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Post Chernobyl surveys of radiocaesium in soil, vegetation, wildlife and fungi in Great Britain

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Abstract

The dataset "Post Chernobyl surveys of radiocaesium in soil, vegetation, wildlife and fungi in Great Britain" was developed to enable data collected by the Natural Environment Research Council after the Chernobyl accident to be made publicly available. Data for samples collected between May 1986 (immediately after Chernobyl) to spring 1997 are presented. Additional data to radiocaesium concentrations are presented where available. The data have value in trying to assess the contribution of new sources of radiocaesium in the environment, providing baseline data for future planned releases and to aid the development and testing of models.

The data are freely available for non-commercial use under Open Government Licence terms and conditions. doi:10.5285/7a5cfd3e-0247-4228-873d-5be563c4ee3b

1 Background

The accident at the Chernobyl Nuclear Power Plant (Ukraine) on 26 April 1986 remains the worst accident in nuclear history (Smith and Beresford, 2005). The explosion released large quantities of radioactivity into the atmosphere, which spread over much of western Union of Soviet Socialist Republics (USSR) and Europe (De Cort et al., 1998).

The radioactive plume reached the UK on 2 May 1986 (Smith and Clark, 1989), with relatively high levels of deposition being recorded in areas which received heavy rainfall during the passage of clouds over the country (Allen, 1986; Smith and Clark, 1989). Areas receiving high levels of deposition tended to be upland areas in the west. The grazing of free-ranging sheep is the predominant land use in many of these areas which are characterised by nutrient-poor, highly organic acidic soils (Beresford et al., 2007). This resulted in radiocaesium in the tissues of sheep that exceeded 1000 Bq kg^{-1} fresh weight, the limit adopted within the UK after Chernobyl to designate meat that could not enter the food chain. Consequently, in 1986 restrictions were placed on the movement and slaughter of animals from 1670 farms in west Cumbria (north-west England), 5100

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farms in north Wales and 2144 farms in Scotland. Subsequently, 122 farms in Northern Ireland were placed under restriction in 1987. Whilst the number of farms under restriction reduced relatively rapidly, in 1991 more than 700 farms were still restricted, and restrictions were only finally lifted on the remaining 334 farms in Wales and 8 Cumbrian farms in June 2012 (26 years after the Chernobyl accident) (Environment Agency et al., 2013).

In May 1986 a survey of radiocaesium in vegetation was carried out (Horrill and Lindley, 1990) by the Institute of Terrestrial Ecology (ITE) (the predecessor of the Centre for Ecology and Hydrology, CEH). Samples were collected from 318 sampling sites which were selected to represent 16 different land classes (described below). This was used to produce the first map of contamination across Great Britain (Allen, 1986). Subsequently, some of these sites were resampled, with soil also being collected and analysed. Over the following years the institute conducted a number of surveys focussed on specific areas or to address questions of long-term management of the affected areas and particular routes of transfer to human consumers. Although a number of radionuclides were observed in the initial fallout in Great Britain (e.g. Howard and Beresford, 1989) the long-term focus was on caesium-134 (^{134}Cs) and caesium-137 (^{137}Cs). The ^{137}Cs : ^{134}Cs ratio from the Chernobyl deposition was circa 2 : 1 in 1986 and hence the ratio could be used to distinguish aged Cs deposits (pre-Chernobyl) from Chernobyl derived Cs (see Beresford et al., 1992, for discussion).

Whilst the resultant data have been used for a number of purposes (e.g. development and testing of predictive models, Gillett et al., 2001; Wright et al., 2003, estimation of the intake of radiocaesium by consumers of wild fungi, Barnett et al., 1999, understanding of the behaviour of radiocaesium in upland farming systems, Beresford et al., 2007) they have never been published in their entirety and in many instances not even summaries were made available in the open literature. In this paper we present a description of the post-Chernobyl data collected by ITE/CEH which has now been made openly available (Chaplow et al., 2014). The data have value in trying to assess the contribution of new sources of radiocaesium in the environment (e.g. as happened

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after the 2011 Fukushima accident, Beresford et al., 2012), providing baseline data for future planned releases (e.g. new nuclear power stations) and to aid the testing of models (e.g. the development of process based models that require spatial data to enable their testing, Hinton et al., 2013). The purpose of this paper is to describe the available data and methodology used to obtain it. However, in instances where the data have not been reported previously in the open literature there is also a limited discussion of the results.

The research focused on caesium-137 which has a half-life of 30 years and was the most abundant and long lived component of the Chernobyl release. Other isotopes were measured in some samples.

2 Data

The datasets available from Chaplow et al. (2014) are:

1. Survey of Chernobyl fallout in graminaceous vegetation across Great Britain conducted in May and June 1986, and re-surveys carried out in October 1986 and spring 1987 (including samples of soil, different vegetation types and wild animals).

2. Survey of radiocaesium in vegetation sampled in Cumbria in 1986, north Wales in 1987, and North Yorkshire (north England) in 1988.

3. Survey of soil and vegetation over three sites in Cumbria conducted in 1989 and 1990.

4. Results of soil and vegetation sampling over the grazing areas of three upland sheep flocks in Cumbria conducted in 1992.

5. Survey of fungi and associated soil sampled from throughout Great Britain between autumn 1995 and spring 1998.

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The above numbers are used in Table A1 and subsequently to identify the datasets. All the data are available as comma separated variable files (Chaplow et al., 2014) and metadata are available as supporting information from the Environmental Information Data Centre (EIDC) Hub (http://eidchub.ceh.ac.uk/). Locations of sampling sites are presented in Fig. 1.

Overview of studies and available data

Dataset 1

To provide a rapid response for regulatory and scientific purposes to the deposition of Chernobyl derived fallout, ITE conducted a survey of ^{137}Cs concentrations in grassy vegetation throughout mainland Great Britain in May–June 1986. The aim of the survey was to determine the pattern of Chernobyl deposition and the range in concentrations over the country, and to establish a base-line for subsequent assessment of the movement and distribution of radionuclides deposited in the terrestrial environment. Representative samples of vegetation needed to be collected quickly before the concentrations of radionuclides were affected by weathering, plant uptake or grazing by animals.

The wide geographical spread of the ITE research stations enabled samples to be collected simultaneously over most of Great Britain (GB) by trained fieldworkers. A stratified sampling scheme was used based on the ITE Land Classification of Great Britain (Bunce et al., 1981; Benefield and Bunce, 1982). This classification used climatic, topographic, geological and land use characteristics to define 1 km square land units; multivariate analysis techniques were used to develop a classification describing Great Britain in terms of 32 land classes (Bunce et al., 1981; Benefield and Bunce, 1982). To respond swiftly to the Chernobyl accident, the classification was simplified (using Indicator Species Analysis) to result in 16 land class groups (Horrill and Lindley, 1990). Twenty one-kilometre square areas for each class were randomly selected, resulting in 320 sampling sites being chosen as representative of the main variation in

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land classes within Great Britain. Grassy or graminaceous vegetation (including sedges and rushes), was sampled by the field surveyors. Graminaceous vegetation samples were in part chosen due to the fact that these plants form an important component of the food of grazing animals. Sample sites were selected to be away from roadsides (to avoid splash) and overhanging vegetation (to avoid throughfall from trees). 318 sites were sampled in total; 2 sites had to be omitted for practical reasons. These data were used to produce the first spatial visualisation of Chernobyl fallout over mainland Great Britain (Allen, 1986; Fig. 2).

The initial survey was repeated in October 1986 and extended to include upper and lower soil horizons and other vegetation types (heather, *Calluna vulgaris* and bracken, *Pteridium aquilinum*) (Horrill et al., 1988). In addition to vegetation, samples of wildlife (rabbit, *Oryctolagus cuniculus*, hare, *Lepus capensis* and *Lepus timidus*, fox, *Vulpes vulpes*, red deer, *Cervus elaphus*, woodcock, *Scolopex rusticola* and grouse, *Tetrao tetrix* and *Lagopus lagopus*) were collected and analysed to assess the uptake of radioactivity by wild herbivores and carnivores. Samples of food ingested shortly before death were also obtained by collecting rumen, stomach and crop contents from the various species. One hundred and forty-six determinations of animal tissue were made and 94 samples of gastrointestinal tract contents were analysed.

In spring 1987, a survey was carried out to re-analyse the 100 areas in Scotland with the highest concentrations of ^{137}Cs in vegetation as determined in May/June 1986. A further 56 sites in the western Isles of Scotland that had not been included in the earlier surveys were also sampled.

For all surveys, vegetation was sampled by clipping from a 1 m square using garden shears to within 1 centimetre of the soil surface. Plant material was oven dried at 85 °C for 24 h, the total mass per metre square was recorded, ground (Christy and Norris mill using a 0.7 mm sieve) and 40 to 50 g material accurately weighed into 150 mL sample containers.

Soil was sampled by taking a 20 cm × 20 cm × 15 cm layer from the centre of the vegetation sampling plot. If a litter layer was present this was removed and stored

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as a separate sample. Where an obvious soil horizon was present the soil block was divided and the dimensions of each subsample were recorded. Fresh soil pH was measured and soil texture was assessed in terms of clay, silt, sand and organic matter. The soil was broken up by hand, oven dried at 60 °C for 24–36 h, sieved and separated into greater and less than (<) 2 mm fractions. Gamma spectrometry was carried out on the < 2 mm fraction. Samples were retained in a cold store at approximately 2 °C until processed.

Sampling of animals and birds relied heavily on the cooperation and goodwill of landowners, stalkers, gamekeepers and Forestry Commission Rangers. Samples of flesh were removed from the necks of deer, legs and backs of hares, rabbits and foxes and from the breast of the bird species. Gastrointestinal tract contents were retained. Samples were weighed and frozen prior to analysis. Samples were thawed for 24 h prior to gamma analysis.

Radiocaesium levels were determined as activity concentrations per unit dry matter (DM) of vegetation or soil and deposition per unit area, by high resolution gamma spectroscopy using either hyperpure germanium or germanium-lithium detectors with relative efficiencies of 20–25 % for between 25 000 to 80 000 s. Detectors were calibrated using British Calibration Service standards incorporated into appropriate matrices (including different soil densities) and volumes. Accuracy was checked against international standards (Horrill et al., 1988). The unit of activity reported, for this and subsequent datasets, was the bequerel (Bq) which equates to 1 disintegration per second (s). All results were decay corrected to the date of sampling. Animal samples reported on a fresh weight basis with the exception of gastrointestinal tract samples which were dried prior to analysis.

Dataset 2

In May/June 1987 graminaceous vegetation was collected from 26 sites in and around the area of North Wales where restrictions were in place on the movement and slaughter of sheep (Beresford et al., 1987a) and 70 sites in and around the restricted area

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of west Cumbria (Beresford et al., 1987b). Sampling sites were those used in the initial GB survey conducted in 1986 as outlined above. Collection and sample preparation was as described above for the GB wide surveys. Samples were analysed using a lead-shielded 3 inch sodium iodide activated with thallium (NaI (Tl)) detector with a sub-set of samples being reanalysed using germanium (Ge) detectors. Soils from each sampling site were categorised on the basis of textural class (i.e. clay, silt, sand, loam or organic) and soil pH determined using the method of Allen (1989). Previously unreported in the open literature, these surveys demonstrated that those sampling sites at which radiocaesium levels had decreased the least had organic soils.

Smith (1986) suggested that areas of North Yorkshire received high rainfall during the passage of the Chernobyl cloud and it was therefore suggested that the area may have received relatively high levels of deposition. Two hundred and fifty-six sites were sampled in January 1988 (Howard et al., 1988). In addition to graminaceous vegetation, new growth *Calluna vulgaris* (heather) was also sampled if present. Heather was collected from 45 of the sampling sites; from 7 sites no graminaceous species were present and only heather was sampled. Samples were prepared as above and initially analysed using a lead-shielded 3″ NaI (Tl) detector to determine ^{137}Cs activity concentrations. Those grass samples with detectable levels of ^{137}Cs were then re-analysed using Ge-detectors to determine ^{134}Cs and ^{137}Cs activity concentrations; all heather samples were analysed using Ge-detectors. Only 9 grass samples had activity concentrations in excess of 340 Bq kg^{-1} DM. Activity concentrations in heather were higher than those in grass with a maximum of 650 Bq ^{134}Cs kg^{-1} and 3300 Bq ^{137}Cs kg^{-1} being recorded. From the ^{134}Cs : ^{137}Cs ratio it was evident that a significant proportion of the ^{137}Cs in some samples originated from sources other than the Chernobyl accident (i.e. predominantly above grounds nuclear weapons testing fallout). A sigmoidal relationship was derived between reported rainfall and the ^{134}Cs : ^{137}Cs ratio in North Yorkshire heather samples ($r^2 = 0.7$).

$$^{134}\mathrm{Cs} : {}^{137}\mathrm{Cs} = \frac{0.3}{1 + 1.26 \times \mathrm{rainfall}^{-0.71}}$$

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Dataset 3

A helicopter survey of radiocaesium deposition identified areas with unexpected $^{137}Cs:{}^{134}Cs$ ratios in Cumbria in July and August 1988 (Sanderson and Scott, 1989). A ground survey of radiocaesium activity concentrations in soils was conducted in 3 areas (each 3 km × 5 km) in the south western corner of the restricted area of west Cumbria in 1989 and 1990, to compare with the aerial survey results (Beresford et al., 1990). The areas were predominantly unimproved grassland (fell) with some enclosed pastures, woodland or areas of heather (*Calluna vulgaris*). Thirty-six samples of soil (down to a maximum of 40 cm depth) were collected from each area. Soil bulk density was determined for the complete profile by weighing a known volume of soil. In two of the areas additional sampling was conducted to study radiocaesium uptake by vegetation (Beresford et al., 1990, 1992). Vegetation was clipped to the level of plant bases from an area of 1 m^2. Samples of soil were taken to a depth of 40 cm or bedrock if the soil was shallower than 40 cm. The 0–4 cm layer was separated from the rest of the profile. Soil and vegetation samples were weighed fresh, dried at 80 °C and then reweighed to determine moisture content. Bulk density was measured for both soil layers by drying and weighing a known volume of soil. Samples were then ground and weighed into plastic containers (150 mL) or Marinelli beakers (750 mL) for gamma analyses using high resolution Ge-detectors. Count times ranged from 40 000 to 170 000 s depending upon the radioactivity present to give a counting error of $< 5\,\%$ (95 % confidence level; note an evaluation of factors such as weighing of sample, analyst, positioning on detector head, etc. demonstrate that the counting error (which includes errors for the standard and radionuclide decay) dominates the total error). The detectors were calibrated using mixed gamma standards (National Physical Laboratory, Teddington, UK).

In the areas studied, aged radiocaesium, primarily from nuclear weapons fallout and the 1957 Windscale accident, accounted for 30–60 % of the ^{137}Cs deposit (Beresford et al., 1990). The uptake by graminaceous vegetation of aged and

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Chernobyl radiocaesium deposited onto soil (differentiated using the $^{134}Cs:^{137}Cs$ ratio) was estimated and found to be similar (reported in Beresford et al., 1992).

Dataset 4

Beresford et al. (1996, 1998, 2000, 2007) describe a series of studies conducted at three upland sheep farms in west Cumbria to investigate the reasons for variability in the radiocaesium activity concentration in sheep within an individual flock. To support this work, samples of soil and graminaceous vegetation were collected during the summer of 1992 (Beresford et al., 2007). The farm codes (A, B and C) used here are consistent with those used in previous publications (Beresford et al., 1996, 2000) and the accompanying dataset (Chaplow et al., 2014).

Eighty sites selected randomly from a 100 m grid based on Ordnance Survey national grid squares were sampled over the grazing areas of each of Farms A and C (both circa 3 km^2); whereas, over the larger (circa 7 km^2) grazing area of Farm B, 100 randomly selected sites were sampled.

Soil maps (1 : 25 000 scale) were produced by soil surveyors using soil augers and small pits along a transect (Beresford et al., 2007). Data from transects was extrapolated to the rest of the sampling area using aerial photographs (ADAS 1 : 10 000 and 1 : 18 000 scale). Soils were classified according to Avery (1980). Soil samples were collected to a depth of 30 cm or bedrock/stone layer using a spade, trimmed and divided into two layers, 0–4 cm and > 4 cm. Vegetation was collected from a known area to a height of circa 1 cm above the ground surface using hand shears.

Soil and vegetation were dried at 80 °C, weights were recorded and samples were ground. Soil bulk density and loss on ignition (LOI) were determined as described above. The ^{134}Cs and ^{137}Cs activity concentrations in soil and vegetation samples were determined using hyper-pure Ge-detectors with relative efficiencies ranging from 25 to 40 %. Resultant spectra were analysed using the Canberra "Apogee" software package; analysis times (ranging from 5000 to 60 000 s depending upon sample activity)

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were sufficient to achieve a counting error of less than 5 % of the reported result for ^{137}Cs at 1.65 sigma. The detectors were calibrated using mixed gamma standards (National Physical Laboratory).

Dataset 5

This dataset contains information from two studies. In the first study samples of the most frequently eaten (as identified in a survey by Barnett et al., 1999) autumn and spring fruiting fungi species were sampled throughout Great Britain between spring 1995 and spring 1996 to assess radiocaesium contamination levels and geographical variation (Barnett et al., 1999). The dataset contains information from fruiting species where, in most cases, soil samples were also collected from the same location as the fruiting bodies. In the second study, further samples were collected as part of the European Commission (EC) SAVE project (No. F14PCT950015) in autumn 1997 and spring 1998; species targeted were spring fruiting and those that had showed higher levels of ^{137}Cs accumulation in the Barnett et al. (1999) study.

From each site approximately 200 g fresh wt (FW) of fruiting bodies (425 samples representing 37 different species) were sampled. Soil samples were taken from beneath the fruiting body using a metal soil corer with a diameter of 61 mm of either 100 mm or 200 mm in length, depending upon the anticipated total soil depth. The litter layer, if present, was included within the soil sample.

The fruiting bodies were weighed and dried at 60 °C to a constant weight. The dried samples were then ground in a small mill and the ground material transferred to plastic containers of a suitable size (between 10 and 150 mL) prior to gamma analysis. The soil samples were carefully removed from the corers and visually divided into an "organic" layer (including the litter layer, if present) and mineral layer. Each portion of the soil was dried at 60 °C to a constant weight and the bulk density determined. The dried soil samples were then ground in a ball mill before being weighed into suitably sized containers (between 10 and 700 mL) for subsequent gamma analyses.

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Samples were analysed on hyper-pure germanium detectors calibrated using mixed gamma standards (National Physics Laboratory) using the Canberra APOGEE or PRO-COUNT software packages to determine the activity concentration of gamma-emitting radionuclides. Analyses times ranged from 1500 to 240 000 s depending upon the activity present. Results were decay corrected to the date of sampling and where possible, a counting error of less than 5 % of the result for ^{137}Cs was achieved.

3 Access and conditions of use

The data described here have a Digital Object Identifier doi:10.5285/7a5cfd3e-0247-4228-873d-5be563c4ee3b and are freely available from the CEH Environmental Information Data Centre Hub (http://eidchub.ceh.ac.uk/) under the Open Government Licence. These must be referenced fully for every use of the data as: Chaplow, J. S., Beresford, N. A., Barnett, C. L. (2014). Post Chernobyl surveys of radiocaesium in soil, vegetation, wildlife and fungi in Great Britain. NERC-Environmental Information Data Centre doi:10.5285/7a5cfd3e-0247-4228-873d-5be563c4ee3b and where appropriate the source references as cited above.

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Table A1. A description of the data included in this paper.

Dataset ID	Years	What was sampled?	Radionuclides reported	Other parameters reported	Number of samples
1	1986, 1987	Vegetation, (grass, heather, bracken) soil and wildlife (game birds, hare species, fox, rabbit and red deer)	Cs-137, Cs-134, Ru-106, Ru-103, K-40	Soil pH, depth, LOI*, texture and wet and dry weight of vegetation	1585
2	1986, 1987, 1988	Vegetation (grass and heather)	Cs-137, Cs-134	Soil type, pH, ITE Land Class	381
3	1989, 1990	Soil (0–4 cm and > 4 cm) and vegetation	Cs-137, Cs-134, K-40, Pu-238, Pu-239, Pu-240	Soil depth and bulk density	168
4	1992	Soil (0–4 cm and > 4 cm) and vegetation	Cs-137, Cs-134	Soil type, bulk density, depth, pH, LOI*, K and NH_4 and vegetation group (determined by TWINSPAN analysis) and weight	258
5	1995, 1996, 1997, 1998	Fungi (37 species) and soil (organic and mineral layers)	Cs-137, Cs-134	Soil depth and bulk density	423

* LOI – Loss on ignition.

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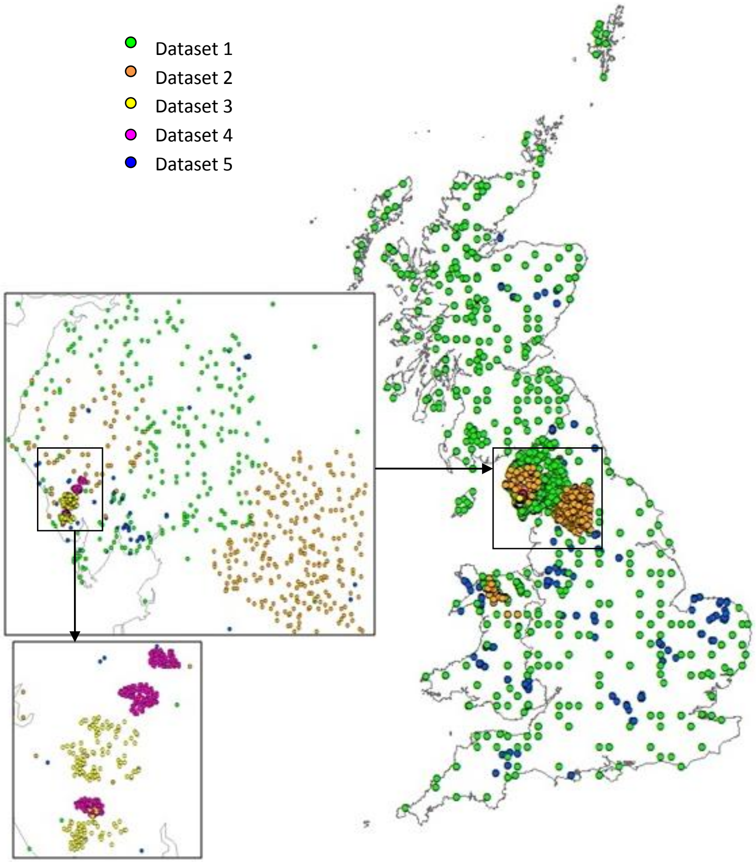

Figure 1. Location of sampling sites. Larger inset shows intensive sampling that was carried out in Cumbria and North Yorkshire; smaller inset shows sampling locations of Datasets 3 and 4 in Cumbria. Copyright© NERC (CEH) 2014. Contains Ordnance Survey data© Crown Copyright and Database Right.

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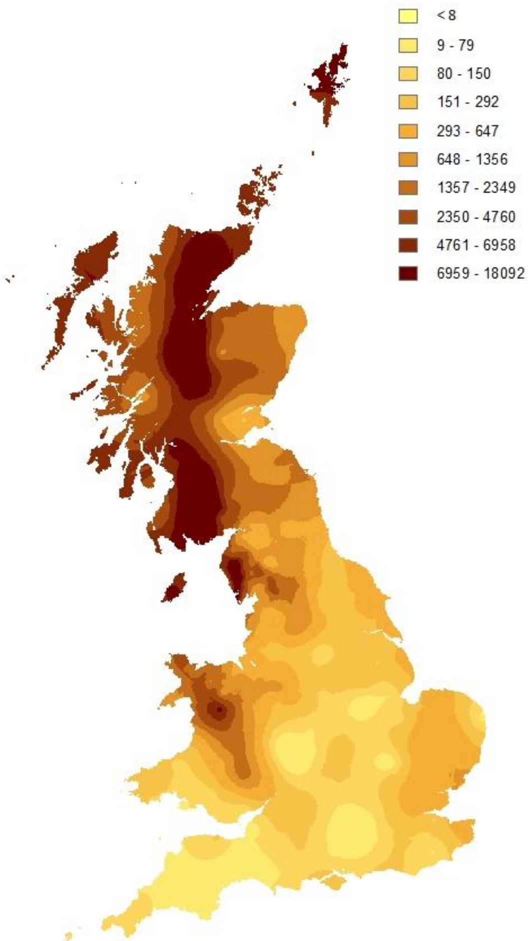

Figure 2. Spatial visualisation of Chernobyl fallout over Great Britain in May 1986 (^{137}Cs activity concentrations (Bq kg^{-1} Dry Matter, DM). Copyright© NERC (CEH) 2014.

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