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Spatial datasets of radionuclide contamination in the Ukrainian Chernobyl Exclusion Zone

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Abstract. The data set "Spatial datasets of radionuclide contamination in the Ukrainian Chernobyl Exclusion Zone" was developed to enable data collected by the Ukrainian Institute of Agricultural Radiology after the Chernobyl accident to be made publicly available. Data for samples collected between May 1986 (immediately after Chernobyl) to 2014 are presented. The data set includes, results from comprehensive soil sampling across the Exclusion Zone (includes ^{137}Cs, ^{90}Sr and soil property data), Pu-isotopes activity concentrations in soils (including distribution in the soil profile), analyses of 'hot' (or fuel) particles from the CEZ (data from across Europe are also included), results of monitoring in the Ivankov region adjacent to the Exclusion Zone. Recently, the CEZ was suggested as a 'Radioecological Observatory' i.e., a radioactively contaminated site that will provide a focus for joint, long-term, radioecological research which will help address challenges identified for the field of radioecology. For this to be successful, relevant data for the CEZ need to be made openly available; indeed the deficiency of open data has been highlighted as one of the causes for the lack of scientific consensus with regard to published studies from within the CEZ and more recently areas affected by the 2011 Fukushima accident. The data presented here are a first step in this process.

The data and supporting documentation are freely available from the Environmental Information Data Centre under the terms and conditions of the Open Government Licence: https://doi.org/10.5285/782ec845-2135-4698-8881-b38823e533bf.

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1 Background

The accident in reactor number four at the Chernobyl nuclear power plant (ChNNP), Ukraine, on the 26th April 1986 remains the worst in the history of nuclear power generation. Starting on the 27th April the human population and farm animals were evacuated from an area of circa 3500 km^2 to create what has become known as the Chernobyl Exclusion Zone (CEZ). Subsequently, the area administered as the CEZ has increased to approximately 4760 km^2. The area of the CEZ in the Ukraine is approximately 2598 km^2, the remainder being in Belarus. The deposition of radionuclides over the CEZ is known to be highly spatially heterogeneous. Releases from the Chernobyl reactor occurred over a period of about ten days. There is a narrow band of high radioactivity to the west of the reactor (often referred to as the western trace) which represents deposition from the initial explosion (Figure 1). Higher levels of contamination to the north and to a lesser extent the south are as a consequence of releases over the days following the accident.

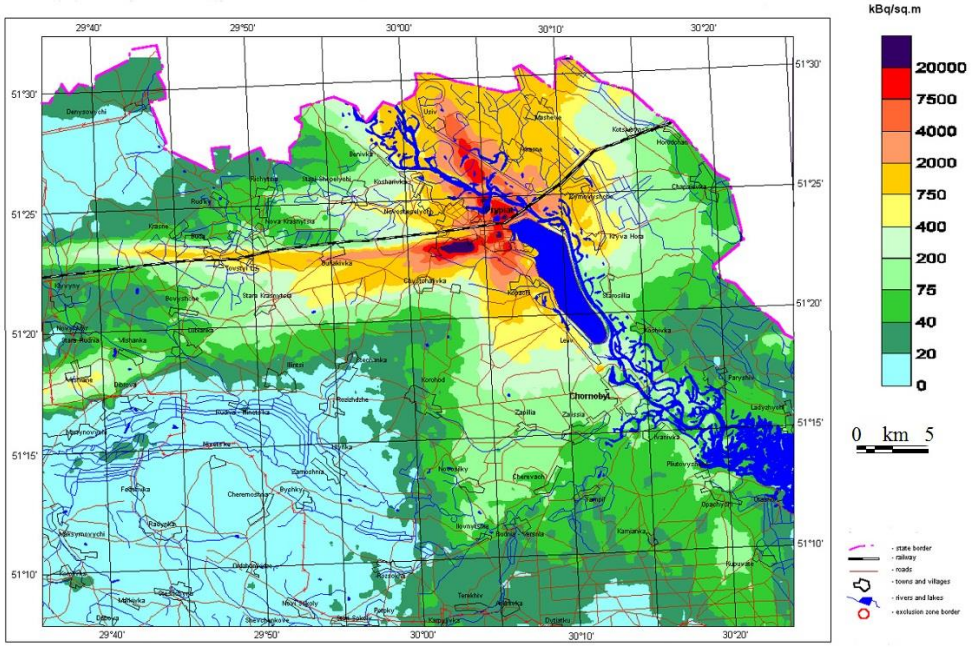

Figure 1. Spatial pattern of ^{90}Sr contamination (kBq m^{-2}) estimated for 1997 (UIAR, 1998)

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The CEZ has many features making it an important radioecological study site:

- Contamination levels are such that the behaviour/transfer/mobility of a number of radionuclides can be studied (^{137}Cs, ^{90}Sr, ^{241}Am, Pu-isotopes, U-isotopes, ^{129}I, ^{14}C, ^{36}Cl and ^{99}Tc).
- The presence of radioactive particles means that their behaviour in the environment can be studied (Kashparov et al., 1999, Beresford et al., 2016). Fuel particles are weathered with time such that mobilisation of ^{90}Sr has been observed to leads to increased contamination of plants (Salbu et al, 1994, Oughton et al., 1993; Kashparov et al., 1999). Fuel particles were released in two different forms (Kashparov et al., 1996): Non-oxidized fuel particles of the initial release (26th April 1986), formed by the mechanical destruction of nuclear fuel. These first particles formed a 100 km long and up to 1 km wide path to the west of the plant, and oxidized fuel particles which were formed during the subsequent reactor fire (from 26th April to 5th May 1986) and deposited to the north and south of the plant.
- Dose rates remain sufficiently high that we may expect to observe effects on wildlife in some areas. Furthermore, published results on radiation effects from the CEZ are contentious with a lack of agreement on interpretation amongst scientists and a high public profile (Beresford and Copplestone, 2011).
- A wide range of terrestrial and aquatic species and habitats are present. The Ukrainian area, the focus of this paper, contains forests, abandoned farmlands, wetlands, flowing and standing waters, deserted villages and urban areas.

Recently, the CEZ was suggested as a 'Radioecological Observatory' (http://www.radioecology-exchange.org/content/radioecological-observatories; Steiner et al. 2013), i.e., a radioactively contaminated site that will provide a focus for joint, long-term, radioecological research which will help address challenges identified for the field of radioecology (Hinton et al. 2013). For this to be successful, relevant data for the CEZ need to be made openly available; indeed the deficiency of open data has been highlighted as one of the causes for the lack of scientific consensus with regard to published studies from within the CEZ and more recently areas affected by the 2011 Fukushima accident (Beresford et al. 2012; Barnett & Welch 2016).

In this paper we describe a series of data sets focussed on radionuclide depositions and radioactive particles within and around the CEZ predominantly from studies conducted by the Ukrainian Institute of Agricultural Radiology (UIAR) (now part of the National University of Life and

Environmental Sciences of Ukraine (NUBIP)). The accompanying data are freely available (https://doi.org/10.5285/782ec845-2135-4698-8881-b38823e533bf) under the terms of the open Government Licence. With respect to radioactive particles, data are also presented from samples collected in Poland in 1986.

The purpose of this paper is to describe the available data and methodology used to obtain them. The data will be valuable to those conducting studies within the CEZ in a number of ways, for instance: (i) helping perform robust exposure estimates to wildlife (Beaugelin-Seiller submitted); (ii) predicting comparative activity concentrations of different key radionuclides; (iii) providing a baseline against which future survey in the CEZ can be compared; (iv) as a source of information on the behaviour of fuel particles; (v) performing retrospective dose assessments; (vi) assessing natural background dose rates in the CEZ.

Whilst many of the results of the studies described have previously been used for various purposes within refereed or other publications (e.g. Kashparov et al. 2001, 2003, 2004, 2006; Zhurba et al. 2009) the complete underlying data sets have not previously been published.

2 Data

There are six data sets available in Kashparov et al., 2017 and an overview of the contents of each is presented below and in Table 1:

1. Spatial data set of radiocaesium, ^{90}Sr and soil chemistry parameters resulting from a soil sampling exercise conducted during the summer of 1997. The data set comprises results from 1200 soil samples reporting ^{134}Cs, ^{137}Cs, ^{90}Sr and some ^{154}Eu activities in soil (kBq m^{-2}), some chemical parameters for soils (e.g. pH, exchangeable Cs, percentage humus), soil type at the sampling site and latitude-longitude co-ordinates. The data set also contains information for additional samples from some of the data sets below (e.g., this data set contains radionuclide activity in soil for all sampling though for some of these there are no corresponding soil chemistry data).
2. Pu isotope measurements in bulk soil samples reporting results for the analyses of ^{238}Pu and ^{239,240}Pu on a subset of 82 of the samples from the above data set and an additional twelve samples collected from within the CEZ and outside it to a distance of 200 km between 2000 and 2001.
3. Pu isotope measurements in sectioned soil samples reporting results from cores sectioned at different depths collected at nine of the 12 sites sampled between 2000 and 2001.

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4. 'Hot' or fuel particle data set presenting radionuclide activity and some physical characteristics of: (i) 1380 'hot particles' (predominantly fuel particles) extracted from soils collected in the CEZ (largely from in the inner 10 km zone) over the period 1987 to 2000; (ii) 206 particles collected from north-east and Warsaw area (Poland) in September 1986 (data courtesy of Warsaw University (Dabrowska et al., 1988, Osuch et al., 1989); (iii) 294 particles collected from within the 'Shelter' (or sarcophagus) in 1992; (iv) data from published literature on particles collected in different European countries between 1986 and 1987.
5. Fuel particle dissolution data set presents results from studies on 115 soil samples, collected within the CEZ between 1995 and 1997, to determine the proportion of undissolved fuel particles.
6. Ivankov region data presents unreported data of a survey of the Ivankov region, immediately to the south of the CEZ conducted in 2014. The data set contains dose rates measured at approximately 3400 sites across the Ivankov region and activity concentrations of ^{90}Sr, ^{137}Cs and natural series gamma-emitting radionuclides in 547 soil samples.

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144 Table 1. A description of the data presented in this paper and available from Kashparov et al., 2017.

Data set ID	Date sampled	Sample description	Radionuclides reported	Other parameters reported
1	1995, 1999, 2000, 2002	Soil	^{134}Cs, ^{137}Cs, ^{90}Sr, ^{154}Eu ^{238}Pu, ^{239,240}Pu	Soil chemistry and soil type
2	1997, 2000, 2002	Soil	^{238}Pu, ^{239,240}Pu	None
3	2000, 2002	Soil	^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239,240}Pu	Depth and mass of soil layers
4	Gamma: 1986, 1987, 1988, 1989, 1991, 1992, 1995, 1997, 1998, 1999, 2000 Alpha: 1986, 1988, 1990, 1991 Beta: 1988, 1994, 1995	'Hot' (or fuel) particles in soil	^{95}Zr, ^{95}Nb, ^{106}Ru, ^{125}Sb, ^{134}Cs, ^{137}Cs, ^{144}Ce, ^{154}Eu, ^{155}Eu, ^{54}Mn, ^{60}Co, ^{241}Am Total alpha ^{90}Sr	Particle description (type, size, view, colour, structure), burn-up value
5	1995, 1996, 1997	Soil and 'hot' (or fuel) particles	^{85}Sr, ^{90}Sr	None
6	2014	Soil and background dose rate	^{137}Cs, ^{90}Sr, ^{40}K, ^{226}Ra, ^{232}Th	Sample mass

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147 Locations of sampling for studies 1, 2, 3, and 5 are presented in Figure 2 and those for the Ivankov
148 region are shown in Figure 3 (below). Reported activity concentrations for all data sets discussed are
149 presented for the date of measurement which is provided in the accompanying data sets.

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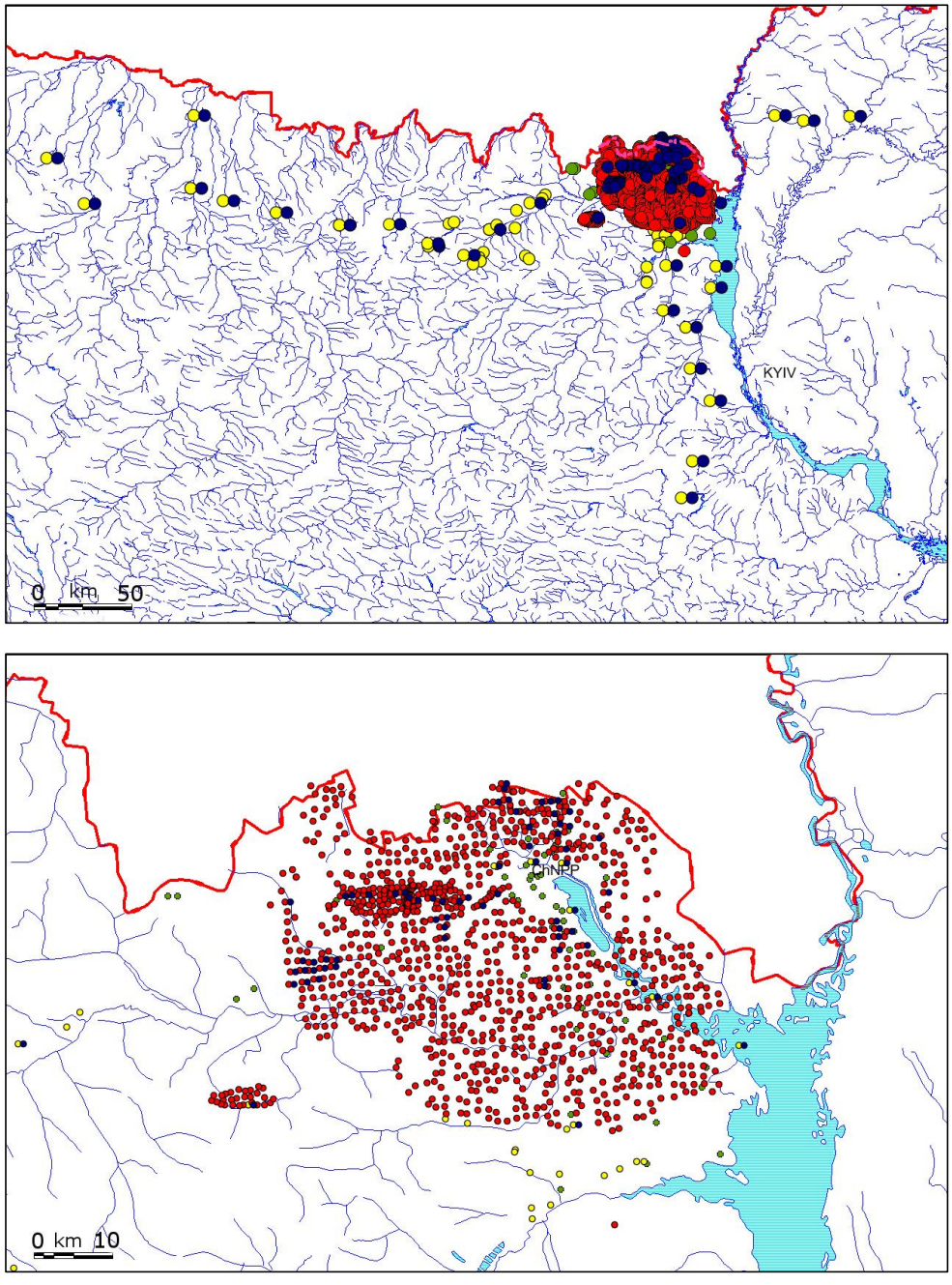

Figure 2. Sampling locations for the different studies in the CEZ (red – 1.Spatial data set of radiocaesium, ^{90}Sr and soil chemistry parameters; - dark blue – 2.Pu isotope measurements in bulk soil samples; yellow - 3.Pu isotope measurements in sectioned soil samples; green - 5. Fuel particle dissolution data set

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157 **1. Spatial data set of radiocaesium, ^{90}Sr and soil chemistry parameters**

158 *Soil sampling*

159 Soil samples were collected at about 1200 sites in the Ukranian CEZ in an area of 36 km radius
160 around ChNPP between July 1995 and September 1997. Sites were selected using a grid and the
161 approximate distance between sampling sites was about 1.2 km. For the narrow western (fuel
162 particle) trace of the radioactive release, where contamination is characterised by a high gradient,
163 the distance between the sampling sites was reduced to 100 to 500 m. Because of the large area of
164 the CEZ, the majority of the sampling was conducted using a helicopter to rapidly move between
165 sites; to sample forest areas motor vehicles were used for transport.
166 At each sampling point, the absorbed dose rate at a height of 1 m above ground surface was
167 measured. Before soil sampling the homogeneity of the site (100 x 100m) was evaluated by
168 measurement of absorbed dose rate at several points using a DRG-01T dosimeter.
169 A composite soil sample was collected that consisted of five sub-samples of cores of 37 mm
170 diameter, taken in the corners and in the centre of 2 to 5 m^2 to 30 cm depth (this approach is
171 referred to as the 'envelope method' and is conducted in accordance to what is now to SOU 74.14-
172 37-425:2006). This approach was developed to obtain a representative soil sample (Khomutinin et
173 al., 2001). The total mass was not less than 2 kg for the five cores. The sampling depth of 30 cm was
174 chosen because it had been shown that ten years after the accident about 95% of the ^{90}Sr activity is
175 associated with the upper 10 to 20 cm layer (Ivanov et al., 1997). To test that the sampling depth
176 was adequate the vertical distribution of ^{90}Sr in sandy soil profiles (up to 1 m depth) was determined
177 and the ^{90}Sr:^{154}Eu activity ratio in the samples was measured. Fuel particles have a characteristic
178 ^{90}Sr:^{154}Eu; if this ratio deviates from that expected it is indicative of ^{90}Sr being leached down the
179 profile. In most of the cores more that 95% of the ^{90}Sr was located in the upper 30 cm layer. Only in
180 a few sites (less than 0.1% of all sites) with a low organic matter content was there a significant
181 percentage of ^{90}Sr (>20%) that had migrated deeper than 30 cm (Shestopalov et al., 2003).
182 The co-ordinates of the sampling sites were determined by means of a GPS-receiver (ScoutMaster)
183 using the WGS-84 (World) system. In order to estimate the accuracy of the co-ordinates and their
184 correspondence to the topographic basis, GPS measurements were performed at several points with
185 known co-ordinates. It was shown that the accuracy of the GPS-receiver was about 100 m with a
186 systematic deviation of about 300 m.
187 The same methodology was used for estimating radionuclide activity concentrations in the other
188 data described below unless otherwise indicated.
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Soil activity measurements

The collected soil samples were dried, sieved through a 1 mm sieve and homogenized. Four sub-samples were taken from each sample for the determination of the total contents of radionuclides: three sub-samples of 100 cm^3 volume and one of 1000 cm^3 volume for the measurement of gamma emitting radionuclides in a Marinelli container. Measurements of gamma emitting radionuclide contents were performed on all four sub-samples (C_i, Bq kg^{-1}) using gamma-spectrometry. If the relative difference of the sub-sample measurements $(C_{max}-C_{min})/(C_{max}+C_{min})$ exceeded 0.15 for the ^{137}Cs activity concentrations, the sub-samples were bulked and remixed. The resultant sample was again divided into replicates which were re-analysed. Once $(C_{max}-C_{min})/(C_{max}+C_{min})$ was <0.15, a 100 cm^3 sub-samples with an approximate average ^{137}Cs activity concentration was chosen for the measurement of the ^{90}Sr specific activity. This procedure was followed to ensure that the sub-sample analysed for ^{90}Sr was representative of the sample as a whole.

Gamma spectrometry measurements were performed using HPGe-detectors, of 30 % relative efficiency and 1.90 keV FWHM for 1333 keV (GEM-30185, EG&G ORTEC, USA) and a multichannel analyser (ADCAM-300, ORTEC, USA) using GammaVision32 software. The efficiency calibration was carried out for the 1 L Marinelli geometry using a spiked soil sample containing ^{241}Am, ^{243}Am, ^{152}Eu, ^{154}Eu, ^{137}Cs, ^{40}K. The average counting time was about 1 hour; for samples of lower activity the counting time was extended to obtain an acceptable error on the measurement (<30 % at 95th percentile confidence interval).

The activity of ^{90}Sr was determined using a radiochemical method (Pavlotskaya, 1997) with treatment of the samples by boiling in 6M nitric acid for four hours. This method was used to dissolve as much of the fuel particles as possible (Kashparov et al, 2003); it has been estimated that for particles collected from the western trace within 5 to 10 km of the ChNPP this method may underestimate ^{90}Sr activity by up to 20 % (Kashparov et al., 2004). After filtration of the solution, hydroxides of high-valence metals (Fe, Al, Ti, Mn, Th, U) were extracted by adding ammonia into the solution. After acidification, the solution was left for three weeks to reach equilibrium between ^{90}Sr and ^{90}Y. Subsequently, stable Y was added to the solution and Y precipitated by addition of ammonia. The precipitate was incinerated to yield Y_2O_3 and after one day (to allow the decay or radium daughters) ^{90}Sr was measured using a low-background beta-counter (CANBERRA-2400, USA). The radiochemical procedure removed ^{137}Cs from the sample which increased the accuracy of the subsequent ^{154}Eu gamma-spectrometric measurements to determine the ^{90}Sr:^{154}Eu ratio (gamma counting to determine the ^{154}Eu activity was conducted after extraction of high-valence metals).

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Determination of the main agrochemical characteristics

Standard methods were used to determine humus content (which equates to organic matter content) (GOST 26213-91), pH_{H2O} (GOST 26423-85), pH_{Kcl} (GOST 26483-85), hydrolytic acidity (determined using the Kappen method (GOST 26212-91)), exchangeable Ca (GOST 26487-85), exchangeable K and P contents were determined using a method appropriate to the specific soil type (GOST 26207-91, GOST26204-91).

Mechanical composition (i.e. percent clay, percent silt, sand) were determined on the soils but not reported in the data. Instead, they have been used to attribute soils to the USSR classification system. Table 2 presents soil types in the CEZ described using USSR countries classification and in accordance with the FAO/UNESCO system (Stolbovoi, 2000). The soil type at each sampling site was attributed to one of these codes and this is presented within the data.

Codes 116 to 143 in the data were sampled from around the village of Bober which was not within the CEZ but at which high deposition values were found. No agrochemical measurements were made at these sites except pH.

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253 Table 2. Soil classifications appropriate to the CEZ.

Code used in data	Ex-USSR (including Ukraine)		FAO-UNESCO
1	Soddy underdeveloped	Soddy-podzolic	Podzoluvisol
2	Soddy slightly- and mid-podzolic sandy and consolidated sandy		
3	Soddy slightly-podzolic sandy-loam and loamy		
4	Soddy mid-podzolic sandy-loam and loamy		
6	Soddy slightly-podzolic gleyic sandy and consolidated sandy		
7	Soddy slightly-podzolic gleyic sandy loam and loamy		
8	Soddy mid- and heavy-podzolic gleyic sandy loamy		
9	Soddy slightly-podzolic gleyic sandy and consolidated sandy		
10	Soddy mid- and heavy-podzolic gleyic sandy loam and loamy		
18	Grey forest	Grey forest	Greyzems
19	Dark-grey forest		
121	Meadow gleyic	Gleyic	Gleysols
122	Meadow and soddy gleyic carbonate	Meadow	Phaeozems
124	Meadow pseudopodzolic and meadow pseudopodzolic gleyic		
131	Meadow-boggy		
133	Boggy	Peat, boggy	Histosols
135	Peaty-boggy		
136	Peat-boggy		
138	Low moor peat		
159	Soddy gleyic sandy and consolidated sandy	Soddy	Arenozols
160	Slightly-soddy low-humus and non-humus sand	Alluvial	Fluvisols
161	Soddy sandy loam and loamy	Soddy	Arenozols
162	Soddy gleyic sandy loam and loamy		
167	Soddy solod		
168	Soddy pseudopodzolic gleyic		

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2. Pu isotope measurements

From the data set described above 82 samples from sites along the main traces of the release were selected for analysis of Pu-isotopes. For determination of plutonium radioisotopes at a given site, one of the homogenized 100 cm^3 samples was used. After heating at 450 °C overnight, the samples were boiled for 4 hours in 6M HNO_3. Plutonium radioisotopes were extracted by a standard radiochemical method (Pavlotskaya, 1997). Chemical yield was calculated using ^{236}Pu or ^{242}Pu as tracers. Plutonium was extracted from the resultant solution using ion-exchange resin (VP-1AP, Russia). After elution and evaporation plutonium was extracted by electrical deposition onto stainless steel plates. Activities on the plates were measured using a Soloist alpha-spectrometer equipped with a Soloist-U0300 detector (EG&G ORTEC, USA).

An additional 12 samples were collected in autumn 2000 and analysed to determine Pu-isotopes using the same methodology. These samples were collected at distances of up to 200 km outside of the CEZ in westerly, southerly and easterly directions.

3. Pu isotope measurements at different depths

Apart from having total Pu isotopes determined as above for the bulked soil sample, cores from eight of these sampling sites were sectioned into layers (0 to 2, 2 to 4, 4 to 6, 6 to 8, 8 to 10, 10 to 15, 15 to 20, 20 to 25 and 25 to 30 cm) and analysed for Pu-isotopes, ^{137}Cs and ^{90}Sr.

A further site was sampled in 2001 to a depth of 110 cm and the core was sectioned into 10 cm slices. Each of these depth samples was analysed to determine ^{238}Pu, $^{239,240}Pu$, ^{137}Cs and ^{90}Sr as described above.

4. 'Hot' or fuel particles

The initial explosion in the Chernobyl reactor released large numbers of uranium dioxide fuel particles with a median radius of 2-3 µm which were deposited in a narrow band up to 100 km to the west of the ChNPP (Kashprov et al. 1999). The explosion also released relatively large fuel fragments ranging from 10's to 100's µm in size. These larger fragments were largely deposited within 2-5 km of the reactor (Kashparov et al. 1999). The deposition of particles was a distinguishing feature of radioactive contamination of the Chernobyl accident and the behaviour of radionuclides associated with such particles in the environment had not previously been considered (Loshchilov et al., 1991, Beresford et al., 2016).

In 1987, the State Committee of Hydrometeorology of the USSR and the Scientific Centre of the Defence Ministry of the USSR created a regular sampling network in the CEZ (Loshchilov et al., 1991).

Fifteen points were chosen on each of 36 transects defined at 10° intervals within 60km of the ChNPP. Soil samples were collected during 1987 and 1989 at each point, except for those located in woods, rivers and lakes; the majority of sampling sites were located within the inner 10 km of the CEZ. The samples (15 cm in diameter and 5 cm deep) were measured in Marinelli beakers using a high-purity Ge detector. From a subset of these samples, all collected within the 30 km zone, more than 1,200 relatively large (size >10 μm) hot particles (activity >100 Bq) were identified and isolated by scanning thin soil layers with a dosimeter (Kuriny et al., 1993). In some instances more than one particle was extracted from the same soil sample (in the data the identifier code starts with the same number).

About 500 additional 'hot' or fuel particles were collected in a similar manner from soils sampled using the same approach in the CEZ during the period 1989–1996 (Kashparov, 2003).

In the inner zone of the CEZ, where these samples were collected, up to 97% of particles comprised finely-dispersed nuclear fuel particles. The other 3% were what are known as 'ruthenium' or condensed particles, a matrix of iron group elements with a high content of ^{103,106}Ru (Kashparov et al., 1996). Condensed particles were formed when highly volatile radionuclides released from the fuel matrix were condensed onto particles of dust, construction materials etc. (Kashparov et al., 1996). Within the database the type of each particle is identified as 'fuel' or 'condensed'.

The specific activity of the fuel particles corresponded to the activity of the fuel at the moment of the accident, excluding the volatile and highly mobile radionuclides such as ^{134,137}Cs and ^{103,106}Ru (Kuriny et al., 1993, Kashparov, 2003). The presence of fissionable material in the particles was demonstrated using neutron-activation analysis, electron probe microanalysis, and laser mass-spectrometry. Particle enrichment with ^{235}U was found to be about 1 to 2% (Kashparov et al, 1996); natural uranium consists of approximately 0.7% ^{235}U (USNRC, 2017). Particle size and radionuclide activity were measured by optical microscopy and by α-, β-, γ-spectrometry respectively. Gamma spectrometry was conducted using high purity germanium detector (GEM-30185, EG&G ORTEC, USA) of about 30% efficiency and 1.85 keV energy resolution (for ^{60}Co, 1332 keV γ-rays) connected to a multichannel analyser (ADCAM™ MCA 350, EG&G ORTEC, USA). Alpha spectrometry was conducted using a gold-silicon detector (DKP-125-IA) linked to a SES-13 spectrometer. Following radiochemical separation using the methodology described above ^{90}Sr activity was determined using an α-β detector (CANBERRA-2400, USA).

An additional 294 particles were obtained from inside of the "Shelter" (or sarcophagus) in the reactor hall of the Chernobyl No4 unit early in 1992 and analysed as described above.

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318 Using the ^{137}Cs and ^{90}Sr results for the particles, Kashparov (2003) estimated the effective duration
319 and temperature for fuel particle annealing to be ~3.5 seconds at 2400 K respectively for particles of
320 >10 µm from Western Trace. This confirmed an explosion-like mechanism of fuel particle formation
321 during the accident (Kashparov, 2003).

322 For particles collected in the CEZ the data set presents 'burn-up' values. Burn-up is used to describe
323 the fraction of fuel atoms having undergone fission. The burn-up distribution of fuel particles larger
324 than 10 µm collected from the CEZ indicated that, at the moment of the accident, particles were
325 released from the less irradiated ('younger') part of the reactor core. Burn-up values were lower in
326 these particles (9.8 to 11 MW day kg^{-1}) in comparison with the estimated most probable fuel burn-up
327 value of 14 MW day kg^{-1} in the 4^{th} unit of the ChNPP (Kuriny et al., 1993, Kashparov et al., 1996,
328 Begichev et al., 1990; Kashparov et al., 1997). Table 3 presents estimated radionuclide activity
329 concentrations at the time of the accident in fuel with different burn-up values. The lower burn-up of
330 fuel contributing to particles within the CEZ resulted in particles with less radioactivity than if
331 deposited particles had been largely formed from 'older' fuel in the reactor.

332 The data set also contains information on 206 particles collected from five areas in the north east of
333 Poland. Particles were collected in autumn 1986 from wasteland and forest clearings; the total area
334 sampled was approximately 6000 square metres (the data are supplied courtesy of Warsaw University;
335 see Dabrowska et al., 1988 and Osuch et al., 1989 for methodology). In contrast to the CEZ, >40% of
336 the particles collected in Poland were of the condensed form; the results for the Polish particles were
337 decay corrected to the day of the accident. The Polish particles were divided into three forms A, B and
338 C. Group A contained mostly ^{103}Ru and ^{106}Ru isotopes, group B particles were rich in other nuclides
339 from the fission product spectrum and group C resembled group B particles in isotope contents but
340 with a higher abundance of ruthenium.

341 For completeness and comparison, we have augmented the data with values from the published
342 literature which present information on particles from: Bialystok in the Masurian Lakes region of
343 northeast Poland (nine particles collected in November 1987) (Schubert and Behrend, 1987);
344 Budapest, Hungary (15 particles collected in July 1986) (Balashazy et al., 1988); Stockholm, Gotland
345 and Gavle in Sweden (41 particles collected in 1987) (Kerekes et al., 1991); Sofia, Bulgaria (five
346 particles) (Mandjoukov et al., 1992). Most of the particles from these studies were also 'ruthenium'
347 condensation particles. Particles were selected in 1986 and activities were decay corrected to the time
348 of the accident.

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5. Fuel particle dissolution data set

Soil Sampling and preparation

Between July 1995 and May 1997, 115 soil samples were collected from sites within the CEZ at varying directions and distances from the ChNPP in order to reflect the variability of the physical and chemical characteristics of the fuel particles deposited (Kashparov et al, 1996, 1999, 2004). The sampling was carried out along the narrow western trace of fallout of the non-oxidized fuel particles of the first release (Chernobyl releases occurred over a period of 10 days (Smith & Beresford, 2005)) and at the southern and northern fuel traces of fallout of oxidized fuel particles (FP). At all sample sites, soddy podzolic sandy soils (type 2 from Table 2) were sampled. Where possible at each site, additional samples were collected from an area of peat soil within a radius of 300 m. It was assumed that within this area the deposited particles would be relatively similar, allowing the effect of soil type on particle behaviour to be studied. Density of contamination by ^{90}Sr varied at the sampling sites from 12 kBq m^{-2} to 60 MBq m^{-2}.

Two soil cores were collected at each sampling point: one sample was collected to a depth of 5 cm and 14 cm diameter; the second was taken to a depth of 30 cm with a 6 cm diameter. The 0 to 5 cm samples (containing the fuel particles) were used to determine soil pH_{H_2O} using the same method as above.

Strontium-85 and ^{90}Sr activity concentrations were determined on the 0 to 30 cm samples which were also used to estimate the fraction of remaining undissolved fuel particles. Soil samples were air-dried (approximately 25 °C), passed through a 1 mm mesh sieve and homogenised prior to analysis.

Estimation of remaining fraction of fuel particles

The fuel particle component in the soil samples was estimated from the fraction of exchangeable ^{90}Sr determined using a 2M NH_4Ac extraction with ^{85}Sr yield monitor (Kashparov et al., 1999). Three or four sub-samples of 100 cm^3 were taken from each 0 to 30 cm soil sample, and 5 ml of ^{85}Sr solution (activity of about 100 Bq) and 25 ml of water were added to each. After 6 to 48 days, the soil was subjected to extraction using 2 M NH_4Ac (solid: liquid ratio 1:10). The soil-extract solution was shaken for 1 hour and left for 1 day. Extractants and soil residues were separated by filtration, and the fraction of radiostrontium was determined in both the solution (A_{sol}) and the residue (A_{res}). The gamma emitting radionuclides ^{137}Cs, ^{154}Eu, ^{241}Am and ^{85}Sr were measured using gamma spectrometry and ^{90}Sr by beta counting after radiochemical separation using the approach outlined above.

The percentages of ^{85}Sr and ^{90}Sr leached from soil ($\Delta^{90}Sr$ and $\Delta^{85}Sr$ respectively) were calculated as follows:

382 $\Delta^{85}Sr = \{A_{sol}(^{85}Sr)/[A_{sol}(^{85}Sr)+A_{res}(^{85}Sr)]\}*100\%$

383 $\Delta^{90}Sr = \{A_{sol}(^{90}Sr)/[A_{sol}(^{90}Sr)+A_{res}(^{90}Sr)]\}*100\%$

384 These percentages were used to calculate the fraction of undissolved fuel particles (ΔFP),
385 estimated as:

386 $\Delta FP = [1 - \Delta^{90}Sr/\Delta^{85}Sr]$.

387 For samples with a low ^{90}Sr:^{154}Eu ratio, it had previously been shown that leached ^{90}Sr had been
388 removed from the 0 to 30 cm layer (Kashparov et al. 1999). This migration was particularly significant
389 for sandy soils with very low humus content. Here, for such samples, the total estimated ^{90}Sr content
390 was used to assess the fraction of undissolved fuel particles rather than $A_{res}(^{90}Sr)$. The total ^{90}Sr content
391 was derived from the vertical distribution of ^{90}Sr and ^{154}Eu activity. From estimates of initial activity
392 concentrations in the fuel the ^{90}Sr:^{154}Eu ratio in 1996 of 56 would be expected (see discussion of
393 predicted activity levels in fuel presented below).

394 The data contains a limited number of negative values which are the consequence of a low percentage
395 of ^{90}Sr in the exchangeable form and a high associated error. Users of the data should assume these
396 values to be zero.

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408 Table 3. The activity concentrations of radionuclides in the ChNPP 4th unit nuclear fuel with different
409 burn-up, Bq g^{-1}.

Burn-up (MW d kg^{-1})	^{90}Sr	^{95}Zr	^{106}Ru	^{134}Cs	^{137}Cs	^{144}Ce	^{125}Sb	^{154}Eu	^{155}Eu
15.20	1.54E+09	3.43E+10	9.66E+09	1.33E+09	1.91E+09	2.73E+10	2.64E+08	5.52E+07	6.48E+07
14.30	1.46E+09	3.46E+10	8.85E+09	1.16E+09	1.80E+09	2.69E+10	2.38E+08	4.91E+07	6.01E+07
13.40	1.38E+09	3.48E+10	8.09E+09	1.01E+09	1.69E+09	2.65E+10	2.13E+08	4.33E+07	5.54E+07
12.50	1.30E+09	3.50E+10	7.36E+09	8.74E+08	1.58E+09	2.59E+10	1.89E+08	3.81E+07	5.12E+07
11.60	1.22E+09	3.51E+10	6.67E+09	7.49E+08	1.47E+09	2.53E+10	1.68E+08	3.32E+07	4.69E+07
10.70	1.14E+09	3.53E+10	6.01E+09	6.35E+08	1.36E+09	2.46E+10	1.47E+08	2.87E+07	4.27E+07
9.84	1.05E+09	3.54E+10	5.38E+09	5.33E+08	1.24E+09	2.37E+10	1.29E+08	2.46E+07	3.86E+07
8.94	9.61E+08	3.55E+10	4.78E+09	4.41E+08	1.13E+09	2.27E+10	1.11E+08	2.09E+07	3.47E+07
8.05	8.72E+08	3.56E+10	4.21E+09	3.60E+08	1.02E+09	2.16E+10	9.48E+07	1.76E+07	3.09E+07
7.15	7.80E+08	3.55E+10	3.66E+09	2.87E+08	9.09E+08	2.03E+10	7.98E+07	1.45E+07	2.72E+07
6.26	6.88E+08	3.53E+10	3.14E+09	2.24E+08	7.96E+08	1.88E+10	6.60E+07	1.17E+07	2.36E+07
5.36	5.93E+08	3.48E+10	2.63E+09	1.69E+08	6.83E+08	1.70E+10	5.34E+07	9.22E+06	2.01E+07
4.47	4.98E+08	3.39E+10	2.15E+09	1.22E+08	5.70E+08	1.50E+10	4.18E+07	7.08E+06	1.66E+07
3.58	4.01E+08	3.23E+10	1.68E+09	8.32E+07	4.57E+08	1.27E+10	3.14E+07	5.15E+06	1.32E+07
2.68	3.03E+08	2.93E+10	1.24E+09	5.17E+07	3.43E+08	1.01E+10	2.20E+07	3.49E+06	9.86E+06
1.79	2.03E+08	2.42E+10	8.07E+08	2.74E+07	2.29E+08	7.19E+09	1.36E+07	2.09E+06	6.53E+06
0.89	1.02E+08	1.53E+10	3.95E+08	1.03E+07	1.15E+08	3.82E+09	6.30E+06	9.27E+05	3.24E+06

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411 ## 6. Survey of Ivankov region

412 Three thousand three hundred and eighty nine sampling points were identified for survey in 2014,
413 using a 1 km grid placed over the Ivankov region and surrounding 3 km (Figure 3) (Kashparov et al.,
414 2014). The co-ordinates of the sampling sites were determined by means of a GPS-receiver (Garmin,
415 GPSmap 78S) using the WGS-84 (World) system with the accuracy of 10 m or less.

416 Measurements of ambient equivalent dose rate (hereafter referred to as dose rate) were carried out
417 at heights of 1 m and 0.1 m above ground surface at 1 km intervals using this grid. Equivalent dose
418 was measured using certified gamma-beta-irradiation dosimeters (RKS-01, Stora-TU, Ukraine). The
419 measurement range of the dosimeters was approximately 0.1 to 1000 μSv hr^{-1} and the time taken to
420 obtain a statistically reliable estimate was approximately 20 s. Seven of the sites selected for survey
421 were inaccessible however the locations have been left in the data for completeness.

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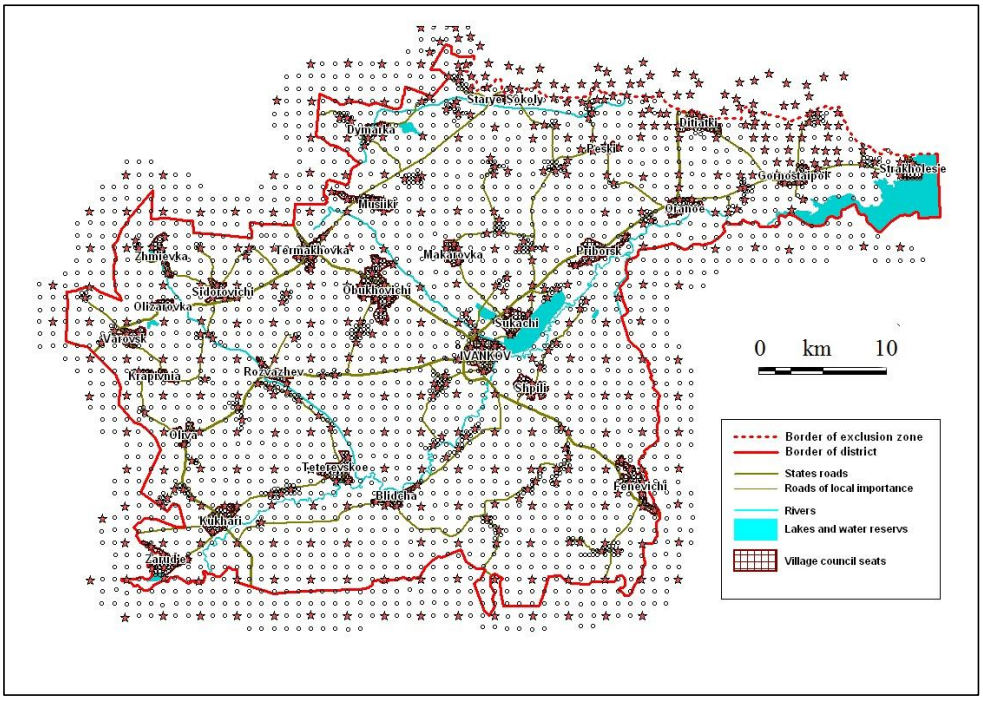

Figure 3. Map of the soil sampling points in Ivankov region: soil sampling and gamma survey (red stars); gamma survey only (grey circles).

An estimation of 'zero background' for the dosimeters was made by taking measurements over a frozen water body in the centre of the Ivankov region. The contribution of gamma irradiation from natural and anthropogenic radionuclides was seen to be negligible and hence the readings were the sum of cosmic radiation and the detectors own background (Kashparov et al, 2014). The background dose rate of the devices themselves were determined in the laboratory by taking measurements within an area shielded by 10 cm lead. Background dose rate of the detectors was estimated to be 0.05 μSv h^{-1} and the 'zero background' determined over the frozen lake was 0.08 μSv h^{-1}. Therefore, the contribution of cosmic radiation in the study area can be estimated to be approximately 0.03 μSv h^{-1}. Contributions from cosmic and instrument background were subtracted from measured values reported in the data set.

Five hundred and forty seven soil samples were collected. Sampling locations were every third dose rate measurement point apart from within settlements where soils were sampled at every measurement point. Sample sites were selected that were undisturbed, relatively flat, with consistent

vegetation cover, and where dose rate estimates did not vary by more than 30%. Samples were taken at a distance from the nearest buildings or trees which equated to two times the height of the buildings or trees. The exception was for samples collected from forest or scrubland where the sample site was positioned equidistantly from the nearest trees or shrubs. Sampling points were located no closer than 20 m to roads or places where accumulation or wash-off of radioactive contamination was possible.

The envelope method was used to collect soil samples using a 37 mm corer down to a depth of not less than 20 cm. The mass was not less than 3 kg for the composite sample.

Soil samples were oven dried at 105°C to constant weight, homogenized and sieved through a 1 mm sieve. Sub-samples of 100 to 150 g were taken for ^{90}Sr analysis and for gamma analysis a 1 L Marinelli container was filled.

The approach used to determine ^{90}Sr activity concentration in soil samples was based on the standard ISO 18589-5:2009 (ISO2009). Strontium-90 activity was estimated through measurement of its daughter product, ^{90}Y. The radiochemical preparation involved digestion of the ashed sample in 8M HNO_3 followed by oxalate precipitation. Strontium was purified using ammonia and saturated sodium carbonate solution. The resultant strontium carbonate was dissolved in 2.5M HNO_3, Y carrier was added and stored for 2 weeks such that ^{90}Y reached equilibrium. Yttrium was then precipitated as oxalate and ^{90}Y was measured using a beta-spectrometer (SEB-70, AKP, Ukraine). The radiochemical separation yield was calculated using carriers such as stable Sr and Y measured using atomic absorption spectroscopy (Varian). Gamma analyses were conducted as described above. Quality assurance of radioanalytical procedures was based on ISO/IEC 17025 standards. The laboratory regularly participated in international and national proficiency tests (e.g. International Atomic Energy Agency).

The data set reports activity concentrations for the natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th in addition to ^{137}Cs and ^{90}Sr. The mean activity concentrations for these radionuclides were 140 Bq kg^{-1} ^{40}K, at 12 Bq kg^{-1} ^{226}Ra and 10 Bq kg^{-1} ^{232}Th. Natural background radionuclides are sometimes cited as being relatively low in the CEZ (Møller & Mousseau, 2011) though there are few data in the International literature. On the basis of soil types we could expect the CEZ to have similar natural radionuclide activity concentrations in soils as the Ivankov region. These activity concentrations do appear to be relatively low compared to average values for e.g., the UK (Beresford et al, 2008).

Caesium-137 activity concentrations in the Ivankov region soils range from 6 to 390 Bq kg^{-1} dry matter. Strontium-90 concentrations range from 1 to 160 Bq kg^{-1} dry matter. These compare to

anticipated global fallout values of approximately 4 Bq kg^{-1} ^{137}Cs and 1 Bq kg^{-1} ^{90}Sr. Activity concentrations of both ^{137}Cs and ^{90}Sr were highest in the east of the Ivankov district (Figure 4). On an aerial basis ^{137}Cs ranged from 2 to 140 kBq m^{-2} and ^{90}Sr <1 to 60 kBq m^{-2}. The ^{137}Cs:^{90}Sr ratio increased to the west and south of the Ivankov region with distance from the ChNPP (Figure 5.)

Dose rates at 1 m across the Ivankov region ranged from 0.1 to 0.24 μSv h^{-1}. From measurements of natural radionuclides in soil and using conversion coefficients by UNSCEAR (1977), a dose rate of 0.01 to 0.07 μSv h^{-1} was estimated. There was a tendency for dose rates to increase with increasing ^{137}Cs activity concentration in soils although correlation was poor (R^2=0.11 for measurements at 1 m).

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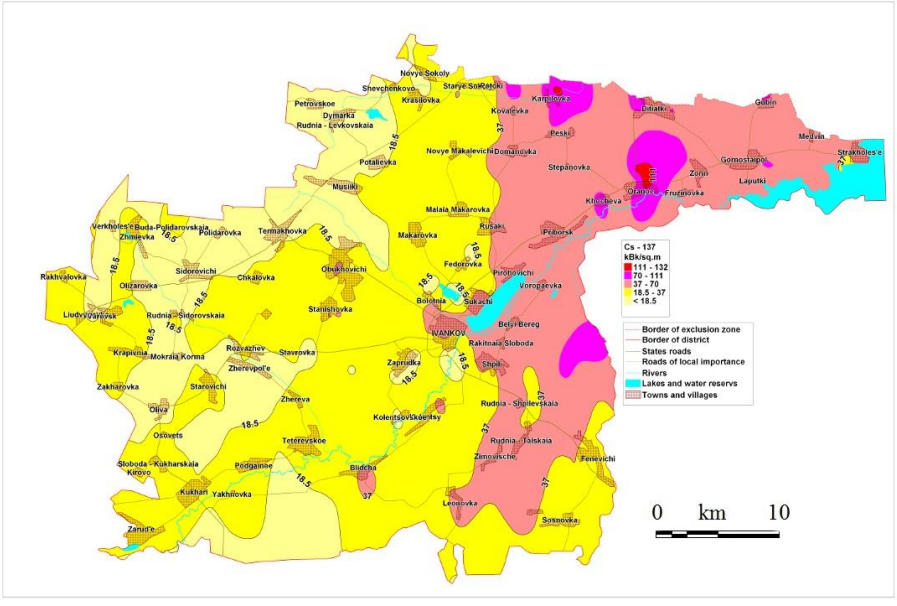

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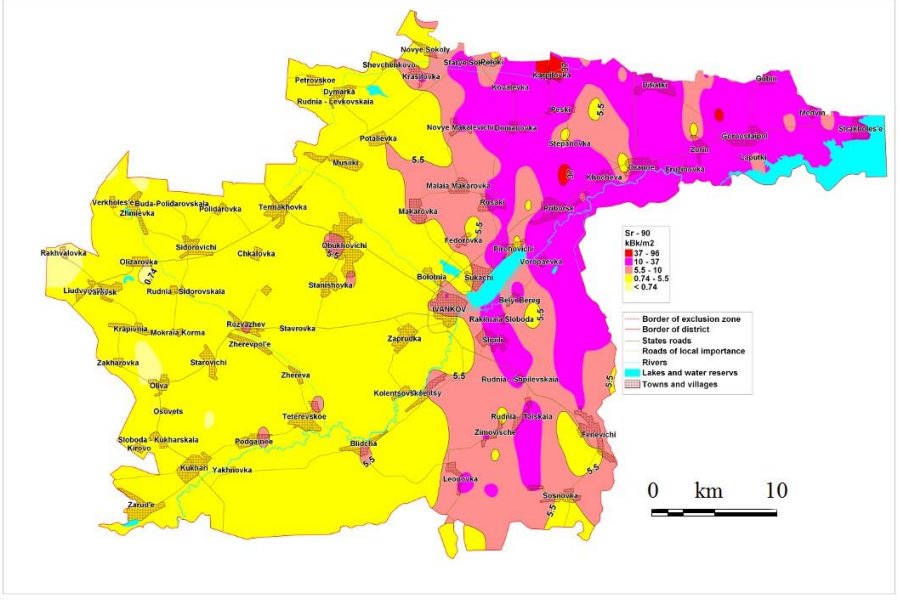

488

489 **Figure 4**. Spatial variation in ^{137}Cs (top) and ^{90}Sr (bottom) soil concentrations (kBq m^{-2}) in the Ivankov
490 district in 2015.

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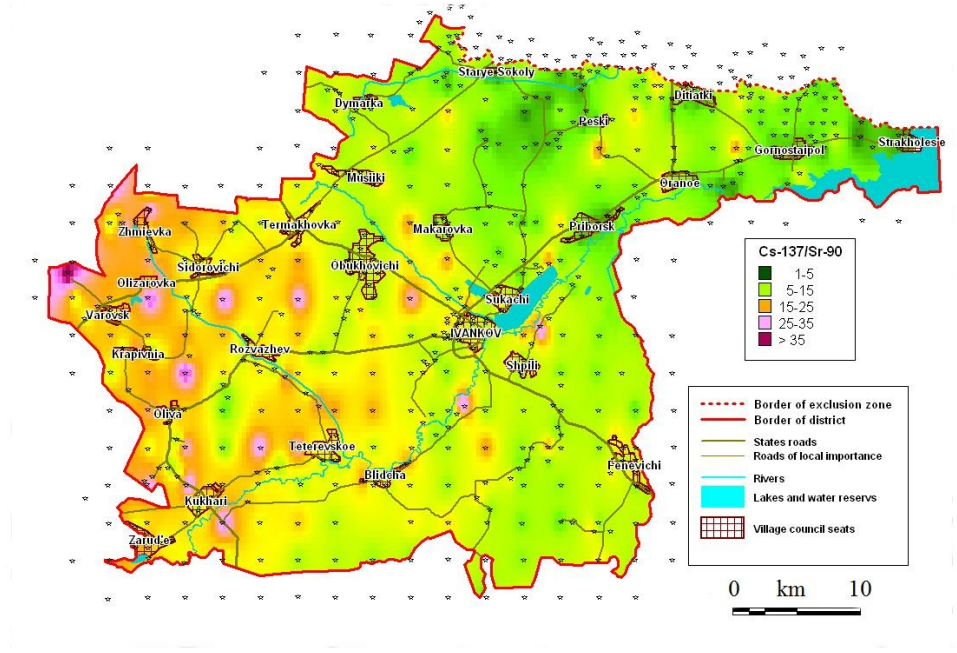

491

492 Figure 5. Spatial variation in 137Cs:90Sr ratio in the Ivankov region (sampling points black stars).

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494 **3. Access and conditions of use.**

495 All samples after gamma-spectrometry measurements are stored at NUBIP and can be made available
496 on request.

497 The data described here have a Digital Object Identifier (doi:10.5285/782ec845-2135-4698-8881-
498 b38823e533bf) and are freely available for registered users from the NERC-Environmental
499 Information Data Centre (http://eidc.ceh.ac.uk/) under the terms of the Open Government Licence.
500 The data must be fully referenced for every use as: Kashparov V., Levchuk S., Zhurba M., Protsak V.,
501 Khomutinin Yu., Beresford N.A. and Chaplow J.S. (2017): Spatial datasets of radionuclide
502 contamination in the Ukrainian Chernobyl Exclusion Zone. NERC-Environmental Information Data
503 Centre doi:10.5285/782ec845-2135-4698-8881-b38823e533bf. Supporting documentation to aid in
504 the reuse of this data is also available from the EIDC.

505

506

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