specific activity below the limit of detection.

- ^{137}Cs specific activities ranged from 1.2- 22.5 (Bq kg^{-1}) in grass, 2.4-85.1 (Bq kg^{-1}) in soil and from below the detection limit to 211 (Bq kg^{-1}) in street dust. The ^{137}Cs street dust data population showed a broadly exponential trend with 2 outliers. These outliers may have been due to differences in sample composition (e.g. a high illitic clay content), but both were observed near potential point sources of ^{137}Cs.

- ^{7}Be, a cosmic ray- produced radionuclide, was detected in grass with specific activities in the range 46-530 (Bq kg^{-1}) and in street dust in the range 44-595 (Bq kg^{-1}). Considerable variation was observed both spatially and temporally, despite having a uniform atmospheric production rate and fairly uniform rainfall over the study area. ^{7}Be detected in soil (0-15 cm) was negligible, probably due to the fact that very little of the soil sampled would have been exposed to the input of ^{7}Be from the atmosphere via wet or dry deposition.

- The highest specific activities of ^{226}Ra , a daughter nuclide of ^{238}U, were observed in soil (11.9- 44.4 Bq kg^{-1}), with fairly consistent results being observed both spatially and temporally. The ranges of specific activities observed in grass and street dust were 1.45- 17.5 (Bq kg^{-1}) and 3.8 –22.2 (Bq kg^{-1}), respectively.

- ^{40}K was detected in the Clyde Estuary waters at levels consistent with those expected for estuarine waters. ^{131}I was detected in the Clyde Estuary, down stream of a hospital with a nuclear medicine facility. This, and the ^{137}Cs outliers in street dust, highlighted that this survey was sensitive enough to detect radionuclides that may be linked to a point source.

- The coniferous soil from Pollok Park was found to have the lowest pH and highest organic content and a ^{210}Pb inventory which was comparable to other ^{210}Pb inventories in the literature for Central Scotland.

- The deciduous soil also had a low pH and high organic content and had a ^{210}Pb inventory approximately a factor of three lower than the coniferous soil.

- An exponential decrease in unsupported ^{210}Pb specific activities with increasing depth in both the coniferous and deciduous soils could be attributed either to accumulation of the soils or to downward migration of the ^{210}Pb. However, the depth distributions of ^{239,240}Pu in both cases were inconsistent with the apparent ^{210}Pb accumulation rates, indicating that the presence of natural and manmade radionuclides at depth in these profiles is almost certainly due to downward migration.

- The grassland soil had a higher pH and lower organic content than both the forest soils and had a ^{210}Pb inventory comparable with the deciduous soil core. The grassland core exhibited a profile indicative of mixing, and accumulation of this soil is unlikely.

- All three cores from Pollok Park suggest the downward movement of ^{137}Cs.

- ^{137}Cs inventories for the deciduous, coniferous and grassland soils and the ^{239,240}Pu and ^{238}Pu inventories for the coniferous and deciduous soils were broadly comparable to values for inventory measurements in the literature.

- Plutonium was found to be associated with the organic phase of the forest soils but considerable revision of the BCR sequential extraction technique would be needed in future for use on such organic soils.

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Appendix 1 – Extended results from gamma spectrometry analysis carried out at GCSS

Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	22.5± 6.5	~	22.4± 4.7	23± 1.2	25.6± 3.8	7.8± 0.5	27.9± 10	21.1± 1.28	20± 1.24	~
2 (Gartnavel Hospital)	19.3± 1.4	49± 16.2	16± 4.8	19.6± 1.1	19.6± 4.7	6.37± 0.49	28.5± 7.12	19.7± 1.35	19.3± 1.41	43.2± 7.77
3 (Strathclyde Uni/ Royal Inf)	22.5± 6.38	45± 19.8	28.2± 5	23.1± 1.2	20.4± 4.08	7.46± 0.46	~	17.9± 1.12	16.9± 1.14	~
4 (Victoria Infirmary)	30.1± 8.7	75.5± 18.12	31.55± 5.9	30.15± 1.51	31.2± 4.21	9.7± 0.56	11.56± 3.87	29.7± 1.69	27.85± 1.6	~
5 (Southern General)	36± 10.44	79± 22.12	39± 6.63	37.7± 1.88	39.7± 5.16	12.7± 0.66	40± 10.4	45.8± 1.14	43± 2.4	38± 17.1
6 Pollok Park (Average)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
7 (Colston Lab)	14.9± 4.47	23± 9.66	18.8± 4.51	14.4± 0.85	14.5± 4.06	5.34± 0.43	~	11± 0.91	12.7± 1.04	16.2± 4.53
8 (Summerston)	22.5± 6.52	56± 21.8	22.9± 4.35	22.5± 1.19	20.2± 3.84	7.29± 0.45	35± 15.75	21.1± 1.29	20.8± 1.31	~
9 Robroyston	41± 12	90± 19	40.7± 6.7	40.3± 21	35.5± 53	1.32± 0.46	33.5± 8	36.8± 2.1	37.8± 2.2	49.4± 7.8

Radionuclides in soil (Bq kg^{-1}), April 2001

Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	29.3± 8.8	88± 22.9	31.1± 6.3	30.3± 2.1	30.5± 5.2	10.2± 0.8	42.8± 8.6	30± 2.2	29.2± 2.2	43.7± 7.4
2 (Gartnavel Hospital)	20.4± 6.0	67± 15.4	23.2± 4.12	20.3± 1.4	19.9± 3.2	6.67± 0.5	31.3± 8.9	24.6± 1.8	23.2± 1.7	24± 15.8
3 (Strathclyde Uni/ Royal Inf)	22.7± 6.8	71± 19.9	17.8± 4.9	22.8± 1.6	24.2± 3.6	7.83± 0.6	28± 13.2	22.6± 1.6	21.9± 1.6	~
4 (Victoria Infirmary)	27.9± 8.3	78± 22	29.6± 6.5	28.5± 2.0	26± 5.6	0.98± 0.5	47.7± 9.5	30.1± 2.3	30.9± 2.5	50± 8.5
5 (Southern General)	27.3± 8.2	79± 21.3	30.8± 6.2	28.4± 1.9	31.7± 5.7	9.44± 0.7	44.2± 8.9	33.7± 2.5	33.8± 2.6	43.3± 7.4
6 Pollok Park (Average)	29.9 ± 8.8	66 ± 10.9	33.5 ± 6.1	30.6 ± 4.3	31.45 ± 8.9	5.2 ± 0.45	36.5 ± 9.6	27.5 ± 2.1	27.3 ± 2.1	16.0 ± 3.2
7 (Colston Lab)	19.1± 5.7	52.3± 16.4	18.36± 4.2	18.86± 1.3	17.1± 2.6	6.33± 0.5	28.03± 8.9	17.9± 1.4	17.26± 1.3	27.1± 2.7
8 (Summerston)	20.9± 6.27	65± 11.05	23.2± 4.64	22.3± 1.56	22.3± 3.34	7.22± 0.54	30.2± 8.75	22.3± 1.65	20.6± 1.52	~
9 Robroyston	24.2± 7.1	55± 16	23.5± 4.5	25.3± 1.8	26.6± 3.5	0.62± 0.18	19.8± 7.7	24.2± 1.8	22.7± 1.7	~

Radionuclides in soil (Bq kg^{-1}), November 2000

Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	~	2.6± 1.3	~	6.9± 1.9	~	~	~	~	~	~
2 (Gartnavel Hospital)	~	~	~	~	~	~	~	~	~	~
3 (Strathclyde Uni/ Royal Inf)	~	~	~	~	~	~	~	~	~	~
4 (Victoria Infirmary)	~	~	~	1.78± 0.6	~	~	39± 6.57	1.47± 0.10	1.43± 0.10	23.73± 1.49
5 (Southern General)	~	~	~	1.83± 0.9	~	~	~	~	~	~
6 Pollok Park	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
7 (Colston Lab)	~	~	~	~	~	~	~	~	~	~
8 (Summerston)	~	~	~	3.6± 1.8	~	~	~	~	~	~
9 Robroyston	~	~	~	~	~	~	~	~	~	~

Radionuclides in grass (Bq kg^{-1}), April 2001

Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	27.6± 9.66	~	~	13.3± 1.99	~	5.8± 1.33	~	14.6± 2.62	20.2± 3.23	~
2 (Gartnavel Hospital)	2.7± 1.56	29± 13.63	~	1.07± 0.42	~	~	24.2± 6.29	1.8± 0.61	2.24± 0.71	19.1± 5.92
3 (Strathclyde Uni/ Royal Inf)	~	~	~	~	~	3.9± 2.14	~	~	~	~
4 (Victoria Infirmary)	30± 11.7	~	~	9.8± 2.35	~	4.9± 1.71	~	15.4± 3.69	19.6± 4.50	~
5 (Southern General)	23.2± 8.81	~	~	14.3± 2.14	~	5± 1.4	~	15.9± 2.86	18.2± 3.09	~
6 Pollok Park (Average)	~	~	~	~	~	~	58± 23.78	~	~	~
7 (Colston Lab)	~	130.25± 63.17	~	0.78± 0.33	~	0.24± 0.15	109.5± 30.66	3.92± 2.19	7.76± 2.94	93.95± 35.96
8 (Summerston)	~	72± 36.72	~	1.99± 0.99	~	1.19± 0.63	33± 20.46	4.1± 1.18	6.3± 1.57	~
9 Robroyston	~	153± 64.26	~	6± 2.1	~	~	135± 32.4	7.1± 2.7	11.7± 3.62	113± 28.25

Radionuclides in grass (Bq kg^{-1}), November 2000

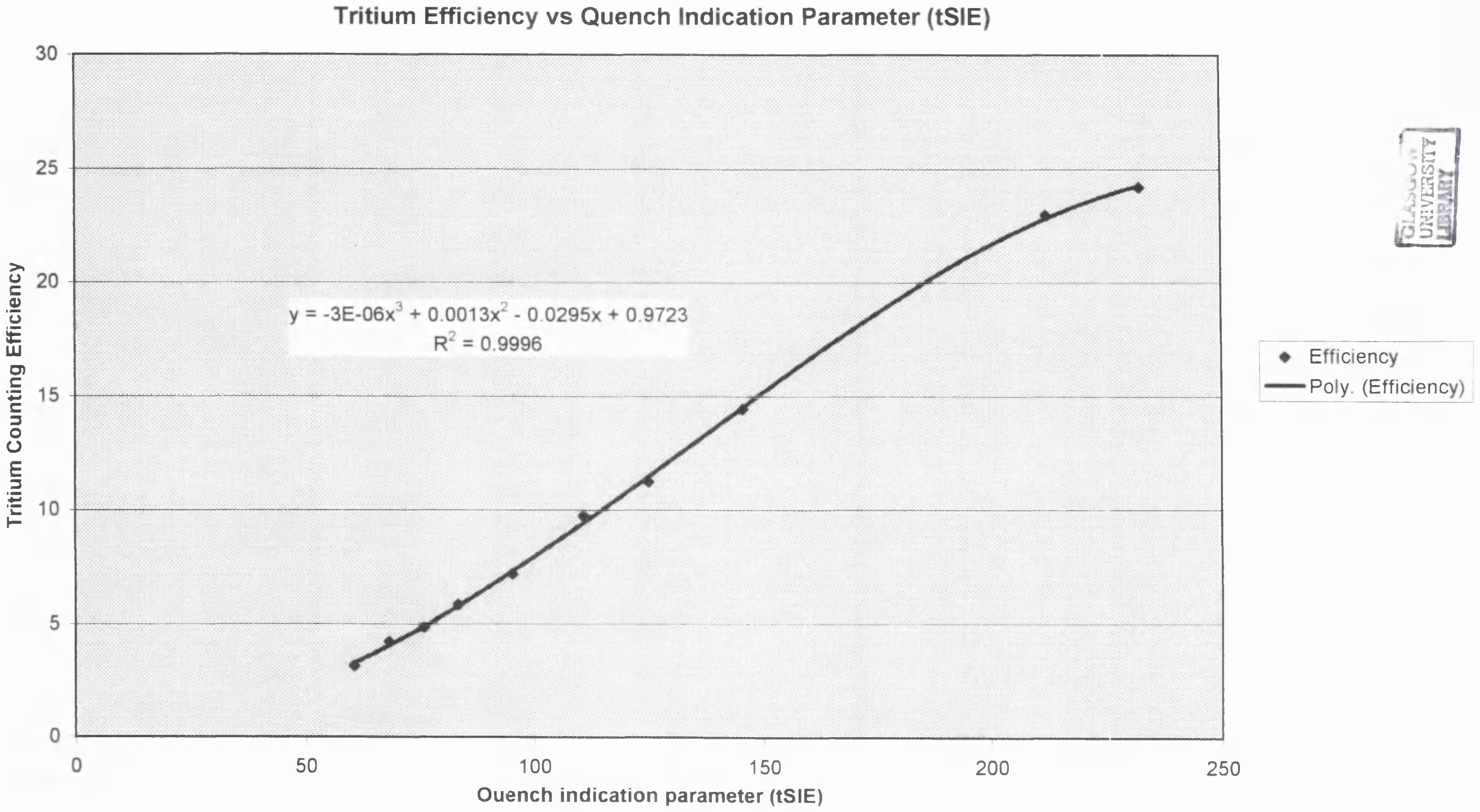
Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	12.5± 6.4	~	~	15.2± 2.2	~	4.9± 1.37	~	9.2± 2.94	~	~
2 (Gartnavel Hospital)	15.6± 7.48	~	~	11± 2.31	~	5± 1.65	126± 28.98	15.5± 3.1	14.6± 3.50	78± 21.06
3 (Strathclyde Uni/ Royal Inf)	11.8± 5.31	~	~	12.8± 1.66	~	3.8± 1.06	~	7.6± 2.28	~	~
4 (Victoria Infirmary)	23.6± 9.91	~	~	20.16± 0.60	~	8.33± 1.92	~	7.53± 1.356	~	18± 3.78
5 (Southern General)	18.6± 7.62	~	~	18± 2.34	~	5.7± 1.31	~	11.7± 3.04	11.4± 3.19	~
6 Pollok Park (Average)	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
7 (Colston Lab)	12.5± 5.87	~	~	13.4± 2.01	25± 3.25	5.2± 1.3	32± 11.84	7.7± 2.77	7.8± 2.96	170± 35
8 (Summerston)	11.6± 5.56	~	~	13.4± 1.742	~	4± 1.12	~	10± 2.2	~	~
9 Robroyston	28± 9.24	~	~	15.6± 1.7	30± 14.1	6.6± 1.12	~	~	16.5± 2.47	~

Radionuclides in street dust (Bq kg^{-1}), April 2001

Sample site	^{228}Ac	^{228}Th	^{224}Ra	^{212}Pb	^{212}Bi	^{208}TL	^{234}Th	^{214}Pb	^{214}Bi	^{210}Pb
1 (Glasgow Uni/ Western Inf)	22.8± 1.86	~	~	16.9± 2.36	~	6.9± 1.31	~	14.5± 2.61	16.5± 2.97	~
2 (Gartnavel Hospital)	18± 1.54	151± 69.46	~	14.9± 2.23	~	5.4± 1.56	9.4± 2.35	16.8± 3.36	20± 3.8	75± 21
3 (Strathclyde Uni/ Royal Inf)	19.6± 8.624	134± 46.9	~	16.9± 2.36	33± 20.46	6.8± 1.836	116± 26.68	17.1± 3.24	16.4± 3.44	67± 17.42
4 (Victoria Infirmary)	18± 7.4	97± 52	~	21.1± 2.6	25± 17	5± 1.2	45± 30	22.9± 3.2	21.6± 3.4	110± 64
5 (Southern General)	24± 2.208	157± 53.38	~	20.3± 2.6	~	2.8± 1.79	127± 27.94	20.6± 3.29	22.9± 3.89	83± 19.09
6 Pollok Park (Average)	18.8 ± 1.2	63 ± 26	~	18 ± 2.3	13.5 ± 9.0	4.1 ± 2.6	24 ± 14	13.3 ± 3.19	15.1 ± 3.9	~
7 (Colston Lab)	17.97± 1.536	46.75± 17	~	19.1± 2.725	16.5± 1.65	6.2± 0.113	22.5± 13.5	15.45± 3.8	7.425± 1.9	160.75± 45.75
8 (Summerston)	20± 7.3	129± 48	~	18.9± 2.1	~	5.5± 1.1	58± 2.7	20.2± 2.6	19.3± 2.9	~
9 Robroyston	~	~	~	20.8± 3.328	~	8.4± 2.436	~	11.4± 4.332	~	~

Radionuclides in street dust (Bq kg^{-1}), November 2000

Appendix 2 - Quench correction curve for ^3H analysis at GCSS


Tritium Efficiency vs Quench Indication Parameter (tSIE)
Tritium Counting Efficiency
30
25
20
15
10
5
0
0
50
100
150
200
250
Quench indication parameter (tSIE)
y = -3E-06x3 + 0.0013x2 - 0.0295x + 0.9723
R2 = 0.9996
Efficiency
Poly. (Efficiency)