Chapter (non-refereed)

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RADIONUCLIDE POLLUTION

Radioactivity in terrestrial ecosystems

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Summary
The types, sources and possible fate of radionuclides released into the Cumbrian environment are discussed. The difficulties associated with this type of environmental monitoring are outlined, and related to the research programmes currently being undertaken at the ITE Merlewood Research Station.

1 Introduction
The activities of man invariably change our natural environment, often in a physical way, by operations such as building, mining, quarrying, and farming. Materials are added to systems both as a supplement to those present naturally or, in some instances, in the release of entirely new materials and elements. Included in these materials are large amounts of low-grade energy released in the form of heat. To some extent, natural systems have an in-built resilience to the addition of energy and materials, yet at a certain point changes begin to take place which cause concern. It is these effects which the public associate with pollution.

Many pollutants are familiar and have reality: people can smell sulphurous fumes, they know car exhausts are dangerous, sewage is unpleasant to see and smell, pesticides and herbicides are familiar everyday items, even if they do have to be treated with some care. On the other hand, radioactivity is not detectable by the human senses. The general public associate radioactivity strongly with atomic weapons, and, indeed, for many this context was their first introduction to a knowledge of the subject. Often people are totally unaware of the different types of radiation and the intrinsic levels present in the environment due to primordial radionuclides deposited when the earth was formed. For instance, an average person contains about 4.44 kBq (kilobecquerel) of naturally occurring potassium-40 (Eisenbud 1973) and, consequently, is emitting about 475 fairly energetic gamma rays every second. It is known that radiation can induce cancers as well as genetic effects, and there is much doubt concerning the minimum dose needed to cause such effects. Indeed, as stated in the Royal Commission on Environmental Pollution report (1976), it is often assumed that there is no threshold value and the effect of radiation is directly proportional to the dose, down to the lowest doses and dose rates. It is facts such as these, and the lack of everyday knowledge, which create a greater sense of alarm concerning radioactivity than that associated with many other pollutants.

2 Radionuclides in Cumbria
The sources of radiation to which the general public are exposed are summarized by Hughes and Roberts in their 1984 review, published by the National Radiological Protection Board (NRPB). Their diagram (reproduced here as Figure 1) shows the sources of radioactivity to which the members of the general public are exposed. The majority of sources are natural and account for 87% of the total, with a large proportion (11.5%) of the remainder being accounted for by medical exposures.

In Cumbria, we are particularly concerned with the 0.1% accounted for by nuclear discharges. The NRPB point out that liquid discharges from the British Nuclear Fuels plc (BNFL) site at Sellafield contribute a major portion of the population dose attributable to radioactive wastes.

Considering the problem as it applies to Cumbria, when the Windscale works were originally sited on the Cumbrian coast, it was probably assumed that the small amount of radioactive effluent discharged into the Irish Sea would become dispersed and diluted within its basin, and eventually pass out into the ocean. However, this predicted pattern of events has not proved as simple as it first appeared.

Emission takes place from the plant as airborne releases and liquid discharges. Airborne releases take place from the stacks on the site and are essentially ventilation air from the process plant. The main component of these releases is the inert gas krypton-85 which originates as a fission product, other materials being in low to nearly undetectable amounts. By far the largest environmental releases are discharged to the sea as low-level liquid waste from the fuel element storage ponds, the reprocessing plant, the laboratories and the Calder Hall power plant. It is worthwhile considering the materials contained in this effluent as described in the annual report (BNFL 1984) on radioactive discharges for 1983 (Table 1). The effluent contains a mixture of radioactive elements which produce all 3 types of radiation: alpha, beta and gamma. The radioactive elements present, of which 28 are listed, range from those already naturally present in the environment, such as cobalt and zinc, to elements never before existing in nature to any extent, such as plutonium and technetium. The half-lives range from a few weeks to thousands of years.

When this mixture of elements and compounds is

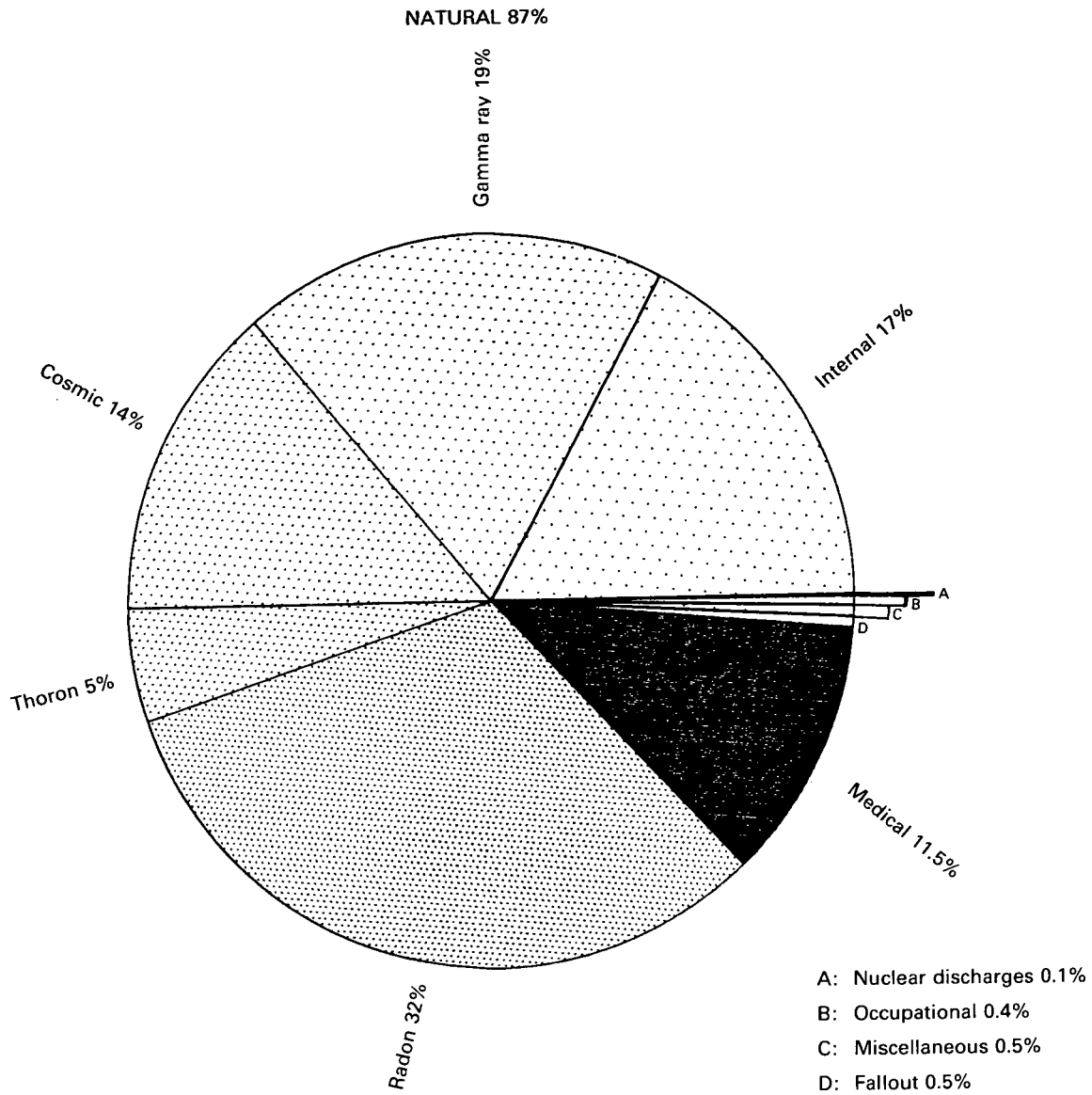

Figure 1. Sources of radiation to which the general public is exposed (source: Hughes & Roberts 1984)

Table 1. Radionuclides discharged by the Sellafield pipeline in 1983

	TB_q		TB_q
Cobalt-6	1.7	Neptunium-237	0.3
Zinc-65	<0.1	Plutonium-238	2.9
Strontium-89	<8.5	Plutonium-239+240	8.7
Zirconium-95	211.4	Americium-241	2.2
Niobium-95	385.1	Curium-242	0.4
Technetium-99	4.4	Curium-243+244	0.1
Ruthenium-103	18.5	Tritium	1831.1
Silver-110	<0.2	Sulphur-35	0.8
Antimony-125	17.6	Manganese-54	<0.2
Caesium-134	89.2	Iron-55	1.1
Caesium-137	1200.2	Nickel-63	1.1
Europium-152	<0.2	Iodine-129	<0.2
Europium-154	<0.5	Promethium-147	24.9
Europium-155	<0.6	Plutonium-241	330.9

released into seawater, a complex set of reactions takes place, which result in the majority of the radioactivity becoming tightly bound to the fine sediments (Hetherington & Jeffries 1974). Offshore of the plant is a large deposit of fine sediments, and recent work has shown that, contrary to original views, these sediments are not being significantly added to by sources such as other sea bed deposits, coastal erosion or material from rivers. Indeed, they are being extensively stirred up and reworked both mechanically and by animal life. The deposits are, in fact, serving as sources of material for deposition in the estuaries of the Irish Sea, such as at Ravenglass, the Solway and Morecambe Bay. Radioactivity is thus being concentrated and returned landwards.

A second pathway (Cambray & Eakins 1980) is indicated by the fact that plutonium of Windscale origin can be detected up to 10 km inland from the coast. It is postulated that, in a very thin micro-layer on the sea surface, radioactive substances are being concentrated and later released by the bursting of bubbles to form aerosols which can be carried inland by the prevailing winds.

Thus, instead of an elimination of radionuclides into the sea, we have mechanisms which are returning

these substances to the land, sometimes concentrating them in environmental materials. The purpose of the work carried out at Merlewood is to study the fate of these radionuclides when returned to the land. We need to know how these substances behave when they enter the terrestrial system, the factors that control their movement, any possible accumulation points in the system, and, of course, the quantities which find their way back to man, both in his food and his surroundings.

3 Radionuclide research at ITE Merlewood Research Station

I would like to outline briefly the problems the ecologist faces when trying to understand the processes involved in radionuclide transfer, and mention specifically some of the work in progress at Merlewood on Cumbrian systems.

There are many schemes representing the movement of radioactivity in ecosystems. A typical one is illustrated in Figure 2. The first problem is that of defining the source, which is a combination of natural, industrial emissions and bomb-test fallout. It is comprised of aerosols both dry deposited and washed out by rain, solid particles moved by wind and mechanical means, and gases diffusing through and being blown by the atmosphere. As a first stage, we need to know a great deal concerning the chemical and physical form of these inputs and the manner in which they interact with the many types of surfaces on which they impinge.

In the simplest of models, we can have a source, some form of resistance pathway and a destination. In general, a progressive dilution takes place as one passes along the chain. However, when biological factors are taken into account, this simple picture no longer holds. Well-illustrated examples exist, such as the series of reactions which convert inorganic lead compounds into biologically active methyl-lead in the environment and the progressive build-up of the radionuclide technetium in the trunks of forest ecosystems. The dilution effect may be reversed particularly when elements or compounds are essential to biological growth, and concentration in parts of the ecosystem can result. Deposition in particular tissues may take place, a classic example being the build-up of organochlorine pesticides in fatty tissue.

Returning to our ecosystem model, we then have to consider the complications introduced by multiple pathways. For instance, materials may enter a plant either directly by way of deposition and absorption through the leaf surface, or they may be deposited on the soil surface, pass through the soil system, be

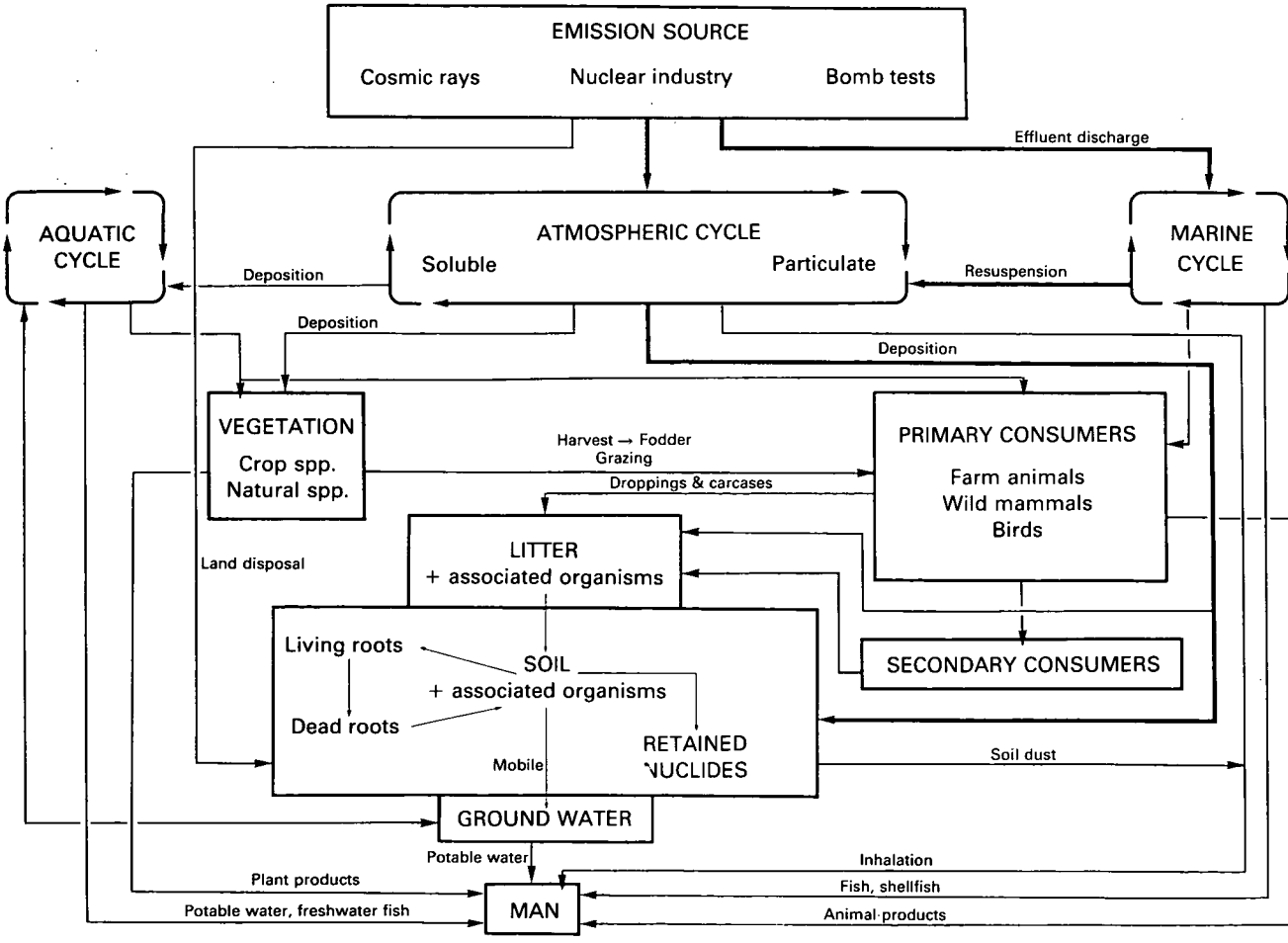

Figure 2. Diagram showing the pathways of radionuclides through the terrestrial environment, from source to man. The thicker lines denote the major routes

captured by the roots, and hence pass into the leaves. In any investigation, we have to be aware of the multiple pathways which exist in ecosystems. There is also the consideration of the different levels within any system, eg the series of food chains leading from the simplest unicellular biota via higher plants and animals to man. There would seem to be 2 possible ways in which these chains could operate. In the classic case of pesticides, there is a progressive build-up through the food chain to the top carnivores, such as the birds of prey. Alternatively, when the concentration factor is less than one, a progressive dilution of the pollutant takes place, particularly if it is a non-essential component of the diet.

A final complication encountered when undertaking studies in the natural environment is the inherent variability found in the systems and in the individuals. This variability is often combined with spatial and temporal fluctuations on both a large and small scale. Despite many attempts, it is not possible to classify biological systems and individuals into distinct units. The biologist has to accept that systems (and in many cases species) intergrade, and sharp boundaries do not exist. As a result, the ecologist has to take many samples in order to obtain reliable figures, and answers are often presented with large degrees of uncertainty, or as probability figures. These results never satisfy the man in the street, as he wants a simple yes/no answer every time.

I would finally like to outline the work programme at Merlewood. We have obviously had to select components of the terrestrial environment for study, and our work can be divided into 5 subject areas: the distribution of radionuclides in saltmarshes; the ingestion of radionuclides by sheep; the role of estuarine birds in the inland transfer of radionuclides; the dynamics of radionuclides in natural and agricultural systems; and plant uptake of radionuclides.

3.1 Distribution of radionuclides in saltmarshes

The saltmarshes and estuarine areas contain by far the largest deposits of radioactive materials and, indeed, these areas must in some respects be regarded as a secondary source of transfer to the inland systems. Concentrations of radionuclides fall dramatically at the tideline over a very short distance (*c*10 metres). For instance, in the Ravenglass area, concentrations of caesium-137 fall from an average of 16 798 Bq kg^{-1} in the surface silts of ungrazed saltmarsh to 40.0 Bq kg^{-1} in the pasture fields immediately behind. These values compare with a range obtained from control sites, such as the Humber and Portsmouth areas, of between 2 and 35 Bq kg^{-1}. The National Radiological Protection Board quote a Generalised Derived Limit (GDL) for soil of 3000 Bq kg^{-1}. It can be seen that, once the tidal limit is reached, levels of contamination are in the order of 1% of the GDL for that element. The deposition of radionuclides in saltmarshes can be related both to tidal immersion, half-life and the form of any vegetation present. In general, short half-life radionuclides such as niobium (35 days) and zirconium (65 days) show a simple deposition pattern related to the amount of tidal immersion. Long-lived material such as americium-241 (458 years) develop complex deposition patterns related to tides, vegetation density and vegetation form (Horrill 1983). For instance, under a bushy shrub of sea-purslane (*Halimione portulacoides*), over 5 times the concentration of americium-241 was found compared with the open mudflats, due to the trapping of particles by the plant.

3.2 Ingestion of radionuclides by sheep

Sheep are a common grazing animal in Cumbria both on the coastal plain and on the high fells. It is therefore appropriate to undertake studies on uptake by these animals. It became apparent during the studies how important the behaviour of the animals and the management practices of the farmer were in determining uptake, and also how seasonal fluctuations in radionuclide levels have to be taken into account. The biological half-life of caesium-137 in an animal is less than one month (Coughtrey & Thorne 1983), and sampling must be carefully matched with periods of food intake, if accurate transfer factors are to be calculated. The mean value for fresh muscle tissue for 3 animals from the coast was 42 Bq kg^{-1} of caesium-137, with a corresponding value for their 3 lambs, in September, of 71 Bq kg^{-1}. These values compare with a GDL for mutton and lamb of 10 000 Bq kg^{-1}. The higher concentration in the young animals is of interest, and further work is in progress to see if this is due either to the mother's milk being a readily absorbable source of caesium or to the young animal being a more efficient accumulator of radionuclides.

3.3 Estuarine birds and radionuclide transfer

If the estuarine areas are regarded as large reservoirs of radionuclides, then the possibility exists that coastal bird populations may act as vectors carrying radionuclides. A secondary interest is the fluctuation in numbers and breeding failures of the bird populations. It has been suggested, totally without evidence, that radionuclides may be the cause of these effects. Merlewood studies are therefore designed to answer these questions. Analyses of many samples of faeces and the tissues of the birds themselves have been carried out, and a joint study on behaviour is in progress with Durham University. Indeed, radionuclides are detectable in the faeces of birds feeding in the estuary, and reflect the place and feeding habits of the bird species. However, only 3 species (black-headed gull (*Larus ridibundus*), greylag goose (*Anser anser*) and starling (*Sturnus vulgaris*)) move from estuary to land to any extent, and the small number of birds and the low concentrations in the faeces indicate that amounts carried inland are very low indeed.

In the breast muscle of the birds, only the 2 isotopes of caesium (^{134}Cs and ^{137}Cs) were detectable and the concentrations were in the order of 70–110 Bq kg^{-1}

fresh weight, with an extreme range of 10–600 Bq kg^{-1} fresh weight. Very little activity, if any, was found in eggs from the Drigg Nature Reserve, less than 1 Bq kg^{-1} of caesium-137 being present in gull eggs, whilst crow eggs from the Eskmeals areas were lower than our limits of detection.

It is difficult to compare these sites with other areas at the moment, but studies in the USA have shown that waterfowl on disposal ponds can have up to 27 000 Bq kg^{-1} fresh weight of caesium-137 in their tissue with no apparent ill effects (Halford *et al.* 1981), whilst the GDL for eggs and chickens are 4000 and 10 000 Bq kg^{-1} respectively.

3.4 Radionuclides in natural and agricultural systems and plant uptake

Studies of radionuclides in natural and agricultural systems and on their uptake by plants can be conveniently brought together. The work aims to investigate the manner in which radionuclides pass through the terrestrial systems, to see if there are any accumulation points and to determine the driving and controlling mechanisms. This is a longer-term project and obviously depends greatly on the goodwill of landowners and farmers in permitting us access to their land. We are sampling a wide range of soils, vegetation and farm crops, as well as obtaining information on farming practices and management.

Initial results indicate that low levels of radionuclides are detectable in the inland systems, and we are concentrating our work on caesium-137 and the plutonium isotopes. Figure 3 shows some initial results for barley samples taken at increasing distances from the Sellafield plant ranging from 0.5 km to 14 km. Caesium-137 concentrations range from 44 Bq kg^{-1} dry weight to just above the level of detection of 0.26 Bq kg^{-1}; the GDL for grain is 1000 Bq kg^{-1}.

I would like to discuss one problem which has emerged from this work, ie soil contamination. The soil is the greatest reservoir of radionuclides. Movement through many soils is slow and the radionuclides accumulated over many years are still in the upper few inches of the profile. This material is resuspended by wind, rain and animals, and tests have shown that much of the radioactivity recorded is stuck to the

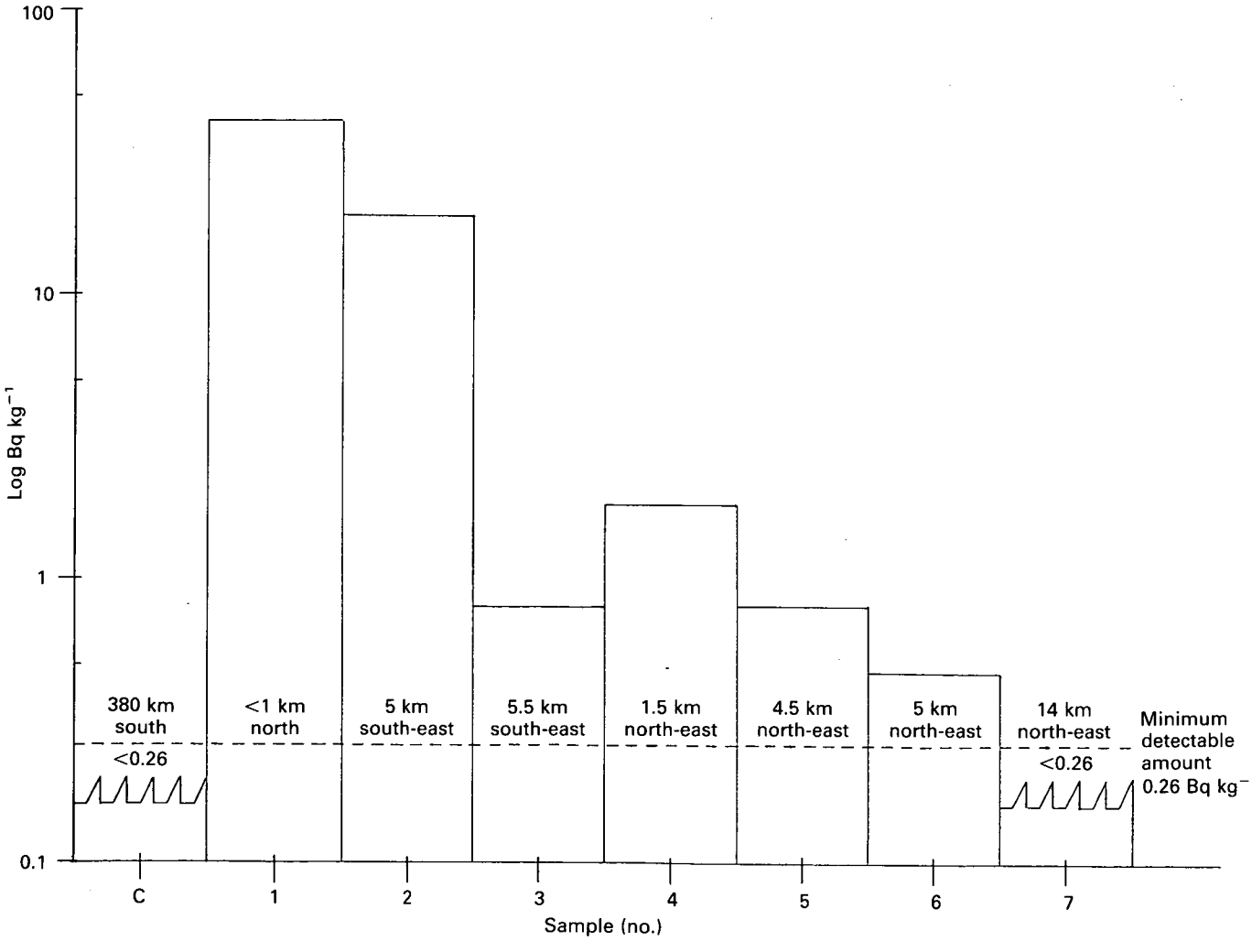

Figure 3. The levels of caesium-137 found in barley grown at varying distances from the Sellafield works. Sample C was a control taken from Taunton, Somerset

outside rather than within the plants. I suspect the same is true for barley, due to dust thrown up by the combine harvester.

We therefore need to know a lot more concerning the uptake by plant roots, in order that we can partition plant contamination into internal and external components. The internal component will be much more available for uptake by animals and man. We also need to know how the soil reservoir responds to environmental and man-made change, in case radionuclide materials fixed in the soil become remobilized and pass either into plants or to water systems.

To summarize, our research programme is designed to look at the fate of radionuclides entering sections of the environment. This information is needed both to identify and quantify the main radionuclide transfer routes to man, and will also help identify key points in the terrestrial system of Cumbria in case of accidental release.

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Radionuclides in Cumbria: environmental issues in the international context

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Summary
Radioactive pollution from the British Nuclear Fuels plc (BNFL) site at Sellafield (Windscale) has attracted concern within the scientific community internationally, but response from British scientists working in the field has been more muted. The responsible UK authorities have claimed that foreign conditions are not comparable and that, in radiological protection terms, the level of concern is not justified. This claim is examined in the light of international experience of reprocessing technology and accepted principles of radiological protection. It is concluded that there is no basis for the UK claim to special conditions and that the Cumbrian situation, unique as it is, reflects policy choices first made in the 1950s and continually reaffirmed in the face of mounting international criticism. The environmental issues are considered with regard to the health implications for Cumbrians, and broader perspectives in the quality of life.

1 Introduction
In 1955, at the UN Conference on 'Peaceful uses of nuclear energy', the international scientific community were first informed of a 'deliberate experiment' in Cumbria, which had taken the form of discharges of radioactive liquid effluent by pipeline into the Irish Sea (Dunster 1956). These experimental discharges were intended to elucidate the fate in the environment of a cocktail of radioisotopes with widely differing chemistries. Elsewhere, in nuclear weapon states which were also developing reprocessing plant for the large-scale production of plutonium (specifically the USA, USSR and France), a more cautious approach prevailed, namely to develop plant which would utilize maximal effluent treatment. Although this contrast of approaches must have been known to scientists involved in the UK nuclear programme, it was not widely discussed until the present controversy over discharges, with its international implications. This paper examines the development of concern for the environment in relation to the science and the politics of the developing controversy. Its aim is to place the response of the UK regulatory authorities in the broader context of international environmental standards, and to address the question of risk to the local population in Cumbria.