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Alpha-emitting 'hot particles' in the vicinity of BNFL Sellafield, Cumbria

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1. Introduction

In 1995, the UK Ministry of Agriculture Fisheries and Food (MAFF) commissioned a scoping study into the current presence and radiological significance of hot particles in the terrestrial environment and food chain close to the BNFL Sellafield site (Whittall & McDonald 1996).

This note describes the methods used to quantify the abundance of alpha-emitting 'hot particles' in the vicinity of Sellafield, and the results of characterisation work on two such particles.

2. Context

Hot particles may occur naturally. In igneous rocks, around 90% of the uranium and thorium is concentrated in accessory minerals such as apatite, fluorite, monazite, sphene or zircon, which usually constitute less than 1% of the total rock volume (Hamilton 1988). Weathering of the parent rock releases these dense, abrasion resistant minerals, typically as particles with mean diameters less than 50 μm.

Anthropogenic hot particles first entered the environment in the atomic detonations of 1945 (*e.g.* Adams *et al.* 1960) and continued to be released throughout the period of atmospheric nuclear weapons testing. A number of accidents at nuclear installations have also led to the release and dispersal of hot particles. Most notably, the Chernobyl disaster in 1986 distributed radioactive particles as far afield as Scandinavia (Raunemaa *et al.* 1987), Germany (Rudhard *et al.* 1992) and Bulgaria (Balashazy *et al.* 1988).

In the UK, between 1951 and 1957, routine operations at the Windscale plant, Cumbria, led to the release of radioactive particles into its immediate vicinity (Chamberlain 1987) and McMahon *et al.* (1994) describe irradiated uranium oxide particles collected from the Windscale area in 1956. After the 1957 fire, which resulted in the shutting down of the Windscale Piles, district survey teams found large numbers of radioactive particles (Dunster *et al.* 1958). More recently, evidence of uranium oxide particles was found in woodland close to Sellafield (Hursthouse *pers. comm.*), and Evans (1997) located alpha-emitting radioactive particles within the BNFL Sellafield site perimeter.

The origin and composition of hot particles released in the Chernobyl and Windscale accidents are discussed in greater detail by Salbu *et al.* (1994, 1998) and Sandalls *et al.* (1993).

3. Method

To quantify the abundance of alpha-emitting radioactive particles in the terrestrial environment in the vicinity of the BNFL Sellafield site, a suite of grass and soil samples was collected. After consultation with local farmers, sample sites for soil and grass were chosen from land that had remained unploughed for at least 30 years. This strategy was adopted to maximise the possibility of detecting particles emitted over the operational life-time of the plant.

At each location, surface scrapes of 0-2 cm depth and area of 0.4 m^2 were collected. Where possible, the underlying soil layer (2-3 cm) was also sampled (Table 1). Grass on the turves was returned to the laboratory, where the grass was trimmed to within 1 cm of the soil surface, collected and dried. The soil was dried, gently disaggregated in a pestle and mortar, and sieved to 2 mm to remove stones and roots.

Samples were also taken in a similar manner from three sites in Ladywood, a small coniferous wood to the north-east of the BNFL Sellafield site. The ground litter of this woodland had been reported to contain radioactive particles in a previous study (Hursthouse *pers. comm.*).

Efforts were then directed toward assessing the presence of alpha-emitting radioactive particles in components of the food chain. Current arable land-use in the Sellafield area is quite restricted. Wheat and barley are grown, but these crops are used solely for animal fodder. Potatoes are the only crop grown commercially for human consumption. Most of the local farmland is pasture for sheep and cattle. Though this might suggest that there is little risk of any alpha-emitting radioactive particles being consumed directly by humans, a habit survey of the local population (Stewart *et al.* 1990) found that, for more than half the sample group, the majority of vegetables consumed were home grown. Thus the consumption of home-grown vegetables may provide a route into the human food chain for radioactive particles.

Accordingly, vegetable plots were established close to the perimeter of the BNFL Sellafield site, in two cottage gardens and at the BNFL meteorological mast compound. A variety of leafy green vegetables were planted out in early July 1995. These consisted of ten plants each of broccoli, cabbage, spinach, lettuce and leek at each plot. Leafy vegetables were selected because they are more likely to intercept and retain freshly deposited or resuspended particles than either root vegetables or grain.

To assess whether or not BNFL Sellafield continued to be a source of fresh hot particles, two grass trays and a 'frisbee' - a large surface area deposition collector (1 metre diameter) - were also deployed at each site. Samples were collected from the frisbees and one of the grass trays at each site on a monthly basis over a four month period. The grass from the other tray was harvested at the end of the four months. These samples were supplemented by blackberries, barley and wheat collected from hedgerows and fields close to the Sellafield perimeter. The full suite of samples collected is described in Table 1. Sample locations ranged from 5 m to 1000m from the Sellafield site perimeter; most locations were within 500 m of the perimeter.

Sub-samples of the soil grass and litter, collected in the initial survey and the monthly sampling of the grass trays, were presented to sections of Kodak LR-115® cellulose nitrate alpha-radiation sensitive film (60 mm × 90 mm), in the form of dried, homogenised material, spread thinly upon adhesive mounting card. Good contact between sample and film was ensured by sandwiching the cards between perspex plates in a press for 4, 8 and 12 weeks.

The vegetable samples were sliced or broken up and then thoroughly washed in 3 litres of water, with the aim of removing particulate material adhering to the plant surfaces. The wash water was filtered (Whatman GF/C), the filters dried, and LR-115® film exposed to the dried filter paper for a period of 4 weeks. The vegetable material was then dried, homogenised and presented to LR-115® film as for the soil and grass samples. The grain samples were washed, and the washings filtered to obtain filter paper samples, whereas the blackberries were dried and homogenised to give card samples. The frisbee boards were rinsed thoroughly, and the rinse-water filtered and processed as for the vegetable rinse water. In this manner, every sample, apart from the washings of the vegetables and the frisbee boards, was analysed in triplicate. Table 2 provides a summary of the sample exposures.

After exposure, the LR-115® plastic exposed to each sample was developed by etching in sodium hydroxide solution. This process preferentially enlarges the sub-microscopic damage tracks produced as incident alpha-particles penetrate the plastic. Each plastic film was then examined under an optical microscope at ×20 magnification.

LR-115® exposures were repeated for any sample cards that produced dense track clusters on the first exposure. These second exposures were limited to 72 hours, to permit resolution and counting of individual damage tracks.

4. Results

No examples of the dense clustering of damage tracks that would indicate the presence of alpha - emitting radioactive particles were observed in any of the films exposed to samples from soils, leaf litter, vegetables, grains or blackberries, or from the frisbees and the grass obtained from the grass trays.

There was evidence of two particles in pasture grass samples which generated alpha-track densities above background. The samples were obtained from site 3 to the east of Sellafield (particle 1) and site 6 to the west of Sellafield (particle 2). Photographs of the 4 week exposures are presented in Figures 1a and 1b. Photographs were also obtained from the 72-hour exposures of the samples, which permitted the "burned-out" areas to be resolved into individual tracks (Figures 2a and 2b). Five-millimetre square sections, encompassing the burn-out zones in the LR-115® film, were cut from the two sample cards for examination under a Cambridge Instruments 360 scanning electron microscope. Initially, each sample section was viewed at low-magnification using back-scattered imaging (BSI). In this mode, the balancing of brightness and contrast highlights regions containing concentrations of heavy elements (Sandalls & Baker 1995). Electron probe microanalysis (EPMA), which involves the bombardment of a specific area with high-energy electrons and the analysis of the resulting x-ray spectrum, was then used to obtain information on the elemental composition of the two alpha-emitting particles.

4.1 Examination of particle no. 1

A secondary electron image of particle 1 is shown in Fig 3a. The image obtained using BSI is shown in Fig 3b. The particle is approximately 20 μm long by 5 μm wide, with a smooth surface. The EPMA spectrum revealed a peak corresponding to thorium. Clear signals for phosphorus, lanthanum and cerium were also detected.

Such an elemental composition suggests that the particle is of natural origin. Thorium, an alpha-emitting nuclide, is often found associated with the lanthanides, in minerals such as monazite, a phosphate of thorium, lanthanum and cerium. This particle may originate from the use of phosphate based fertilisers in the study area or from a phosphoric acid plant at Whitehaven, used to process phosphate ore up to the early 1990's (Poole *et al.* 1995). However, the distance between this plant and the sample area is more than 10 km and it is unlikely that the particle detected could have travelled that far. Consequently, it seems reasonable to deduce that this particle is of local mineral origin. Relatively high concentrations of lanthanum, mostly as monazite, are present in stream sediments close to Sellafield (British Geological Survey 1992).

4.2 Examination of particle no. 2

A secondary electron image of particle 2 (Figure 4a) shows that this particle is similar in size to particle 1, but with a rougher, less weathered, surface. The image obtained using BSI is shown in Fig 4b. The EPMA spectrum revealed that the particle is composed mainly of zirconium, which indicates that the particle may be a fragment of fuel cladding. However, the lack of a peak for uranium or plutonium in the spectrum means that no firm conclusion can be drawn. EPMA provides information only on the surface of the object being studied, so the alpha-emissions may have originated from a region inaccessible to the electron beam. The absence of a signal for silicon eliminates the possibility that the particle could be a mineral fragment, such as zircon.

4.3 Results of alpha-particle and gamma-ray spectrometry

The sample sections containing Particles 1 and 2 were examined by alpha-particle spectrometry *in vacuo* under a silicon surface barrier detector and by gamma-ray spectrometry on a high-purity germanium detector. Although enhanced alpha-activity was readily detected in both samples, the resulting spectra were too degraded to permit nuclide identification. The gamma-ray analyses also provided little useful information.

5. Discussion and Conclusions

No evidence for alpha-emitting radioactive particles was found in the cultivated vegetables, nor in any other samples that might be considered to be acting as collectors for freshly deposited material, such as the frisbees or the grass trays. This suggests that, over the period of the study, BNFL Sellafield was not a source of fresh alpha-emitting radioactive particles to the terrestrial environment.

The discovery of only two alpha-emitting radioactive particles in nearly 400 sample exposures might imply that the abundance of hot particles in the vicinity of Sellafield is low. However, the area sampled in this scoping study is small in comparison to the total area within 1000 m of the Sellafield perimeter (in the order of 10^6 m^2). The surface samples analysed (pasture grass, grass trays, surface leaf litter, frisbees, and vegetables) constitute an area of approximately 14 m^2 (considering only the 'footprint' of the vegetables). If it is tentatively assumed that the area sampled is representative, in terms of alpha-emitting radioactive particle occurrence, of the area surrounding Sellafield, then a surface density can be estimated of 0.07 natural or anthropogenic alpha-emitting hot particles per square metre. Clearly, this is a crude calculation and no confidence limits can be presented.

Given that at least 20 positive results would be required to determine, with a reasonable degree of confidence, the surface density of such particles, this study suggests it would be necessary to analyse samples collected from an area of more than 280 m^2. The collection, processing and analysis of this quantity of sample material would be prohibitively costly in terms of time and resources.

With some assumptions, an estimate of the activities of the two particles can be made. In experimental work on Chernobyl hot particles, Kushin *et al.* (1993) determined a registration efficiency for LR-115® of $\rho = 0.47$, assuming that the particles being studied were spherical and on the surface of the sample. The electron micrographs show that, in both samples, the particles are very close to the sample surface. Some assumptions about the geometric efficiency (γ) must also be made. Examining the immediate environment of each particle, both were in close proximity to other, larger particles. These will occlude some proportion of the alpha-radiation being emitted from the active particles. The geometric efficiency assumed in this study was 0.35 and the activities of the particles then estimated as follows:

$$A = \frac{x}{\rho.\gamma.t}$$

where A = activity of the particle (Bq),

x = number of tracks counted,

ρ = registration efficiency,

γ = geometric efficiency,

t = exposure time (s).

After a 72-hour exposure period, 55 tracks for Particle 1 and 45 tracks for Particle 2 were revealed, suggesting total activities of 1.3 mBq for Particle 1 and 1.1 mBq for Particle 2. This is a crude calculation, but it does give a first order approximation of the activities of the particles in the absence of useful spectrometric data.

The radiological implications of such particles are discussed in Whittall *et al.* (1999). The highest estimated committed effective dose, based on conservative assumptions for resuspension, inhalation and retention in the respiratory tract is less than 5×10^{-4} μSv a^{-1} for either particle.

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Table 1. Sample inventory

Site No.	OS grid reference	Nature of samples
1	3035 5045	Grass, Soil 0-2 cm, 2-3 cm
2	3041 5043	Soil 0-2 cm, (grass too short to obtain sample)
3	3023 5042	Grass, Soil 0-2 cm, 2-3 cm
4	3038 5037	Grass, Soil 0-2 cm
5	3039 5029	Grass, Soil 0-2 cm
6	3038 5029	Grass, Soil 0-2 cm
7	3022 5054	Grass, Soil 0-2 cm
8	3023 5054	Grass, Soil 0-2 cm
9	3036 5033	Pine needle litter 0-2 cm, 2-5 cm
10	3037 5035	Pine needle litter 0-2 cm, 2-5 cm
11	3037 5037	Pine needle litter 0-2 cm, 2-5 cm
12	3022 5045	Grass, Soil 0-2 cm, grass trays, frisbees, 26 vegetables
13	3032 5051	Grass, Soil 0-2 cm, grass trays, frisbees, vegetable plot destroyed by caterpillars.
14	3038 5035	Grass, Soil 0-2 cm, grass trays, frisbees, 28 vegetables
15	3042 5031	Wheat
16	3041 5025	Barley
17	3035 5038	Blackberries

Table 2. Summary of sample exposures

Sample Type	Number	Sample Type	Number
Soils	39	Vegetable	162
Grass (from pasture)	30	Vegetable washings	54
Grass (monthly trays)	27	Wheat	3
Grass (4-month trays)	9	Barley	3
Pine litter (needles)	18	Blackberries	9
Pine litter (fines)	18	Frisbee washings	12
Total Number of Exposures = 381			

List of Captions

Fig 1a. Autoradiograph of Particle 1 (4 week exposure)

Fig 1b. Autoradiograph of Particle 2 (4 week exposure)

Fig 2a. Autoradiograph of Particle 1 (72 hour exposure)

Fig 2b. Autoradiograph of Particle 2 (72 hour exposure)

Fig 3a. Secondary electron image of Particle 1

Fig 3b. Back-scattered electron image of Particle 1

Fig 4a. Secondary electron image of Particle 2

Fig 4b. Back-scattered electron image of Particle 2