Step by step responses and amendments to paper for 4 referees.

General comments Anonymous referee 1

This paper describes in detail the spatial radioactive contamination by condensation and fuel of the fallout caused by the Chernobyl accident in 1986, i.e. comparing 144Ce and 137Cs. Making these data available is, as nicely described in chapter 3 “Use of the data” important for assessing the long term effect of radiation exposure of the surrounding landscape including wildlife. The introduction is well written and interesting to read. As a geologist I was missing that today 137Cs deposited in 1986 is commonly used in areas far away from ChNPP to date sediment layers for environmental reconstructions.

Thanks for your comments. Whilst we accept that Cs-137 is used for sediment dating we do not think adding a comment to this effect to a paper on data close to the Chernobyl accident is required.

I favor Figure 9 because here you can see the development over time (May vs. August 1986), whereas other figures show only the static situation reconstructed for 6th May 1986.

When I looked into the data provided, I found a csv table with 20 parameters listed for 491 measurements between 15.05.1987 and 08.06.1987. 49 entries had ID’s but no data. Metadata provide explanations and units as well as methods for the shown parameters. In the metadata it was described that missing values are due to water bodies. In the manuscript the authors state that the data include northing and easting, but I could not find coordinates in the data set. Is this missing by mistake?

JC No, eastings and northings are not presented. The data are presented as a radial network (i.e. angle and distance from the ChNPP are given). This was a mistake in the text which has been amended.

Overall the study is presented in a good way. My concern is that the data are presented as “corrected to 6th May 1986”, but obviously based on measurements roughly one year later in 1987. If the data are extrapolations back in time, the authors should describe in detail their methods how they calculated/corrected the values presented in the figures.

This information has been added at the end of section 2.2.

Specific comments:

Line 20-21 is this a redundant listing of “caesium-134 and caesium-137” or is there a striking difference? If so, maybe few words explaining why would help.

This is not redundant and text has been clarified

Line 22 You used exactly the same sentences as in the previous paper in ESSD. Please specify “them” in this context.

Text amended

Line 35 Please provide a rough estimate of the vast area size.

Text amended

Line 105-111 Describe how many samples and the spatial resolution of sampling (compare lines 159-160)

This paragraph discusses previous studies (not the work reported here) – text amended to hopefully remove any potential confusion.

Line 168 I could not find Northing or Easting in the data set.

JC Northing or Easting are not in the dataset so these words removed.

Line 178 More precise for “regularly” in which temporal resolution? Did sampling take place at exactly the same locations? The photo shows that the upper column of the soil and grass was sampled. How did the resampling account for accumulation on top of the contaminated layer in subsequent years?

Clarified that these data are not reported here and are not available

Line 198 – 200 – clarification on why data is not available – embargo or not processed?

As noted in the text these samples were sent to laboratories across the Soviet Union – which is no longer one country (and historically was not an ‘open’ nation)

Line 208 – 210 – uncertainty seems to be high - are there other means to check, whether the uncertainty could be limited? How did you calculate the 50%, is it standard deviation between 5 samples??

The text has been amended to describe this more clearly and a reference added to the methodology.

Figure 5b. – Why is the R2 = 0.25 not discussed?

The lower trend for 137Cs with distance was noted in text – but text now amended to acknowledge the R^2 value

Line 230 – 232, which is associated with the plot, does not include specifics.

Apologies – but we do not understand the reviewers comment. We have reviewed the text around what were lines 230-232 and cannot identify an issue.

Technical comments:

Line 33 Is this the correct citation format (Chernobyl, 1996)?

Reference replaced.

Line 70 . . .”radiocaesium”?

JC Spelling mistake corrected

Figure 1. It would help to remove blue color from legend, if it is not used, or use different color instead, because it is too close to the blue of the rivers and lakes. Is there a limit at the top of the legend?

Figure amended as requested.

Table 1. Scientific notation seems not very reader-friendly and the table seems long compared to the intended message. It would help if you could reduce it to a smaller number or highlight entries according to a meaningful criterion.

Format has been changed. However, information in the table is useful to readers and we have not further amended

Line 181-182 repetitive statement to line 162? JC duplicate 489 deleted fr

General comments Anonymous referee 2

General comments

The manuscript “Spatial radionuclide deposition data from the 60 km area around the Chernobyl nuclear power plant: results from a sampling survey in 1987” by Kashparov et al. describes the values of various radionuclides from samples obtained in 1987 in the broader region surrounding of the Chernobyl nuclear power plant. The presented dataset is very valuable by itself due to its uniqueness, but nevertheless, the authors present several options for its utilization in the future. The manuscript is well written: I especially appreciate that the authors provide an extensive introduction/background.

In the following review I only state a few comments and technical correction from which the manuscript could benefit.

The landing page of the dataset is well prepared and contains all the relevant information for future users. The dataset itself is well prepared and contains all the data, which is described in the manuscript, except geospatial data (see Specific Comments below). I compliment the authors on the carefully prepared and very clear metadata file.

I do have two comment regarding the access to the dataset and the provided data itself, which are posted in the Specific comments section of the review.

Specific comments – Manuscript

Fig. 1 is not very clear. If possible, I suggest the authors modify the original map in a way, that the figure will be readable (enlarge text, indicate all the locations that are mentioned in the manuscript).

Figure amended as requested

L168: Easting and northing data is not included in the dataset! For details see the last Specific comment regarding the dataset.

Please see response to Reviewer 1

L168-170: Personally, I think you did a really nice job in creating the Table that is included in the “Spatial_radionuclide_deposition_metadata” document, which accompanies the dataset. I suggest you to include it in this part of the manuscript or in Section 4, as it allows the reader to rapidly understand the meaning of the column headers and the used units (without reading the supporting material). I also suggest to the authors to include in the manuscript a few sentences describing the used data format (e.g. the data is presented in a form of an Excel table etc.).

We have added as supplementary information to the paper

Specific comments – Dataset

At present (2nd half of February), the data repository requires registration in order to access the dataset and accompanying metadata. As ESSD recommends “two-click” access (see Section 3.1 in

https://doi.org/10.5194/essd-10-2275-2018), I suggest the authors consult the Editor, if access in its present state is acceptable.

As the editor knows we have previous published in ESSD linking to data on the INSPIRE compliant and Core Trust Seal approved EIDC repository.

The dataset in its present state does have one shortcoming, which hinders its use by other users, as it does not contain geospatial data (despite the description at L168 in the manuscript). If the authors will not add northing and easting, they should at least state the coordinates of point zero (ChNPP), so later users can use the provided angles and distance to geolocate the datapoints. If the authors will add northing and easting, a short statement specifying the used coordinate system (possibly by stating the EPSG number) should be added for clarity in the manuscript and in the metadata description.

See response above

Technical corrections

L2: remove dot after 1987

JC Full stop removed

L14-15: Replace "Spatial radionuclide deposition data from the 60 km area around the Chernobyl nuclear power plant: results from a sampling survey in 1987" with "Spatial radionuclide deposition data from the 60 km radial area around the Chernobyl nuclear power plant, 1987", as the latter is the name of the dataset provided at https://doi.org/10.5285/a408ac9d-763e-4f4c-ba72-73bc2d1f596d.

JC Replaced here and in the title of the manuscript

L19: Should ". . . include information on sample sites, dose rate . . ." be ". . . include information from sample sites, such as: dose rate . . ."?

JC. No, we mean site information such as unique identifier and location in relation to the ChNPP. I have changed this to 'include sample site information, dose rate,…'

L33: "Chernobyl, 1996" should be "Chernobyl Nuclear Power Plant and RBMK reactors, 1996"?

JC Amended

L57-58: I suggest changing ". . . the closest observations were for a distance of more than 100 km away to the west . . ." to ". . . the closest observations were more than 100 km away to the west . . ."

JC. 'for a distance' deleted

L64: I would omit "fission products" as it is a repetition from L62.

JC 'fission products' deleted

L69: What does the "c." refer to?

JC c means circa (from Latin, meaning 'around, about, roughly, approximately') – frequently abbreviated to c. For clarity I have replaced with approximately.

L76: Should ". . . including, 40 . . ." be ". . . including 40 . . ."?

JC ',' removed

L78: ". . . Ukraine. . . ." should be ". . . Ukraine, . . ."

JC Yes, updated

L82: ". . . Ukraine. . . ." should be ". . . Ukraine, . . ."

JC Yes, updated

L102: ". . . 60-km . . ." should be ". . . 60 km . . ."

JC Corrected

L117: 14.00-17.00 hours?

JC 'hours' added for clarity

L129: I would use ". . . to identify areas . . ." instead of ". . . to identifying areas . . ."

JC 'ing' deleted

L162: "Figure" should be "Figures"

JC Changed to (Figures 3 and 4).

L163: The authors already describe the acronym UIAR in L18

JC. Agreed - text updated

L177: "Figure" should be "Figures"

JC Changed to (Figures 3 and 4).

L387: The hyperlink includes the ";" symbol and consequentially does not work. Make sure to provide a working link in the revised manuscript.

JC. I checked the link - there is no ";" and the link provided opens correctly

L389: The hyperlink includes the ";" symbol and consequentially does not work. Make sure to provide a working link in the revised manuscript.'

JC. I checked the link - the ";" is not part of the link and the link provided opens correctly

Anonymous referee 3.

General Comments

The reviewed manuscript presents the results of radionuclide activity surveys conducted on surficial soils in April and May of 1987 within a 60 km radius of the Chernobyl nuclear power plant, which experienced a catastrophic release of fuel and fission products beginning on April 26, 1986. The stated goal of the authors is to provide the resultant dataset and methodological details specifically to inform dose reconstructions oriented toward human and wildlife impact evaluations and management. Overall the manuscript is well structured and written. The authors presented a detailed overview of the accident and radionuclide emission timeline, including sufficient information to orient the reader on the fuel emission and remediation, meteorological and depositional processes that contributed to the resultant spatio-temporal pattern of fuel/fission product fallout in the study area.

Methodological details were clear, but too brief (a moderate issue), and the connectivity between the dataset and the target applications were well articulated. The data access portal is easy to use, and the dataset and attendant metadata are well organized, but spatial data reporting was insufficient (a moderate issue). Figures were used effectively throughout the manuscript, but in some cases were difficult to read (a minor issue). For these reasons (detailed below) I recommend publication after major revisions.

Specific Comments

The following moderate to minor issues should be addressed in the revised manuscript:

1. Methodological details were insufficient to fully evaluate the gamma spectrometry analyses used to estimate radionuclide activities (moderate revisions). The authors only reported on gamma spectrometer device and sample geometry, however further details on instrument calibration and spectral analysis procedures are necessary to evaluate the approach used to estimate activities and measurement error.

Text added

2. Sample location were chosen by superimposing this scheme on 'maps and [the] local landscape,' and reported using only the study's local polar coordinate system. The precision of sample locations generated in this manner is likely quite low. Furthermore, without any additional information, dataset users that convert these local coordinates to values in a geographic coordinate system will each introduce further error. I suggest that the authors report their study locations using a specified geographic coordinate system, and detail the manner in which this conversion was produced, including an estimate of location error.

Information on precision of sample location was given.

Technical Corrections

Many of the figures are difficult to read and/or have minor structural issues Please do not include any text that is unreadable because of size/resolution issues. If text is necessary, then it must be large enough to read (e.g. Figure 1 lat/long, scale, legend labels, etc.). Also, a small panel illustrating the study location in the broader geographic region would be helpful in Figure 1. In Figure 5 please label each axis in the same fashion.

Figures amended.

Author response to Anonymous Referee #4

We thank the anonymous referee for their positive feedback and constructive suggestions.

Referee comment: Yet, it is still not entirely clear to me what the affiliation of the authors was at the time of sample acquisition, and how responsibilities were distributed. An "author contribution" section, if supported by the journal, might be a good addition.

Author comment: author contribution section added below 4. Data availability section.

Author contribution. Soil samples were collected by the USSR Ministry of Defence and delivered to UIAR. Sample preparation, analysis and data interpretation was carried out by UIAR staff contributing as follows: Kashparov, Levchuk, Protsak, - sample preparation, measurement of radionuclide activity concentrations in samples; Kashparov - analysis of results; Zhurba - database creation and preparation of the manuscript figures (maps). The manuscript was prepared by Chaplow, Beresford, Kashparov, Levchuk and Zhurba.

Specific comments:

Referee comment 1) The information that I was missing most was a more detailed description of the gamma spectrometry methods. It would be important to know which emission lines were used for which nuclide; which emission probability (if included in the calibration), and which half-lives were used for correction to the release date. These missing pieces are listed in the order of importance. Emission lines are crucial; emission probabilities are optional; and for half-lives, the information is basically there, just not stated explicitly where the correction in mentioned. The more background information there is, the more likely it gets that the dataset can be made comparable with other, similar datasets. If the same emission line, same emission probability and same half-life have been used, one has a much better handle on comparability. One should also consider the aspect that this dataset may become a template for organising similar monitoring programmes in the future, in which case it would be most useful to have the right emission lines at hand.

Author comment: the manuscript has been amended to include further information on the gamma spectrometry methods - both in the methods text and also with the addition of extra information as Appendix 2/

2.2 Analysis

Using a high-purity germanium detector (GEM-30185, ORTEC, USA) and a multichannel analyser "ADCAM-300" (ORTEC, USA), the activity concentration of gamma emitting radionuclides (zirconium-95 (^{95}Zr), niobium-95 (^{95}Nb), ruthenium-106 (^{106}Ru), caesium-134 (^{134}Cs), caesium-137, (^{137}Cs) cerium-144 (^{144}Ce)) was determined in one soil sample from each sampling site. Information on gamma lines used in the analyses and radioisotope half-lives assumed for decay correction are presented in Appendix 2. Soil samples were analysed in a 1 litre Marinelli container. The other four cores were sent to different laboratories in the Soviet Union (data for these cores are unfortunately not available). Using a 1M NH_4Ac solution (pH 7) a 100 g subsample of soil was leached (solid: liquid ratio 1:5). The resultant leachate solution was shaken for 1 hour and then left at room temperature for 1 day before filtering through ashless filter paper (3-5 µm). The filtrate was then put into a suitable container for gamma analysis to determine the fraction of exchangeable ^{134,137}Cs. Measured activity concentrations were reported at 68% confidence level (which equates to one standard deviation).

Decay radiation information from the master library, integrated in spectrum analysing software tool Gelicam (EG&G ORTEC, USA), was used in gamma-analyses. Activities of ^{106}Ru and ^{137}Cs in samples were estimated via their gamma radiation emitting progenies ^{106}Rh and ^{137m}Ba, respectively.

Calibration of the spectrometer was conducted using certified standards (soil equivalent multi-radionuclide standard, V. G. Khlopin Radium Institute, Russia). Quality assurance/quality control procedures included regular monitoring of the system performance, efficiency, background and full width at half maximum (FWHM) for the ^{144}Ce, ^{137}Cs and ^{95}Nb photo peaks. To validate accuracy and precision of the method employed for ^{137}Cs activity concentration measurements, quality control samples (i.e., different matrix samples including water, soil and sawdust spiked with known certified activities of radionuclides) and Certified Reference Materials (CRM) were analysed alongside the samples. Analysis of IAEA CRMs showed satisfactory results for radionuclide mean activity concentrations with results being

within the 95% confidence interval; the limit of detection for ^{137}Cs in all samples was 1 Bq. Subsamples were analysed in a different laboratory (USSR Ministry of Defence) and results for the two laboratories were within the error of determination.

Appendix 2. Decay radiation information from the master library, integrated in spectrum analysing software tool Gelicam (EG&G ORTEC, USA), used in gamma-analyses. Activities of ^{106}Ru and ^{137}Cs in samples were estimated via their gamma radiation emitting progenies ^{106}Rh and ^{137m}Ba, respectively

Target radionuclide	Measured radionuclide	Energy, keV	Emission probability %	Half life of target radionuclides
95Zr	95Zr	724.20 756.72	44.10 54.50	64.02 days
95Nb	95Nb	765.79	99.79	34.97 days
106Ru	106Rh	621.84 1050.47	9.812 1.73	368.2 days
134Cs	134Cs	604.70 795.85	97.56 85.44	753.1 days
137Cs	137mBa	661.66	85.21	30.174 years
144Ce	144Ce	133.54	10.8	284.3 days

Referee comment 2) In line 115: A source for these very specific numbers is missing.

Author comment: Reference added to manuscript as Aleksakhin et al., 2001 and added to References section.

In the initial phase after the accident (before 7th May 1986) 99195 people were evacuated from 113 settlements including 11358 people from 51 villages in Belarus and 87 837 people from 62 settlements in Ukraine (including about 45 thousand people evacuated between 14.00-17.00 hours on April 27 from the town of Pripyat located 4 km from the ChNPP) (Aleksakhin et al., 2001).

References

Aleksakhin R.M., Buldakov L.A., Gubanov V.A., Drozhko E.G., Ilyin L.A., Kryshev I.I., Linge I.I., Romanov G.N., Savkin M .N., Saurov M.M., Tikhomirov F.A., Kholina Yu.B. 2001. Major radiation accidents: consequences and protective measures. Edited by L.A. Ilyin and V.A. Gubina. book published in Moscow, Publishing House IzdAT. 752 p. (data from p. 481). ISBN 5-86656-113-1 http://elib.biblioatom.ru/text/krupnye-radiatsionnye-avarii_2001/go,0/

Referee comment 3) In line 168: Please remove northing, easting- this is not contained in the dataset I downloaded. The angle and distance are sufficient to reconstruct the location, once a central co-ordinate is given. Northing and easting would be nice to have, but are no reason to delay publication.

Author comment: Apologies, this was a mistake; eastings and northings are not available as noted by all reviewers and this has been removed.

Referee comment 4) Figure 8 and 9: I struggle a bit with the interpolation. To me it looks like a large number of measurement points cause a local anomaly, mostly a decrease, in the interpolated values. Why is the algorithm (which algorithm, by the way) overestimating values over such large areas? Have missing values been actually excluded, or do they go in as zero?

Author comment: The reviewer's comment was not totally clear to us as they seem to contradict 'mostly a decrease' and then 'overestimation'. However, we have reviewed the text and added some information about the interpolation method and also a short clarifier in the figure legends with regard to the white area in the centre of the interpolate surface. For sites from which no samples were collected (e.g. waterbodies) nothing was included in the interpolation (I.e. no assumed value of zero was used as the reviewer questions).

As an example of the application of the data in this manner, Figure 8 presents the estimated deposition of ^{238}Pu; Figure 8 was prepared using the TIN (triangulated irregular network) interpolation within MAPINFO. The first maps of ^{90}Sr and $^{239+240}$Pu surface contamination from the Chernobyl accident were prepared in the frame of an international project (IAEA, 1992) in a similar way.

Figure 8. The fallout density of ^{238}Pu (kBq m^{-2}) corrected to 6th May 1986; estimated from measurements of ^{144}Ce in soil and estimated activity concentrations in the fuel of the ChNNP reactor number four (note no data were available for less than 5 km from ChNPP and no interpolation for this area has been attempted).

Figure 9. Spatial distribution, interpolated as for Figure 8, of effective dose rate within the 60 km zone around the ChNPP on 10th May 1986 (a) and 10th August 1986 (b). Note no data were available for less than 5 km from ChNPP and no interpolation for this area has been attempted.

End of reviewer comments, Track changed manuscript below.

Spatial radionuclide deposition data from the 60 km area around the Chernobyl nuclear power plant: results from a sampling survey in 1987.

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Abstract. The dataset "Spatial radionuclide deposition data from the 60 km area around the Chernobyl nuclear power plant: results from a sampling survey in 1987" is the latest in a series of data to be published by the Environmental Information Data Centre (EIDC) describing samples collected and analysed following the Chernobyl nuclear power plant accident in 1986. The data result from a

survey carried out by the Ukrainian Institute of Agricultural Radiology (UIAR) in April and May 1987 and include information on sample sites, dose rate, radionuclide (zirconium-95, niobium-95, ruthenium-106, caesium-134, caesium-137 and cerium-144) deposition, and exchangeable caesium-134 and 137.

The purpose of this paper is to describe the available data and methodology used for sample collection, sample preparation, and analysis~~The purpose of this paper is to describe the available data and methodology used to obtain them.~~ The data will be useful in the reconstruction of doses to human and wildlife populations, answering the current lack of scientific consensus on the effects of radiation on wildlife in the Chernobyl Exclusion zone and in evaluating future management options for Chernobyl impacted area of Ukraine and Belarus.

The data and supporting documentation are freely available from the Environmental Information Data Centre (EIDC) under the terms and conditions of the Open Government Licence (Kashparov et al., 2019 https://doi.org/10.5285/a408ac9d-763e-4f4c-ba72-73bc2d1f596d).

1 Background

The dynamics of the releases of radioactive substance from the number four reactor at the Chernobyl nuclear power plant (ChNPP) and meteorological conditions (Chernobyl, 1996) over the ten days following the accident on the 26th April 1986 resulted in a complex pattern of contamination over a vast area (~~De Cort et al., 1998~~; IAEA, 2006).

The neutron flux rise and a sharp increase in energy emission at the time of the accident resulted in heating of the nuclear fuel and leakage of fission products. Destruction of the fuel rods caused an increase in heat transfer to the surface of the superheated fuel particles and coolant, and release of radioactive substances into the atmosphere (Kashparov et al., 1996). According to the latest estimates (Kashparov et al., 2003; UNSCEAR, 2008) 100% of inert radioactive gases (largely ^{85}Kr and ^{133}Xe), 20-60% of iodine isotopes, 12-40% of ^{134,137}Cs and 1.4-4% of less volatile radionuclides (^{95}Zr, ^{99}Mo, ^{89,90} Sr, ^{103,106} Ru, ^{141,144} Ce, ^{154,155} Eu, $^{238\text{-}241}$ Pu etc.) in the reactor at the moment of the accident were released to the atmosphere.

As a result of the initial explosion on 26th April 1986, a narrow (100 km long and up to 1 km wide) relatively straight trace of radioactive fallout formed to the west of the reactor in the direction of Red Forest and Tolsty Les village (this has subsequently become known as the 'western trace'). This trace was mainly finely dispersed nuclear fuel (Kashparov et al., 2003, 2018) and could only have been formed as a consequence of the short-term release of fuel particles with overheated vapour to a comparatively low height during night time (the accident occurred at 01:24) stable atmospheric conditions. At the time of the accident, surface winds were weak and did not have any particular direction; only at a height of 1500 m was there a south-western wind with the velocity 8-10 m·s^{-1} (IAEA, 1992). Cooling of the release cloud, which included steam, resulted in the decrease of its volume, water condensation and wet deposition of radionuclides as mist (as the released steam cooled) (Saji, 2005). Later the main mechanism of fuel particle formation was the oxidation of the nuclear fuel (Kashparov et al., 1996; Salbu et al., 1994). There was an absence of data on meteorological conditions in the area of ChNPP at the time of the accident (the closest observations were for a distance of more than 100 km away to the west (Izrael et al., 1990)). There was also a lack of source term information and data on the composition of dispersed radioactive fallout. Consequently, it was not possible to make accurate predictions of deposition for the area close to the ChNPP (Talerko, 2005).

The relative leakage of fission products of uranium (IV) oxide in an inert environment at temperatures up to 2600 °C decreases in the order: volatile (Xe, Kr, I, Cs, Te, Sb, Ag), semi-volatile (Mo, Ba, Rh, Pd, Tc) and nonvolatile (Sr, Y, Nb, Ru, La, Ce, Eu) fission products (Kashparov et al., 1996; Pontillon et al., 2010). As a result of the estimated potential remaining heat release from fuel at the time of the accident (~230 W kg^{-1} U) and the heat accumulation in fuel (National Report of Ukraine, 2011), highly mobile volatile fission products (Kr, Xe, iodine, tellurium, caesium) were released from the fuel of the reactor and raised to a height of more than 1 km on 26th April 1986 and to *c*. 600 m over the following days (IAEA, 1992; Izrael et al., 1990). The greatest release of radiocesium occurred during the period of maximum heating of the reactor fuel on 26-28th April 1986 (Izrael et al., 1990). This caused the formation of the western, south-western (towards the settlements of Poliske and Bober), north-western (ultimately spreading to Sweden and wider areas of western Europe), and north-eastern condensed radioactive traces. Caesium deposition at distances from Chernobyl was largely determined by the degree of precipitation (e.g. see Chaplow et al. (2015) discussing deposition across Great Britain). After the covering of the reactor by dropping materials (including, 40 t of boron carbide, 2500 t of lead, 1800 t of sand and clay, 800 t of dolomite) from helicopters over the period 27th April–10th May 1986 (National Report of Ukraine. 2011), the ability for heat exchange of the fuel reduced, which caused a rise of temperature and consequent increase of the leakage of volatile fission products and the melting of the materials which had been dropped onto the reactor. Subsequently, there was a sharp reduction in the releases of radionuclides from the destroyed reactor on 6th May 1986 (National Report of Ukraine. 2011) due to aluminosilicates forming thermally stable compounds with many fission products and fixing caesium and strontium at high temperature (a process known prior to the Chernobyl accident (Hilpert & Nurberg, 1983)).

The changes of the annealing temperature of the nuclear fuel during the accident had a strong effect on both the ratio of different volatile fission products released (the migratory properties of Xe, Kr, I, Te, Cs increased with the temperature rise and were influenced by the presence of UO_2) and the rate of destruction of the nuclear fuel which oxidised forming micronized fuel particles (Salbu et al., 1994; Kashparov et al., 1996). The deposition of radionuclides such as ^{90}Sr, $^{238\text{-}241}Pu$, ^{241}Am, which were associated with the fuel component of the Chernobyl releases was largely limited to areas relatively close to the ChNPP. Areas receiving deposition of these radionuclides were the Chernobyl Exclusion Zone (i.e. the area of approximately 30 km radius around the ChNPP), and adjacent territories in the north of the Kiev region, in the west of the Chernihiv region, and the Bragin and Hoyniki districts of the Gomel region (Belarus). Deposition was related to the rate of the dry gravitational sedimentation of the fuel particles caused by their high density (about 8-10 $g \cdot cm^{-3}$ (Kashparov et al., 1996)); sedimentation of the lightweight condensation particles, containing iodine and caesium radioisotopes, was lower and hence these were transported further.

After the Chernobyl accident, western Europe and the Ukrainian-Belorussian Polessye were contaminated with radionuclides (IAEA, 1991, 1992, 2006). However, the area extending to 60-km around the ChNPP was the most contaminated (Izrael et al., 1990). Work on the assessment of the radiological situation within the zone started within a few days of the accident; the aim of this work was the radiation protection of the population and personnel.

Subsequently, further quantification of terrestrial dose rates was carried out by aerial-gamma survey by the State Hydrometeorological Committee together with Ministry of Geology and

Ministry of Defence of USSR (as reported in Izrael et al., 1990). ~~Further quantification of terrestrial dose rates was carried out by aerial-gamma survey by the State Hydrometeorological Committee together with Ministry of Geology and Ministry of Defence of USSR (Izrael et al., 1990).~~ Large-scale sampling of soil was also conducted, with samples analysed using gamma-spectrometry and radiochemistry methods (Izrael et al., 1990). These studies showed high variability in dose rates and radionuclide activity concentrations, with spatial patterns in both radioactive contamination and the radionuclide composition of fallout (Izrael et al., 1990). ~~The first results showed high variability in dose rates and radionuclide activity concentrations, with spatial patterns in both radioactive contamination and the radionuclide composition of fallout (Izrael et al., 1990).~~

The initial area from which the population was evacuated was based on an arbitrary decision whereby a circle around the Chernobyl nuclear power plant with a radius of 30 km was defined (IAEA, 1991). In the initial phase after the accident (before 7th May 1986) 99195 people were evacuated from 113 settlements including 11358 people from 51 villages in Belarus and 87 837 people from 62 settlements in Ukraine (including about 45 thousand people evacuated between 14.00-17.00 on April 27 from the town of Pripyat located 4 km from the ChNPP) (Aleksakhin et al., 2001).~~.~~

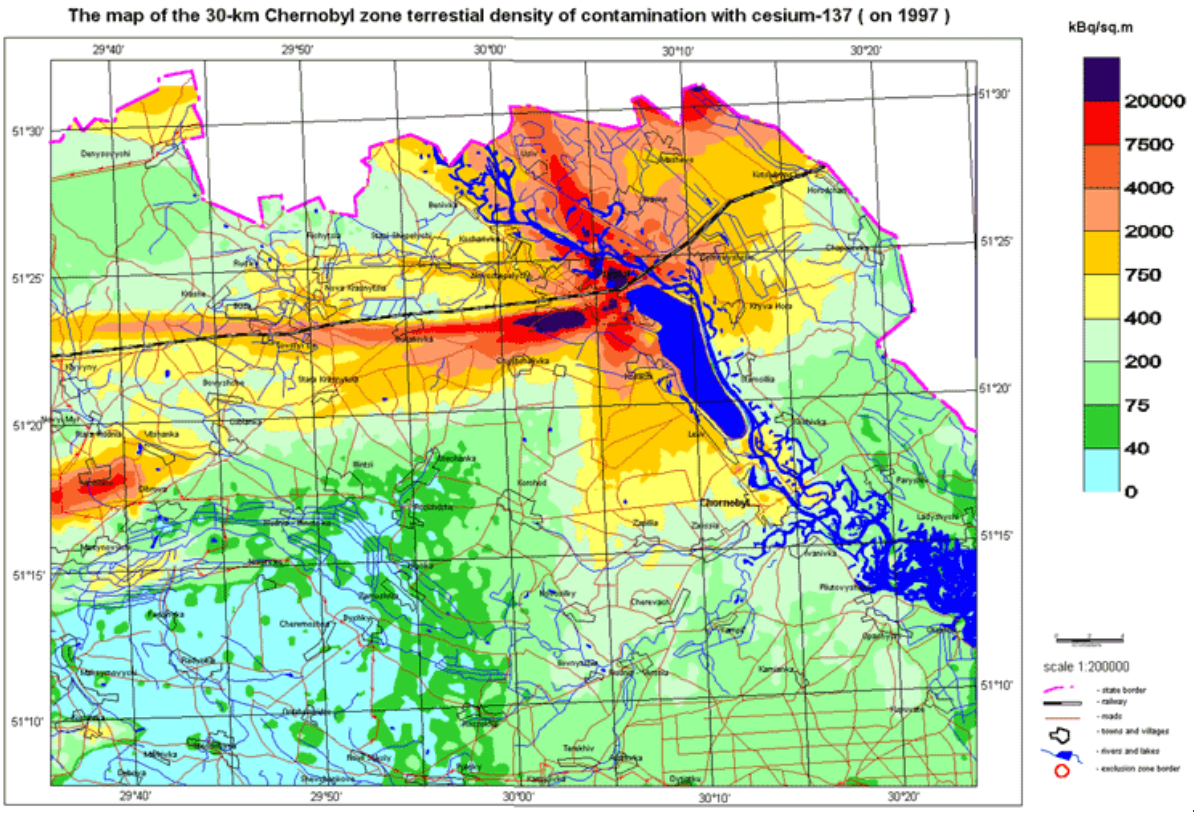
The analysis of data available in May 1986 showed that the extent of the territory with radioactive contamination where comprehensive measures were required to protect the population extended far beyond the 30-km Chernobyl Exclusion Zone (CEZ). A temporary annual effective dose limit of 100 mSv for the period from 26th April 1986 to 25th April 1987 (50 mSv from external and 50 mSv from internal exposure) was set by the USSR Ministry of Health. To identify areas outside of the CEZ where the population required evacuation, dose criteria had to be defined. It was proposed to use the average value of the dose rate of gamma radiation in open air for an area (estimated for 10th May, 1986) to help define an evacuation zone. An exposure dose rate of 5 mR h^{-1} estimated for 10th May 1986 (approximating to an effective dose rate (EDR) of gamma radiation in air of 50 μSv h^{-1}) equated to an external annual dose of 50 mSv for the period from 26th April 1986 to 25th April 1987.

At the end of May 1986 an approach to identifying areas where evacuation was required using estimated internal dose rates was proposed. This used the average density of the surface contamination of the soil with long-lived biologically significant nuclides (^{137}Cs, ^{90}Sr, ^{239,240}Pu) in a settlement and modelling to estimate the contamination of foodstuffs and hence diet. The numerical values suggested to identify areas for evacuation were: 15 Ci km^{-2} (555 kBq m^{-2}) of ^{137}Cs, 3 Ci km^{-2} (111 kBq m^{-2}) of ^{90}Sr and 0.1 Ci km^{-2} (3.7 kBq m^{-2}) of ^{239,240}Pu; this equated to an internal dose of 50 mSv over the first year after the accident.

However, in reality the main criterion for the evacuation was the exposure dose rate (R h^{-1}) and where the exposure dose rate exceeded 5 mR h^{-1} (EDR in air of about 50 μSv h^{-1}) the evacuated population were not allowed to return.

Hence, in 1986 the boundary of the population evacuation zone was set at an exposure dose rate of 5 mR h^{-1} (EDR of about 50 μSv h^{-1}). However, the ratio of short-lived gamma-emitting radionuclides (^{95}Zr, ^{95}Nb, ^{106}Ru, ^{144}Ce) deposited as fuel particles to ^{134,137}Cs deposited as condensation particles, was inconsistent across the evacuated areas. Therefore, after the radioactive decay of the short-lived radionuclides the residual dose rate across the evacuated

areas varied considerably and was largely determined by the pattern of long-lived ^{137}Cs deposition (e.g. Figure 1) (Kashparov et al., 2018).



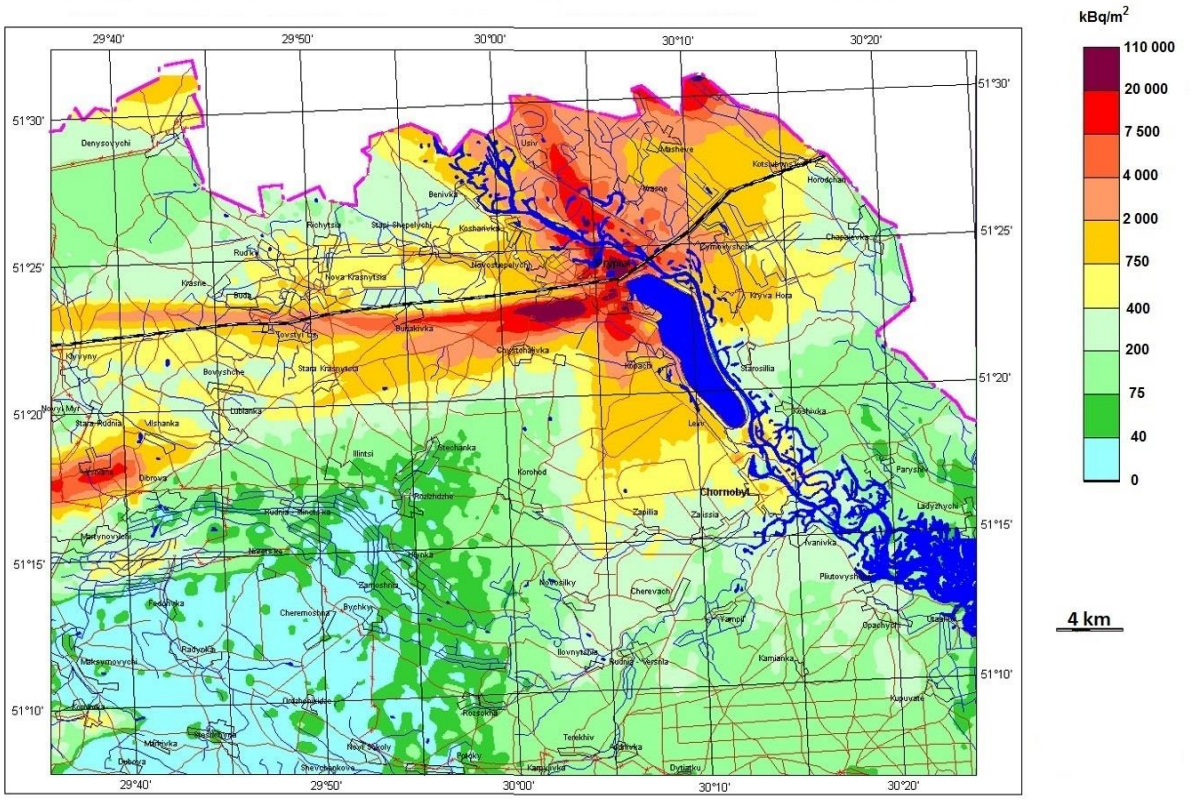


Figure 1. Caesium-137 deposition in the Ukrainian 30-km exclusion zone estimated for 1997 (from UIAR, 1998).

The first measurements of activity concentration of radionuclides in soil showed that radionuclide activity concentration ratios depended on distance and direction from the ChNPP (Izrael et al., 1990). Subsequent to this observation a detailed study of soil contamination was started in 1987 (Izrael et al., 1990). Taking into account the considerable heterogeneity of terrestrial contamination with radioactive substances in a large area, sampling along the western, southern and northern traces was carried out in stages finishing in 1988.

In 1987 the State Committee of Hydrometeorology of the USSR and the Scientific Centre of the Defence Ministry of the USSR established a survey programme to monitor radionuclide activity concentrations in soil. For this purpose, 540 sampling sites were identified at a distance of 5 km to 60 km around the ChNPP using a polar coordinate system centred on the ChNPP. Fifteen sampling sites were selected on each of the 36 rays drawn every 10 degrees (Loshchilov et al., 1991) (Figure 3, 4). Radionuclide activity concentrations in 489 soil samples collected on the radial network were determined by the Ukrainian Institute of Agricultural Radiology (UIAR) and used to calculate the radionuclide contamination density. These data are discussed in this paper and the full dataset is freely available from Kashparov et al. (2019).

2 Data

The data (Kashparov et al., 2019) include location of sample sites (~~easting, northing,~~ angle and distance from the ChNPP), dose rate, radionuclide deposition data, counting efficiency and information on exchangeable ^{134,137}Cs.

The data are presented in a table with 21 columns and 540 rows of data (plus column headings) as one Microsoft Excel Comma Separated Value File (.csv) as per the requirements of the

Environmental Information Data Centre. Appendix 1 presents an explanation of the column headings and units used in the data (Kashparov et al., 2019).

2.1 Sampling

To enable long-term monitoring and contamination mapping of the 60- km zone around the ChNPP 540 points were defined and sampled in April – May 1987. The sampling strategy used a radial network with points at every 10° (from 10° to 360°); sampling points were located at distances of 5 km, 6 km, 7 km, 8.3 km, 10 km, 12 km, 14.7 km, 17 km, 20 km, 25 km, 30 km, 37.5 km, 45 km, 52.5 km and 60 km (Figure 3, 4). The locations of sampling points were identified using military maps (1:10000 scale) ~~maps~~ and local landscape. Sites were resampled regularly until 1990 and sporadically thereafter, however, data for these subsequent samplings are not available (including to the UIAR).

Samples were not collected from points located in swamps, rivers and lakes; in total 489 samples were collected. A corer with a diameter of 14 cm was used to collect soil samples down to a depth of 5 cm from five points at each location using the envelope method (with approximately 5-10 m between sampling points) (Figure 2) (Loshchilov et al., 1991). Soil cores were retained intact during transportation to the laboratory. At each sampling point, the exposure dose rate was determined 1 m above ground level.

Figure 2. Soil sampling using a ring of 14 cm diameter to collect a 5 cm deep soil core (courtesy of UIAR, 1989).

2.2 Analysis

Using a high-purity germanium detector (GEM-30185, ORTEC, USA) and a multichannel analyser "ADCAM-300" (ORTEC, USA), the activity concentration of gamma emitting radionuclides (zirconium-95 (^{95}Zr), niobium-95 (^{95}Nb), ruthenium-106 (^{106}Ru), caesium-134 (^{134}Cs), caesium-137, (^{137}Cs) cerium-144 (^{144}Ce)) was determined in one soil sample from each sampling site. Information on gamma lines used in the analyses and radioisotope half-lives assumed for decay correction are presented in Appendix 2. Soil samples were analysed in a 1

litre Marinelli container. The other four cores were sent to different laboratories in the Soviet Union (data for these cores are unfortunately not available). Using a 1M NH_4Ac solution (pH 7) a 100 g subsample of soil was leached (solid: liquid ratio 1:5). The resultant leachate solution was shaken for 1 hour and then left at room temperature for 1 day before filtering through ashless filter paper (3-5 µm). The filtrate was then put into a suitable container for gamma analysis to determine the fraction of exchangeable $^{134,137}Cs$. Measured activity concentrations were reported at 68% confidence level (which equates to one standard deviation).
Decay radiation information from the master library, integrated in spectrum analysing software tool Gelicam (EG&G ORTEC, USA), was used in gamma-analyses. Activities of ^{106}Ru and ^{137}Cs in samples were estimated via their gamma radiation emitting progenies ^{106}Rh and ^{137m}Ba, respectively.

Calibration of the spectrometer was conducted using certified standards (soil equivalent multi-radionuclide standard, V. G. Khlopin Radium Institute, Russia). Quality assurance/quality control procedures included regular monitoring of the system performance, efficiency, background and full width at half maximum (FWHM) for the ^{144}Ce, ^{137}Cs and ^{95}Nb photo peaks. To validate accuracy and precision of the method employed for ^{137}Cs activity concentration measurements, quality control samples (i.e., different matrix samples including water, soil and sawdust spiked with known certified activities of radionuclides) and Certified Reference Materials (CRM) were analysed alongside the samples. Analysis of IAEA CRMs showed satisfactory results for radionuclide mean activity concentrations with results being within the 95% confidence interval; the limit of detection for ^{137}Cs in all samples was 1 Bq. Subsamples were analysed in a different laboratory (USSR Ministry of Defence) and results for the two laboratories were within the error of determination.

The density of soil contamination (Bq m^{-2}) was calculated from the estimated radionuclide activity concentrations in soils. It has been estimated that uncertainty from using a single soil sample (of area 0.015 m^2) to estimate the value of contamination density of a sampling site ~~(i.e. the area from which five cores were collected)~~ may be up to 50% (IAEA, 2019).

The data described in this paper (Kashparov et al., 2020) comprise exposure dose rate (mR/h), date of gamma activity measurement, density of contamination (Bq m^{-2}) of ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{134}Cs, ^{137}Cs and ^{144}Ce (with associated activity measurement uncertainties) and density of contamination of $^{134+137}Cs$ in exchangeable form. Reported radionuclide activity concentration values are for the date of measurement (samples were analysed within 1.5 months of collection).

For presentation below, radionuclide activity concentrations have been decay corrected to 6th May 1986 (the date on which releases from the reactor in-effect stopped) using the equation:
$A_T = A_0/e^{-\lambda t}$ where A_T equals the radionuclide activity concentration at the time of measurement (t); A_O is the activity concentration on 6th May 1986, and λ is the decay constant (i.e. 0.693/radionuclide physical half-life (see Table 1 for radionuclide half-lives)).
~~Using a high-purity germanium detector (GEM-30185, ORTEC, USA) and a multichannel analyser "ADCAM-300" (ORTEC, USA), the activity concentration of gamma-emitting radionuclides (^{95}Zr+^{95}Nd, ^{106}Ru, $^{134,137}Cs$, ^{144}Ce) was determined in one soil sample from each sampling site. Soil samples were analysed in a 1 litre Marinelli container. The other four cores were sent to different laboratories in the Soviet Union (data for these cores are unfortunately not available). Using a 1M NH_4Ac solution (pH 7) a 100 g subsample of soil was leached~~

~~(solid: liquid ratio 1:5). The resultant leachate solution was shaken for 1 hour and then left at room temperature for 1 day before filtering through ashless filter paper (3-5 µm). The filtrate was then put into a suitable container for gamma analysis to determine the fraction of exchangeable ^{134,137}Cs. Measured activity concentrations were reported at 68% confidence level (which equates to one standard deviation).~~

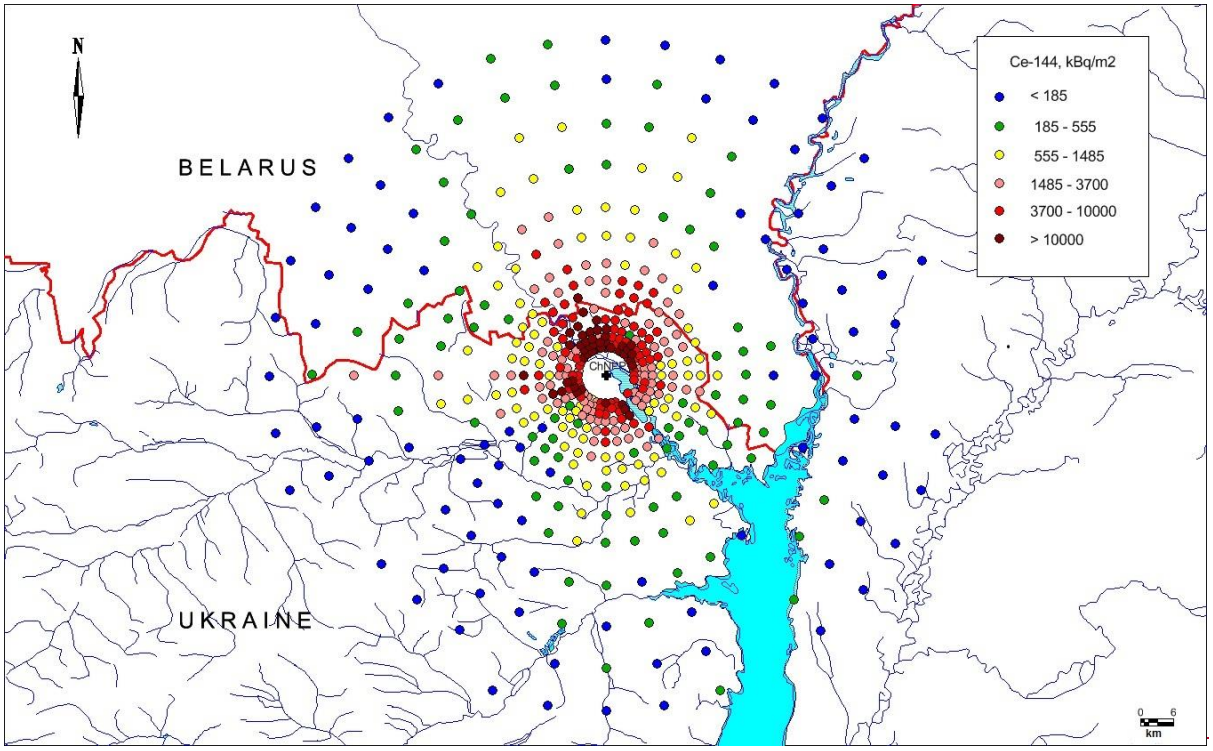
~~The density of soil contamination (Bq m^{-2}) was calculated from the estimated radionuclide activity concentrations in soils. It has been estimated that using one soil sample (of area 0.015 m^2) is used to estimate a value of contamination density of the sampling site (i.e. the area from which five cores were collected) the uncertainty may be up to 50% (Khomutinin et al., 2019).~~

~~The data described in this paper (Kashparov et al., 2019) comprise exposure dose rate (mR/h), date of gamma activity measurement, density of contamination (Bq m^{-2}) of ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{134}Cs, ^{137}Cs and ^{144}Ce (with associated activity measurement uncertainties) and density of contamination of $^{134+137}$Cs in exchangeable form. Reported radionuclide activity concentration values are for the date of measurement (samples were analysed within 1.5 months of collection).~~

2.3 Results

The contamination density of ^{144}Ce and ^{137}Cs are presented in Figures~~.~~ 3 and 4; the activity concentrations as presented in the figures have been decay corrected to 6th May~~ 1986 the date on which releases from the reactor in-effect stopped~~. The density of ^{144}Ce contamination decreased exponentially with distance (Figures 3 and 5), because ^{144}Ce was released in the fuel particles, which had a high dry deposition velocity (Kuriny et al., 1993). The fallout density of ^{144}Ce decreased by 7-9 times between the 5 km and 30 km sampling sites, and by 70-120 times between the 5 km and 60 km sampling sites (Figure 5).

The fallout density of ^{137}Cs decreased similarly to that of ^{144}Ce along the southern 'fuel trace' (Figure 5a). The contamination density of ^{137}Cs along the western trace decreased less than the ^{144}Ce contamination density due to the importance of the condensation component of the fallout in this direction (with a resultant R^2 value for the relationship between ^{137}Cs and distance lower than seen for ^{144}Ce and ^{137}Cs in different directions) (Figure 5b). The comparative decrease of ^{137}Cs contamination density along the northern trace (mixed fuel and condensation fallout) was in between that of the southern and western traces (Figure 5c) although there were caesium hotspots in the northern condensation trace (Figures 4 and 5c). The activity ratio of ^{144}Ce to ^{137}Cs decreased with distance from the ChNPP due to the condensation component being more important for ^{137}Cs; the condensation component had a lower deposition velocity compared with fuel particles (with which ^{144}Ce was associated) (Figure 6). The ratio ^{144}Ce/^{137}Cs for Chernobyl reactor fuel on 6th May 1986 can be estimated to be 15 from data presented in Table 1. The ratio was about 11 (geometric mean of 1167 measurements) in Chernobyl fuel particles larger than 10 µm due to caesium escape during high-temperature annealing (Kuriny et al., 1993). The ratio of ^{144}Ce/^{137}Cs in deposition exceeded five in the south-east and in the south up to 60 km and 30 km from the NPP respectively (Figure 6). Thus, activities of ^{134,137}Cs in the condensate and in the fuel components in these directions were of approximate equal importance. The condensation component of caesium was more important in the north and dominated in the west (Figure 8) (Loshchilov et al., 1991; Kuriny et al., 1993); the more rapidly changing ^{144}Ce/^{137}Cs ratios in these directions are reflective of this (Figure 6).



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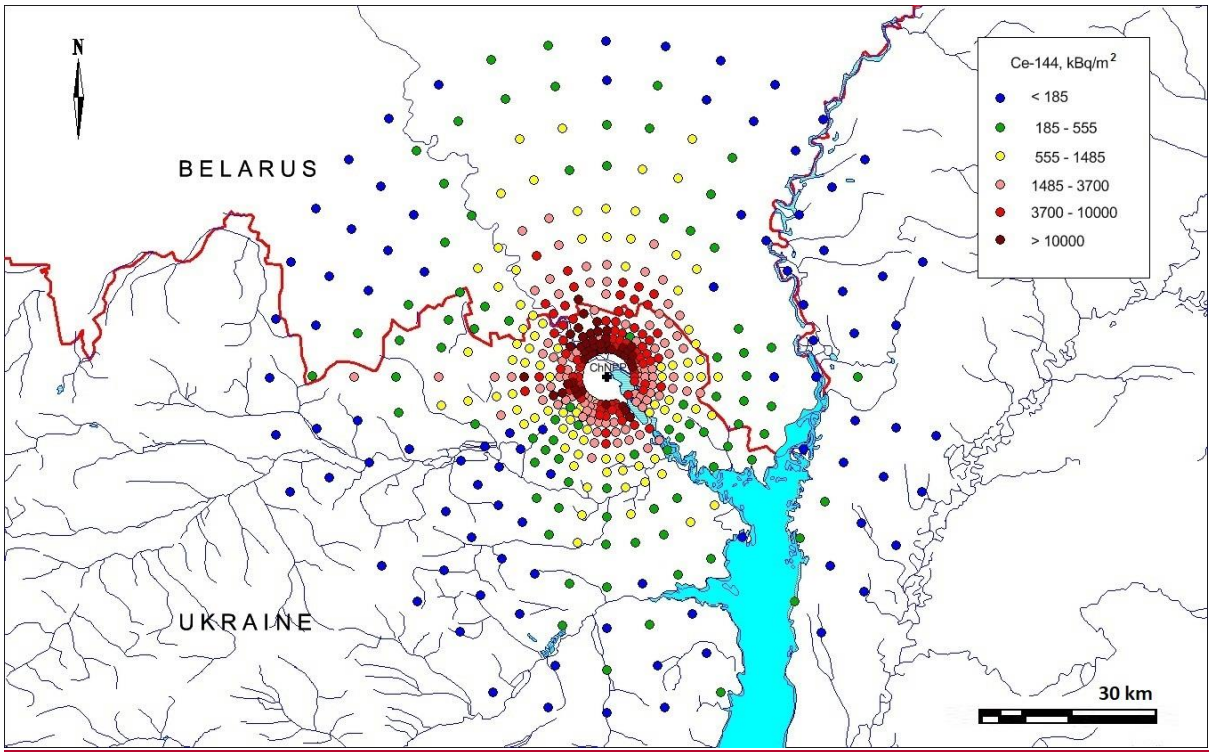

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641 Figure 3. The fallout density of ^{144}Ce (kBq/m^2) within the 60 km zone around the ChNPP
642 decay corrected to 6th May 1986.

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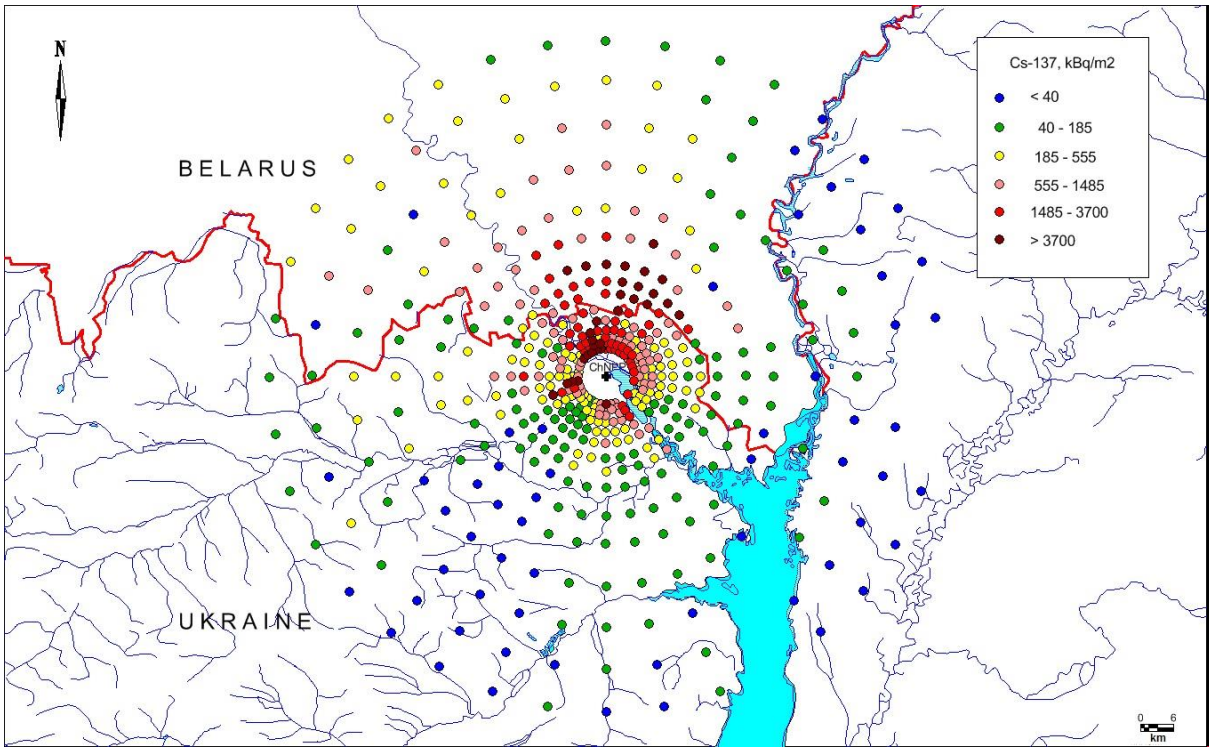

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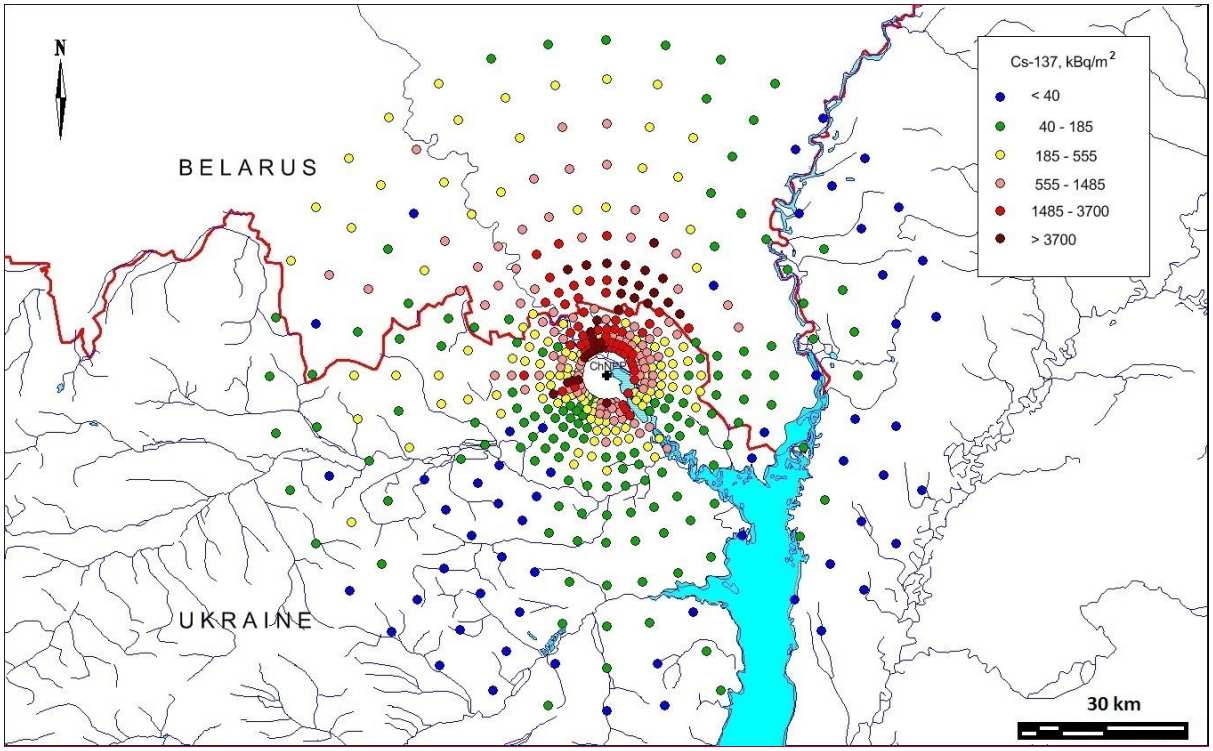

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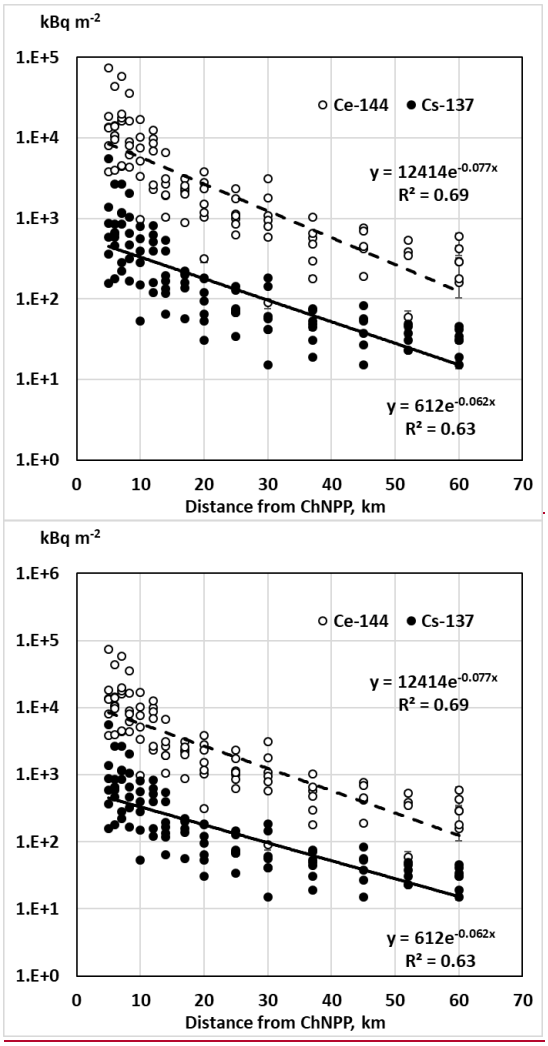
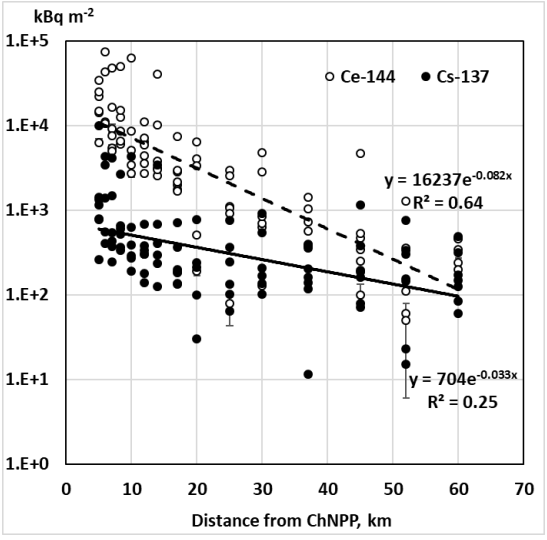
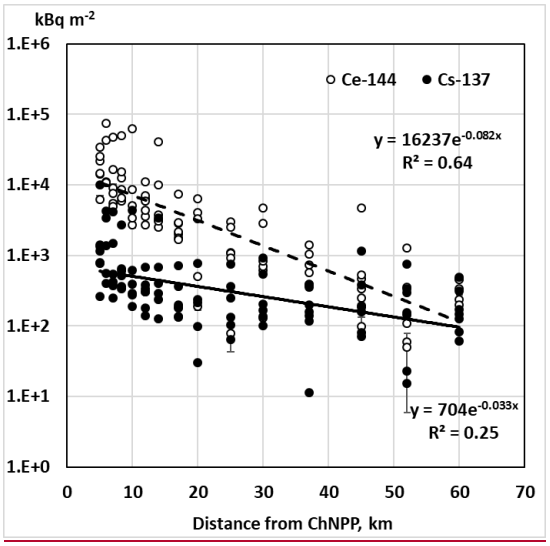
646 Figure 4. The fallout density of ^{137}Cs (kBq/m^2) within the 60 km zone around the ChNPP
647 decay corrected to 6th May 1986.


kBq m-2
1.E+5
1.E+4
1.E+3
1.E+2
1.E+1
1.E+0
Ce-144
Cs-137
y = 12414e-0.077x
R² = 0.69
y = 612e-0.062x
R² = 0.63
0
10
20
30
40
50
60
70
Distance from ChNPP, km
kBq m-2
1.E+6
1.E+5
1.E+4
1.E+3
1.E+2
1.E+1
1.E+0
Ce-144
Cs-137
y = 12414e-0.077x
R² = 0.69
y = 612e-0.062x
R² = 0.63
0
10
20
30
40
50
60
70
Distance from ChNPP, km

a


kBq m-2
1.E+5
1.E+4
1.E+3
1.E+2
1.E+1
1.E+0
Ce-144
Cs-137
y = 16237e-0.082x
R² = 0.64
y = 704e-0.033x
R² = 0.25
0
10
20
30
40
50
60
70
Distance from ChNPP, km



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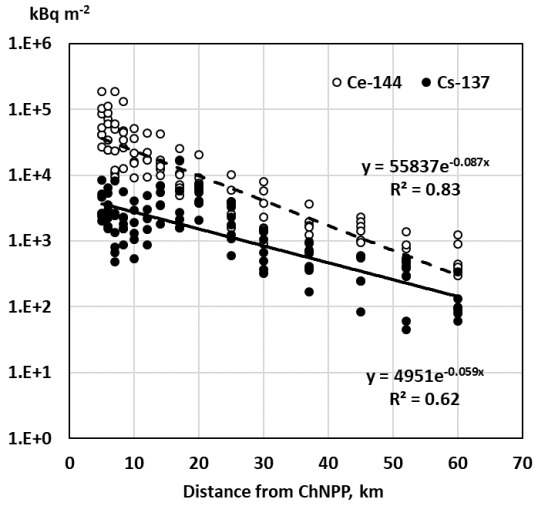


c

Figure 5. Relationship between fallout density of ^{144}Ce (1) and ^{137}Cs (2) and distance from the ChNPP towards the south (a) (150-210°), the west (b) (240-300°) and the north (c) (330-30°).

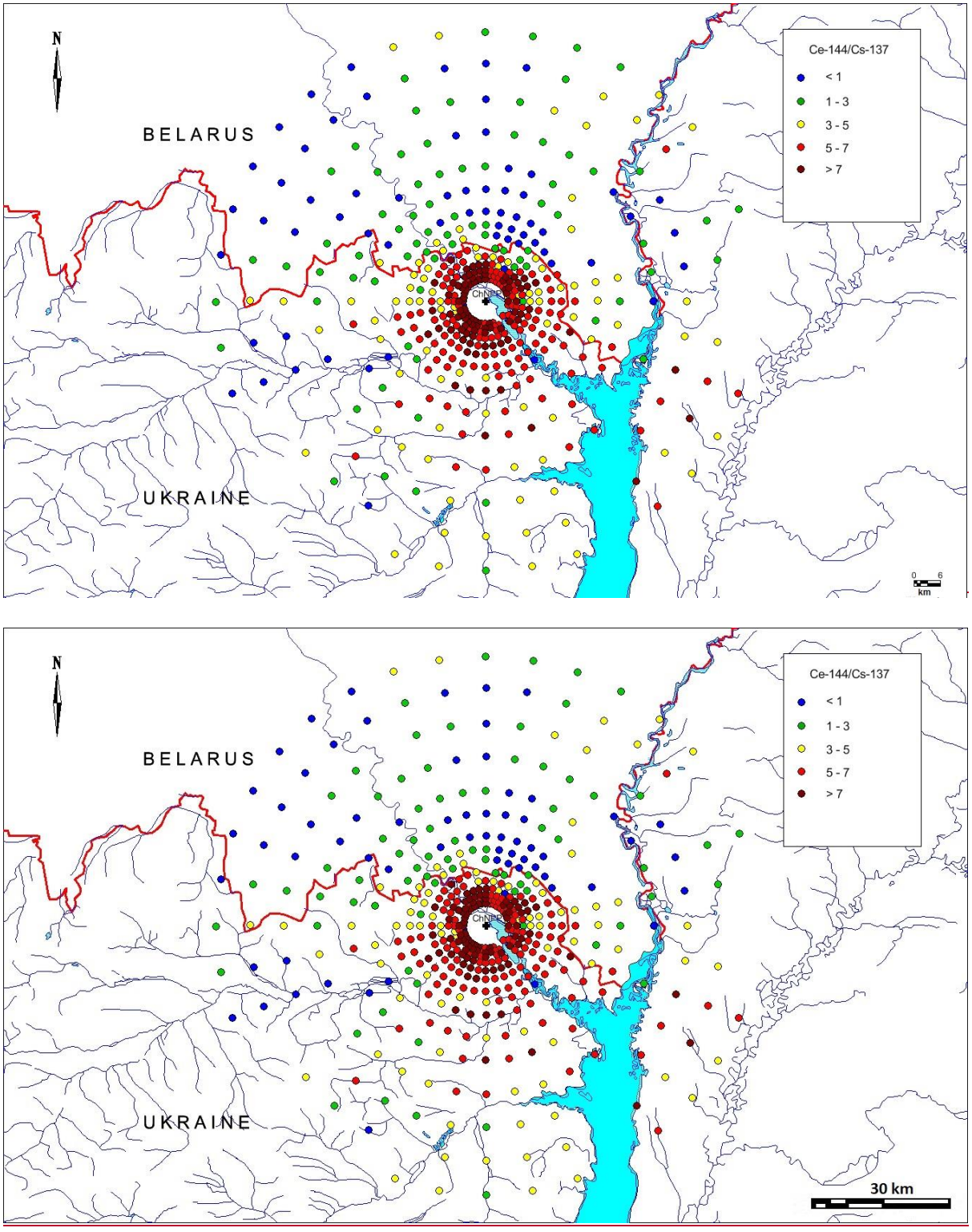


Figure 6. $^{144}Ce/^{137}Cs$ ratio within the 60 km zone around the ChNPP decay corrected to 6th May 1986.

Table 1. The average activity concentrations of radionuclides with half-life ($T_{1/2}$) >1 day estimated in the fuel of the ChNPP number four reactor recalculated for 6th May 1986 (Begichev et al., 1993).

~~Radionuclide~~	~~Half-life (days)~~	~~Average activity concentration (Bq g^{-1})~~	~~Radionuclide~~	~~Half-life (days)~~	~~Average activity concentration (Bq g^{-1})~~
~~^{75}Se~~	~~1.2E+02~~	~~5.40E+06~~	~~^{132}Te~~	~~3.3E+00~~	~~2.40E+10~~
~~^{76}As~~	~~1.1E+00~~	~~1.70E+07~~	~~^{133}Xe~~	~~5.2E+00~~	~~3.40E+10~~
~~^{77}As~~	~~1.6E+00~~	~~4.10E+07~~	~~^{134}Cs~~	~~7.6E+02~~	~~8.90E+08~~
~~^{82}Br~~	~~1.5E+00~~	~~1.80E+09~~	~~^{135}Cs~~	~~5.5E+07~~	~~1.90E+04~~
~~^{85}Kr~~	~~3.9E+03~~	~~1.50E+08~~	~~^{136}Cs~~	~~1.3E+01~~	~~3.30E+10~~
~~^{86}Rb~~	~~1.9E+01~~	~~8.70E+09~~	~~^{137}Cs~~	~~1.1E+04~~	~~1.40E+09~~
~~^{89}Sr~~	~~5.1E+01~~	~~2.10E+10~~	~~^{140}Ba~~	~~1.3E+01~~	~~3.20E+10~~
~~^{90}Sr~~	~~1.1E+04~~	~~1.20E+09~~	~~^{141}Ce~~	~~3.3E+01~~	~~2.90E+10~~
~~^{90}Y~~	~~1.1E+04~~	~~1.20E+09~~	~~^{143}Ce~~	~~1.4E+00~~	~~2.90E+10~~
~~^{91}Y~~	~~5.9E+01~~	~~2.60E+10~~	~~^{144}Ce~~	~~2.8E+02~~	~~2.10E+10~~
~~^{95}Zr~~	~~6.4E+01~~	~~3.10E+10~~	~~^{147}Nd~~	~~1.1E+01~~	~~1.10E+10~~
~~^{95}Nb~~	~~3.5E+01~~	~~3.00E+10~~	~~^{147}Pm~~	~~9.5E+02~~	~~4.20E+09~~
~~^{96}Nb~~	~~9.8E-01~~	~~3.10E+10~~	~~^{148m}Pm~~	~~4.1E+01~~	~~8.50E+09~~
~~^{99}Mo~~	~~2.7E+00~~	~~3.20E+10~~	~~^{149}Nd~~	~~2.2E+00~~	~~5.80E+09~~
~~^{99m}Tc~~	~~2.7E+00~~	~~2.80E+10~~	~~^{151}Pm~~	~~1.2E+00~~	~~2.60E+09~~
~~^{103}Ru~~	~~3.9E+01~~	~~2.00E+10~~	~~^{151}Sm~~	~~3.3E+04~~	~~3.40E+07~~
~~^{105}Rh~~	~~1.5E+00~~	~~1.00E+10~~	~~^{153}Sm~~	~~1.9E+00~~	~~1.10E+09~~
~~^{106}Ru~~	~~3.7E+02~~	~~4.50E+09~~	~~^{154}Eu~~	~~3.1E+03~~	~~3.70E+07~~
~~^{110m}Ag~~	~~2.5E+02~~	~~5.30E+08~~	~~^{155}Eu~~	~~1.7E+03~~	~~4.85E+07~~
~~^{111}Ag~~	~~7.5E+00~~	~~4.40E+08~~	~~^{156}Eu~~	~~1.5E+01~~	~~1.90E+08~~
~~^{115m}In~~	~~1.9E-01~~	~~8.60E+07~~	~~^{160}Tb~~	~~7.2E+01~~	~~1.00E+07~~
~~^{117m} Sn~~	~~1.4E+01~~	~~8.30E+07~~	~~^{237}Np~~	~~7.8E+08~~	~~1.40E+03~~
~~^{123}Sn~~	~~1.3E+02~~	~~9.90E+07~~	~~^{239}Np~~	~~2.4E+00~~	~~3.10E+11~~
~~^{124}I~~	~~4.2E+00~~	~~1.40E+08~~	~~^{236}Pu~~	~~1.0E+03~~	~~6.00E+02~~
~~^{125}Sb~~	~~1.0E+03~~	~~7.80E+07~~	~~^{238}Pu~~	~~3.2E+04~~	~~6.80E+06~~
~~^{125m}Te~~	~~5.8E+01~~	~~1.60E+07~~	~~^{239}Pu~~	~~8.8E+06~~	~~5.00E+06~~
~~^{126m}Sb~~	~~1.2E+01~~	~~4.40E+08~~	~~^{240}Pu~~	~~2.4E+06~~	~~7.80E+06~~
~~^{126}Sb~~	~~1.2E+01~~	~~6.10E+07~~	~~^{241}Pu~~	~~5.1E+03~~	~~9.60E+08~~
~~^{127}Sb~~	~~3.8E+00~~	~~1.10E+09~~	~~^{242}Pu~~	~~1.4E+08~~	~~1.50E+04~~
~~^{127}Te~~	~~1.1E+02~~	~~8.90E+08~~	~~^{241}Am~~	~~1.6E+05~~	~~8.70E+05~~
~~^{129m}Te~~	~~3.3E+01~~	~~5.50E+09~~	~~^{243}Am~~	~~2.7E+06~~	~~5.10E+04~~
~~^{131}I~~	~~8.0E+00~~	~~1.60E+10~~	~~^{242}Cm~~	~~1.6E+02~~	~~2.30E+08~~
~~^{131m}Xe~~	~~1.2E+01~~	~~1.80E+08~~	~~^{244}Cm~~	~~6.6E+03~~	~~2.20E+06~~

668

Radionuclide	Half-life (days)	Average activity concentration (Bq g^{-1})	Radionuclide	Half-life (days)	Average activity concentration (Bq g^{-1})
^{75}Se	1.2 x 10^{2}	5.4 x 10^{6}	^{132}Te	3.3 x 10^{0}	2.4 x 10^{10}
^{76}As	1.1 x 10^{0}	1.7 x 10^{7}	^{133}Xe	5.2 x 10^{0}	3.4 x 10^{10}

^{77}As	1.6 x 10^{0}	4.1 x 10^{7}	^{134}Cs	7.6 x 10^{2}	8.9 x 10^{8}
^{82}Br	1.5 x 10^{0}	1.8 x 10^{9}	^{135}Cs	5.5 x 10^{7}	1.9 x 10^{4}
^{85}Kr	3.9 x 10^{3}	1.5 x 10^{8}	^{136}Cs	1.3 x 10^{1}	3.3 x 10^{10}
^{86}Rb	1.9 x 10^{1}	8.7 x 10^{9}	^{137}Cs	1.1 x 10^{4}	1.4 x 10^{9}
^{89}Sr	5.1 x 10^{1}	2.1 x 10^{10}	^{140}Ba	1.3 x 10^{1}	3.2 x 10^{10}
^{90}Sr	1.1 x 10^{4}	1.2 x 10^{9}	^{141}Ce	3.3 x 10^{1}	2.9 x 10^{10}
^{90}Y	1.1 x 10^{4}	1.2 x 10^{9}	^{143}Ce	1.4 x 10^{0}	2.9 x 10^{10}
^{91}Y	5.9 x 10^{1}	2.6 x 10^{10}	^{144}Ce	2.8 x 10^{2}	2.1 x 10^{10}
^{95}Zr	6.4 x 10^{1}	3.1 x 10^{10}	^{147}Nd	1.1 x 10^{1}	1.1 x 10^{10}
^{95}Nb	3.5 x 10^{1}	3.0 x 10^{10}	^{147}Pm	9.5 x 10^{2}	4.2 x 10^{9}
^{96}Nb	9.8 x 10^{1}	3.1 x 10^{10}	^{148m}Pm	4.1 x 10^{1}	8.5 x 10^{9}
^{99}Mo	2.7 x 10^{0}	3.2 x 10^{10}	^{149}Nd	2.2 x 10^{0}	5.8 x 10^{9}
^{99m}Tc	2.7 x 10^{0}	2.8 x 10^{10}	^{151}Pm	1.2 x 10^{0}	2.6 x 10^{9}
^{103}Ru	3.9 x 10^{1}	2.0 x 10^{10}	^{151}Sm	3.3 x 10^{4}	3.4 x 10^{7}
^{105}Rh	1.5 x 10^{0}	1.0 x 10^{10}	^{153}Sm	1.9 x 10^{0}	1.1 x 10^{9}
^{106}Ru	3.7 x 10^{2}	4.5 x 10^{9}	^{154}Eu	3.1 x 10^{3}	3.7 x 10^{7}
^{110m}Ag	2.5 x 10^{2}	5.3 x 10^{8}	^{155}Eu	1.7 x 10^{3}	4.85 x 10^{7}
^{111}Ag	7.5 x 10^{0}	4.4 x 10^{8}	^{156}Eu	1.5 x 10^{1}	1.9 x 10^{8}
^{115m}In	1.9 x 10^{1}	8.6 x 10^{7}	^{160}Tb	7.2 x 10^{1}	1.0 x 10^{7}
^{117m} Sn	1.4 x 10^{1}	8.3 x 10^{7}	^{237}Np	7.8 x 10^{8}	1.4 x 10^{3}
^{123}Sn	1.3 x 10^{2}	9.9 x 10^{7}	^{239}Np	2.4 x 10^{0}	3.1 x 10^{11}
^{124}I	4.2 x 10^{0}	1.4 x 10^{8}	^{236}Pu	1.0 x 10^{3}	6.0 x 10^{2}
^{125}Sb	1.0 x 10^{3}	7.8 x 10^{7}	^{238}Pu	3.2 x 10^{4}	6.8 x 10^{6}
^{125m}Te	5.8 x 10^{1}	1.6 x 10^{7}	^{239}Pu	8.8 x 10^{6}	5.0 x 10^{6}
^{126m}Sb	1.2 x 10^{1}	4.4 x 10^{8}	^{240}Pu	2.4 x 10^{6}	7.8 x 10^{6}
^{126}Sb	1.2 x 10^{1}	6.1 x 10^{7}	^{241}Pu	5.1 x 10^{3}	9.6 x 10^{8}
^{127}Sb	3.8 x 10^{0}	1.1 x 10^{9}	^{242}Pu	1.4 x 10^{8}	1.5 x 10^{4}
^{127}Te	1.1 x 10^{2}	8.9 x 10^{8}	^{241}Am	1.6 x 10^{5}	8.7 x 10^{5}
^{129m}Te	3.3 x 10^{1}	5.5 x 10^{9}	^{243}Am	2.7 x 10^{6}	5.1 x 10^{4}
^{131}I	8.0 x 10^{0}	1.6 x 10^{10}	^{242}Cm	1.6 x 10^{2}	2.3 x 10^{8}
^{131m}Xe	1.2 x 10^{1}	1.8 x 10^{8}	^{244}Cm	6.6 x 10^{3}	2.2 x 10^{6}

A good correlation (R^2=0.98) was observed between fallout densities of ^{95}Zr (estimated from the activity concentration of daughter product ^{95}Nb)[1] and ^{144}Ce (Figure 7a) because both radionuclides were released and deposited as fuel particles (Kuriny et al., 1993; Kashparov et al., 2003; Kashparov, 2003). The fallout density ratio of ^{144}Ce/^{95}Zr=0.73±0.05, decay corrected to 6th May 1986, was similar to that estimated for Chernobyl reactor fuel (^{144}Ce/^{95}Zr=0.68) (Table 1).

The activity ratio of ^{144}Ce to ^{106}Ru in fallout was correlated (R^2=0.93) and was 3.9±0.4 decay corrected to 6th May 1986 (Figure 7b). The value was close to the ratio of ^{144}Ce/^{106}Ru estimated for fuel in the ChNPP number four reactor (4.7) (Table 1). Excess ^{106}Ru activity relative to

[1] Niobium-95 ($T_{1/2}$=34 days) is the daughter radionuclide of ^{95}Zr ($T_{1/2}$=65 days) and the ratio of their activities at an equilibrium equals ^{95}Nb/^{95}Zr=2.1.

^{144}Ce activity in some soil samples was observed likely due to the presence of "ruthenium particles" (a matrix of iron group elements with a high content of ^{103,106}Ru (Kuriny et al., 1993; Kashparov et al., 1996)).

There was a weak correlation (R^2=0.41) between ^{144}Ce and ^{137}Cs activities in the fallout because, as already discussed, caesium was largely deposited as condensation particles while cerium was deposited in fuel particles only. However, in highly contaminated areas close to the ChNPP a significant part of the ^{137}Cs was deposited as fuel particles and the activity ratio of ^{144}Ce/^{137}Cs of 9.1 (Figure 7c) broadly corresponded to that of 15 in the reactor fuel (Table 1).

Different radioisotopes of caesium escaped from nuclear fuel and were deposited in the same way. This similar behaviour of ^{134}Cs and ^{137}Cs resulted in a strong correlation (R^2=0.99) between their activities in soil samples and the ratio of ^{134}Cs/^{137}Cs=0.57±0.07 was similar to that estimated for the reactor fuel (0.64, Table 1).

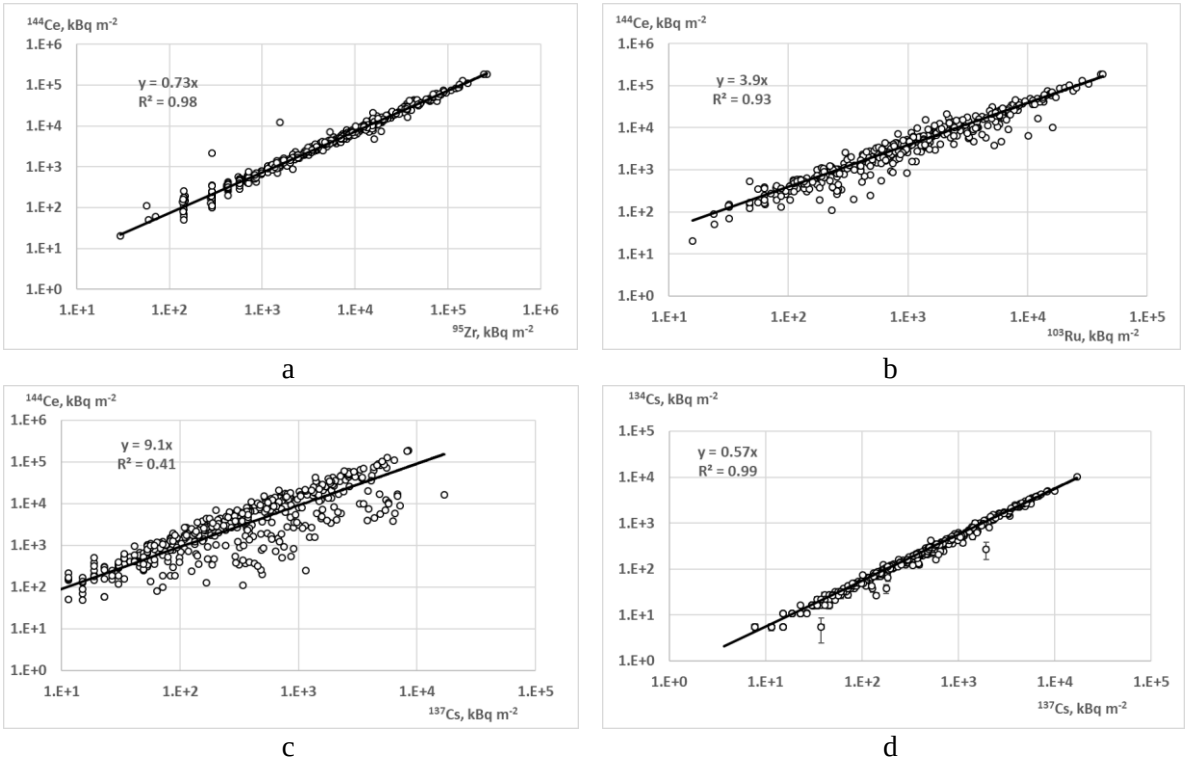


Figure 7. Correlation between deposition densities of different radionuclides decay corrected to 6th May 1986.

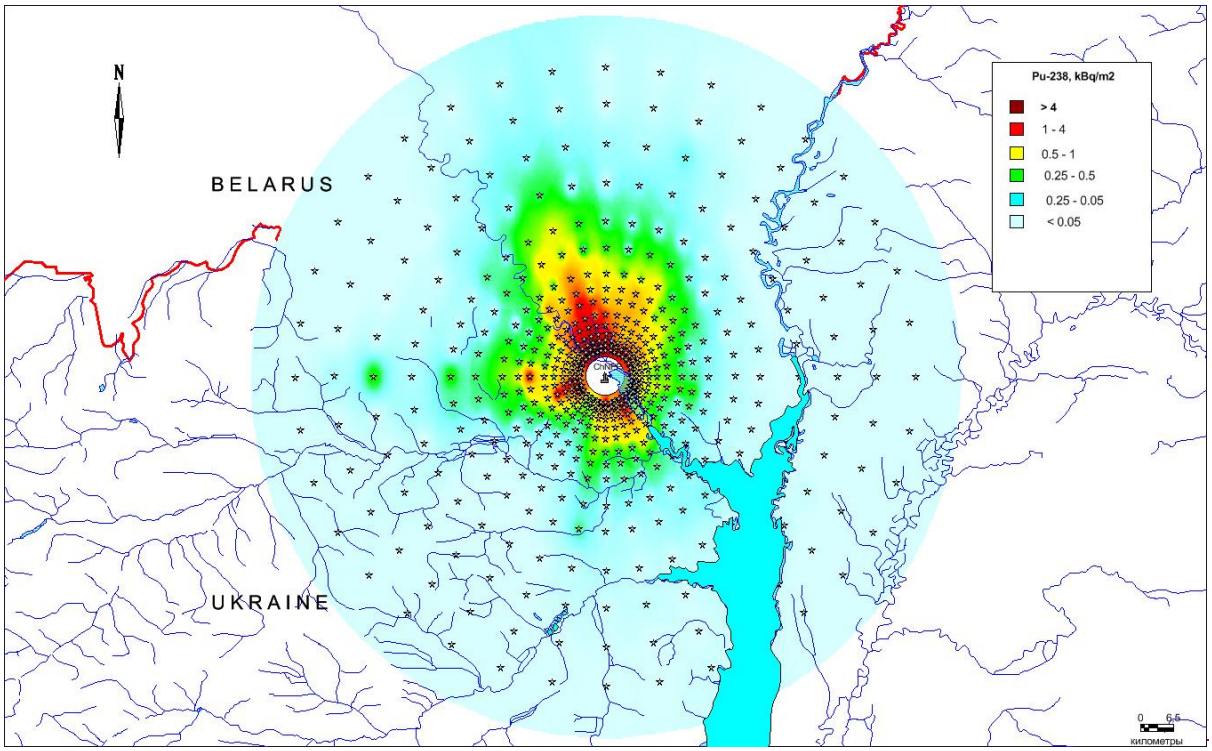
3 Use of the data

Apart from adding to the available data with which contamination maps for the CEZ and surrounding areas can be generated (e.g. Kashparov et al., 2018) the data discussed in this paper can be used to make predictions for less well studied radionuclides.

The determination of beta and alpha emitting radionuclides in samples requires radiochemical extraction which is both time consuming and relatively expensive. Large-scale surveys of the deposition of alpha and beta emitting radionuclides are therefore more difficult than those for

gamma-emitting radionuclides and are not conducive with responding to a large-scale accident such as that which occurred at Chernobyl. Above we have demonstrated that the deposition behaviour of different groups of radionuclides was determined by the form in which they were present in the atmosphere (i.e. associated with fuel particles or condensation particles).

We propose that ^{144}Ce deposition can be used as a marker of the deposition of fuel particles; fuel particles were the main deposition form of nonvolatile radionuclides (i.e. Sr, Y, Nb, Ru, La, Ce, Eu, Np, Pu, Am, Cm). Therefore, using ^{144}Ce activity concentrations determined in soil samples and estimates of the activities in reactor fuel, we can make estimates of the deposition of radionuclides such as Pu-isotopes and Cm that have been relatively less studied. For example, activity ratios of ^{238}Pu, ^{239}Pu ^{240}Pu and ^{241}Pu to ^{144}Ce, at the time of measurement would be $8.4\times10^{-4,}$ 6.2×10^{-4}, 9.7×10^{-4} and 1.1×10^{-1} respectively (estimated by decay correcting data presented in Table 1). Fallout densities of these plutonium isotopes can therefore be calculated for all sampling points where deposition density of ^{144}Ce was measured either in this study (e.g. Figure 3) or in other datasets. As an example of the application of the data in this manner, Figure 8 presents the estimated deposition of ^{238}Pu. The first maps of ^{90}Sr and $^{239+240}$Pu surface contamination from the Chernobyl accident were prepared in the frame of an international project (IAEA, 1992) in a similar way.



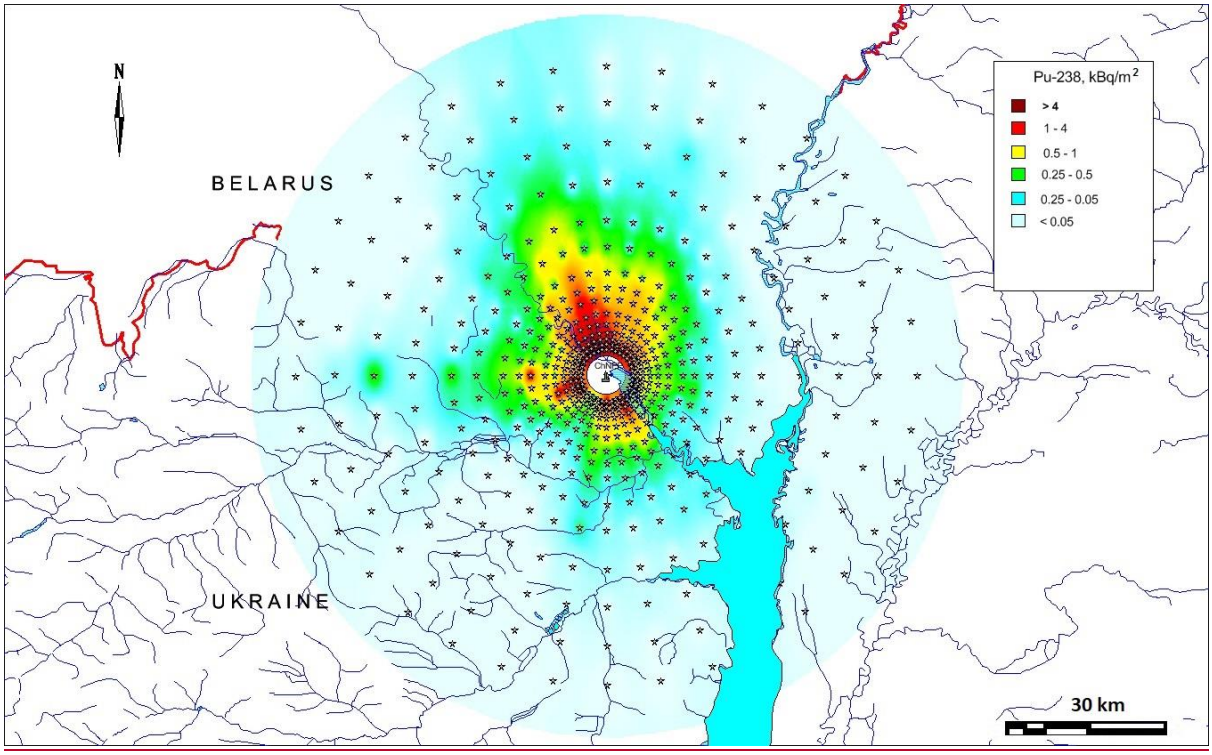


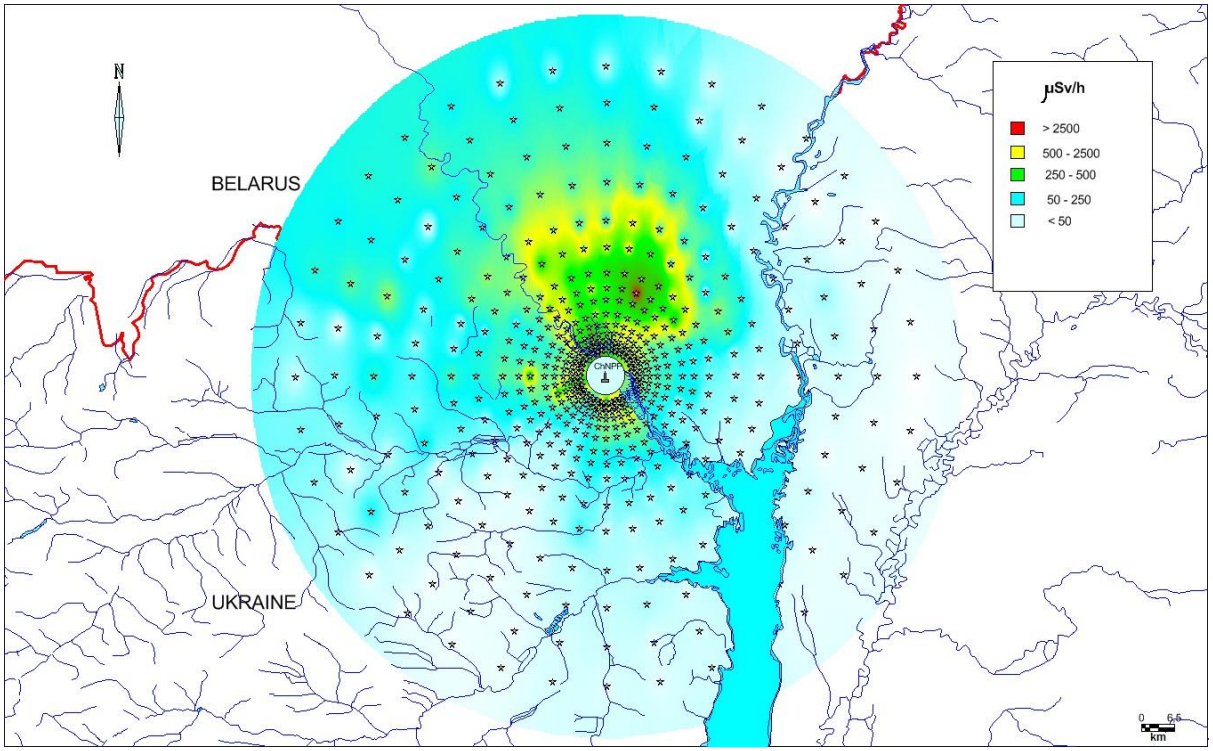
Figure 8. The fallout density of ^{238}Pu (kBq m^{-2}) corrected to 6th May 1986; estimated from measurements of ^{144}Ce in soil and estimated activity concentrations in the fuel of the ChNNP reactor number four (note no data were available for less than 5 km from ChNPP and no interpolation for this area has been attempted).

~~Figure 8. The fallout density of ^{238}Pu (kBq m^{-2}) corrected to 6th May 1986; estimated from measurements of ^{144}Ce in soil and estimated activity concentrations in the fuel of the ChNNP reactor number four.~~

The dynamic spatial distribution of gamma dose rate can be reconstructed using the data on radionuclide contamination densities (Kashparov et al, 2019) in combination with the ratios between activities of radionuclides in fuel and in condensed components of Chernobyl fallout (Table 1) and also dose coefficients for exposure to contaminated ground surfaces, (Sv s^{-1}/Bq m^{-2}) (Eckerman & Ryman, 1993). Five days after deposition the following radionuclides were major contributors (about 95 %) to gamma dose rate: ^{136}Cs, ^{140}La, ^{239}Np, ^{95}Nb, ^{95}Zr, ^{131}I, ^{148m}Pm, ^{103}Ru, ^{140}Ba, ^{132}Te. After three months the major external dose contributors were: ^{95}Nb, ^{95}Zr, ^{148m}Pm, ^{134}Cs, ^{103}Ru, ^{137m}Ba, ^{110m}Ag, ^{136}Cs, ^{106}Rh. Three years after the major contributors were ^{137m}Ba, ^{134}Cs, ^{106}Rh, ^{110m}Ag, ^{154}Eu. At the present time the gamma dose can be estimated to be mainly (99%) due to the gamma-emitting daughter radionuclide of ^{137}Cs (^{137m}Ba). Bondar (2015) from a survey of the CEZ along the Ukrainian-Belarussian border, showed a good relationship between ^{137}Cs contamination ($A_{Cs\text{-}137}$, in the range of 17-7790 kBq m^{-2}) and ambient dose rates at 1m above the ground (D_{ext}, in the range of 0.1-6.0 μSv h^{-1}**)**. The relationship was described by following equation with correlation coefficient of 0.99:

$$D_{ext} = 0.0009 \cdot A_{Cs\text{-}137} + 0.14.$$

As an example of the application of the data in this manner, Figure~~.~~ 9 presents the estimated external effective gamma dose rate five and 95 days after the cessation of the radioactive releases from the reactor on 6th May 1986.



747

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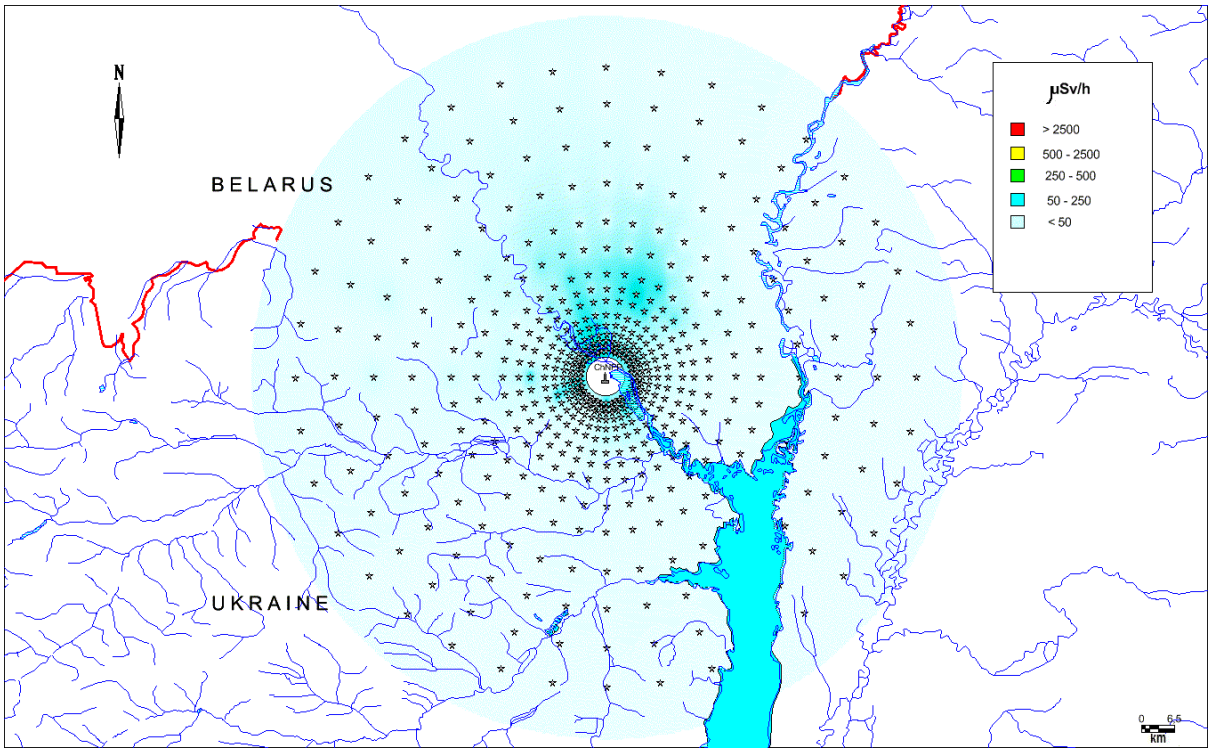
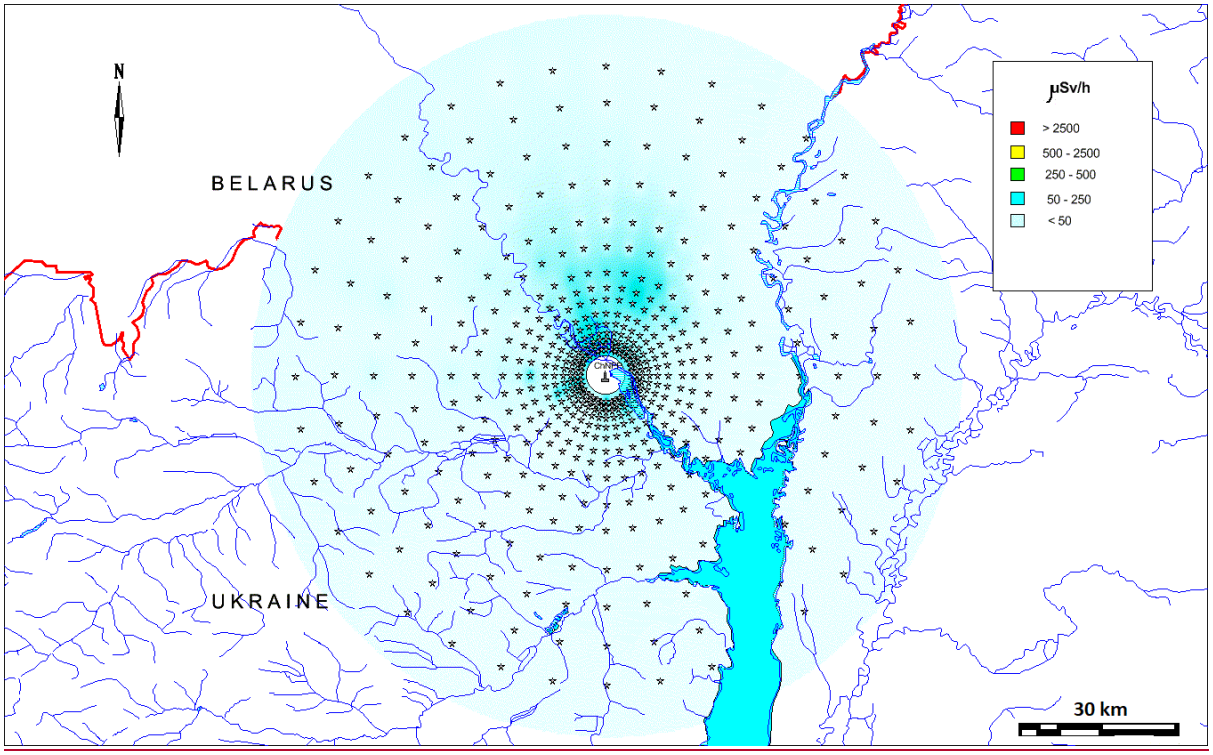
749 a



750



b

Figure 9. Spatial distribution, interpolated as for Figure 8, of effective dose rate within the 60 km zone around the ChNPP on 10th May 1986 (a) and 10th August 1986 (b). Note no data were available for less than 5 km from ChNPP and no interpolation for this area has been attempted.

~~Figure 9. Spatial distribution of effective dose rate within the 60km zone around the ChNPP on 10th May 1986 (a) and 10th August 1986 (b).~~

The estimated effective dose rate values exceed the evacuation dose criteria of 50 µSv h^{-1} over a large area (especially in the north and west) of the 60 km area around the ChNPP on 10th May 1986 (Figure 9a); as discussed above a dose rate of 50 µSv h^{-1} on 10th May 1986 equated to a total dose over the first year after the accident of 50 mSv - the value used to define areas for evacuation. On the 10th August 1986 the area estimated to exceed 50 µSv h^{-1} was restricted to the north (Figure 9b). The dose rate decreased quickly after the accident due to the radioactive decay of short-lived radionuclides. The dominance of these short-lived radionuclides and a lack of knowledge of the radionuclide composition of the fallout made it difficult in 1986 to estimate external dose rates to the public for an evaluation date of 10th May 1986 (most dose rate measurements being made after the 10th May). This likely resulted in the overestimation of dose rates for some villages in 1986 leading to their evacuation when the external dose rate would not have been in excess of the 50 mSv limit used by the authorities.

There is a need for deposition data for the CEZ and surrounding areas for a number of reasons. These include exploring risks associated with future management options for the CEZ (e.g. management of the water table, forest fire prevention, increased tourism, etc.) and also the return of abandoned areas outside of the CEZ to productive use. The long-term effect of radiation exposure on wildlife in the CEZ is an issue of much debate (e.g. see discussion in Beresford et al., 2019). Improved data which can be used to map the contamination of a range of radionuclides will be useful in improving dose assessments to wildlife (including

retrospective assessments of earlier exposure rates). The CEZ has been declared a 'Radioecological Observatory' (Muikku et al., 2018) (where a Radioecology Observatory is defined as a radioactively contaminated field site that provides a focus for joint, long-term, radioecological research). The open provision of data as described in this paper fosters the spirit of collaboration and openness required to make the observatory site concept successful and joins a growing amount of data made available for the CEZ (Kashparov et al., 2017; Fuller et al., 2018; Kendrick et al., 2018; Gaschak et al., 2018; Beresford et al., 2018; Lerebours and Smith, 2019).

4 Data availability

The data described here have a digital object identifier (doi: 10.5285/a408ac9d-763e-4f4c-ba72-73bc2d1f596d) and are freely available for registered users from the NERC Environmental Information Data Centre (http://eidc.ceh.ac.uk/) under the terms of the Open Government Licence (Kashparov et al., 2019).

Competing interests. The authors declare that they have no conflict of interest.

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Author contribution. Soil samples were collected by the USSR Ministry of Defence and delivered to UIAR. Sample preparation, analysis and data interpretation was carried out by UIAR staff contributing as follows: Kashparov, Levchuk, Protsak, - sample preparation, measurement of radionuclide activity concentrations in samples; Kashparov - analysis of results; Zhurba - database creation and preparation of the manuscript figures (maps). The manuscript was prepared by Chaplow, Beresford, Kashparov, Levchuk and Zhurba.

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Appendix 1. A detailed explanation of the column headings and units (where applicable) which accompanies the data (Kashparov et al., 2019).

Column_heading	Explanation	Units
Identifier	Unique identification number	not applicable
Angle_degree	A number between 10 and 360 indicates the direction from the ChNPP in degrees; 90 degrees is due east, 180 degrees is due south, 270 degrees is due west and 0/360 degrees is due north. See Figure 1.	degree
Distance_from_ChNPP_km	Distance from the Chernobyl Nuclear Power Plant (ChNPP) reactor number 4 in kilometres	kilometres
Date_gamma_measurement	Date of gamma measurement. An empty cell indicates a network point	dd-month-yyyy

	located in a water body where sample collection was not possible	
Exposure_dose_rate_mR/h	Dose rate in air at a height of 1 metre	milliroentgen per hour
Absorbed_dose_rate_microGray/h	Absorbed dose rate is the energy deposited in matter by ionizing radiation per unit mass	Micro Gray per hour
Zr-95_Bqm^2	Density of soil contamination with zirconium-95	Becquerel per square metre
Zr-95_relative_error	Relative uncertainty in determination of Zr-95 (at 68% confidence interval)	percentage
Nb-95_Bqm^2	Density of soil contamination with niobium-95	Becquerel per square metre
Nb-95_relative_error	Relative uncertainty in determination of Nb-95 (at 68% confidence interval)	percentage
Ru-106_Bqm^2	Density of soil contamination with ruthenium-106	Becquerel per square metre
Ru-106_relative_error	Relative uncertainty in determination of Ru-106 (at 68% confidence interval)	percentage
Cs-134_Bqm^2	Density of soil contamination with caesium-134	Becquerel per square metre
Cs-134_relative_error	Relative uncertainty in determination of Cs-134 (at 68% confidence interval)	percentage
Cs-137_Bqm^2	Density of soil contamination with caesium-137	Becquerel per square metre
Cs-137_relative error	Relative uncertainty in determination of Cs-137 (at 68% confidence interval)	percentage
Ce-144_Bqm^2	Density of soil contamination with cerium-144	Becquerel per square metre
Ce-144_relative_error	Relative uncertainty in determination of Ce-144 (at 68% confidence interval)	percentage

Exch_Cs-134+Cs-137_Bqm^2	Density of soil contamination with the exchangeable form of caesium	Becquerel per square metre
Note on empty cells	An empty cell means that data is not available	
Instrument	Gamma spectrometer with a semiconductor detector GEM-30185 ORTEC (results reported at 68% confidence level)	

Appendix 2. Decay radiation information from the master library, integrated in spectrum analysing software tool Gelicam (EG&G ORTEC, USA), used in gamma-analyses. Activities of ^{106}Ru and ^{137}Cs in samples were estimated via their gamma radiation emitting progenies ^{106}Rh and ^{137m}Ba, respectively

Target radionuclide	Measured radionuclide	Energy, keV	Emission probability %	Half life of target radionuclides
^{95}Zr	95Zr	724.20 756.72	44.10 54.50	64.02 days
^{95}Nb	95Nb	765.79	99.79	34.97 days
^{106}Ru	106Rh	621.84 1050.47	9.812 1.73	368.2 days
^{134}Cs	134Cs	604.70 795.85	97.56 85.44	753.1 days
^{137}Cs	137mBa	661.66	85.21	30.174 years
^{144}Ce	144Ce	133.54	10.8	284.3 days