



# Reconstructing the Chernobyl Nuclear Power Plant (CNPP) accident 30 years after. A unique database of air concentration and deposition measurements over Europe<sup>☆</sup>



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## ABSTRACT

30 years after the Chernobyl Nuclear Power Plant (CNPP) accident, its radioactive releases still remain of great interest mainly due to the long half-lives of many radionuclides emitted. Observations from the terrestrial environment, which hosts radionuclides for many years after initial deposition, are important for health and environmental assessments. Furthermore, such measurements are the basis for validation of atmospheric transport models and can be used for constraining the still not accurately known source terms. However, although the “Atlas of cesium deposition on Europe after the Chernobyl accident” (hereafter referred to as “Atlas”) has been published since 1998, less than 1% of the direct observations of <sup>137</sup>Cs deposition has been made publicly available. The remaining ones are neither accessible nor traceable to specific data providers and a large fraction of these data might have been lost entirely. The present paper is an effort to rescue some of the data collected over the years following the CNPP accident and make them publicly available. The database includes surface air activity concentrations and deposition observations for <sup>131</sup>I, <sup>134</sup>Cs and <sup>137</sup>Cs measured and provided by Former Soviet Union authorities the years that followed the accident. Using the same interpolation tool as the official authorities, we have reconstructed a deposition map of <sup>137</sup>Cs based on about 3% of the data used to create the Atlas map. The reconstructed deposition map is very similar to the official one, but it has the advantage that it is based exclusively on documented data sources, which are all made available within this publication. In contrast to the official map, our deposition map is therefore reproducible and all underlying data can be used also for other purposes. The efficacy of the database was proved using simulated activity concentrations and deposition of <sup>137</sup>Cs from a Langrangian and a Eulerian transport model.

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## 1. Introduction

The most serious nuclear disaster in human history took place in the nuclear complex of Chernobyl on April 26th, 1986 in the Former

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Soviet Union (FSU, nowadays near the northern border of Ukraine with Belarus) (IAEA, 2006; Yablokov et al., 2009). The accident was caused by a catastrophic power increase in the reactor unit 4 during a scheduled test procedure, leading to explosions and a nuclear fire, which resulted in dispersion of vast quantities of radioactive fuel and core materials into the atmosphere (Medvedev, 1990). Moreover, the combustible graphite moderator was ignited, increasing the emission of radioactive particles, which were transported by the rising smoke as the reactor was not covered by any kind of hard

containment vessel (Burakov et al., 1996).

Consequently, around 10 EBq ( $10^{19}$  Bq) of radioactivity were released and transported over long distances following the prevailing winds (De Cort et al., 1998). The most severe contamination occurred in FSU countries (Ukraine, Belarus and Russia). For instance, it has been estimated that around 47% of the aerosol-bound components deposited over Belarus (IAEA, 2006). According to conservative estimates, 350,000 emergency workers were initially involved in cleaning activities in 1986–1987. Among them, about 200,000 recovery workers (and 600,000 the following years), known as “liquidators”, took part in major mitigation activities in the 30-km zone surrounding the reactor. Following the definition given by the International Atomic Energy Agency (IAEA, 2005, 2009), any area with deposition density of more than 40 kBq m<sup>-2</sup> (for beta and gamma emitters) is classified as “contaminated”. Hence, around 6.8 million people lived in contaminated areas and of these about 270,000 people lived in “more contaminated” areas (>185 kBq m<sup>-2</sup>). Of this population, 135,000 people were evacuated from the area surrounding the CNPP (designated as the “Chernobyl Exclusion Zone – CEZ”) to non-contaminated areas. This gives a total population that directly suffered from the accident of more than 7 millions (Cardis et al., 1996; Fairlie and Sumner, 2006). Outside Ukraine, Belarus and Russia, several research groups and monitoring facilities across Europe began reporting high radioactivity levels following the disaster (e.g., in parts of Norway, Sweden and Finland, in Austria, Switzerland, North Italy, Slovenia, Poland, Romania and Greece) (JRC, 2015). Given the unprecedented magnitude of radioactive emissions, it was of great importance for the official authorities to record the exact radioactive contamination all over Europe, in order to develop and implement policies related to public health in the region.

Therefore, shortly after the accident, the REM (Radioactivity Environmental Monitoring) project started with the aim to improve the exchange of information between the European Union (EU) Member States (JRC, 2015). The main line of activities concerned the improvement of procedures for the collection, evaluation and harmonization of environmental radioactivity concentrations and the modeling of the migration of radioactivity in the environment for routine and emergency conditions, as well as making this information available to the general public. In parallel, several research groups worldwide started independently reporting observations of surface atmospheric concentrations and deposition taken for research purposes. According to Scopus (Scopus, 2015), more than 19,000 scientific publications contain the term “Chernobyl” in the title with an annual average of 475 since 1996 (maximum: 821 in 2014). This number is even higher when including non peer-reviewed publications and reports. Since the REM database was founded, there has not been any update in the public dataset concerning Chernobyl. This means that measurements from areas not belonging to the EU countries (such as the FSU countries) have not been included, albeit several observations have become available. In addition, such measurements existed (e.g., De Cort et al., 1998; Izrael et al., 1996) as they were used to create the Atlas deposition map but they are unavailable or have been lost.

More specifically, very few gridded observations from the areas near Chernobyl, such as Russia, Belarus and Ukraine, in which a significant amount of the fallout was deposited (47%), are available within the public domain. Such measurements are very important when assessing health consequences from the accident (both for human and non-human biota), for model validation, as well as for inverse modeling for the calculation of the Chernobyl source–term. In this paper, we present a unique database with emphasis on observations from Eastern Europe (Ukraine, Belarus, Russia). For the first time, we include observations of surface air activity

concentration of <sup>131</sup>I ( $t_{1/2} = 8.02$  d), <sup>134</sup>Cs ( $t_{1/2} = 2.01$  y) and <sup>137</sup>Cs ( $t_{1/2} = 30.2$  y) and deposition (<sup>134</sup>Cs and <sup>137</sup>Cs) from several high-radioactivity areas inside the CEZ, in the rest of Ukraine, in highly contaminated areas of Belarus (e.g., Gomel) and Russia, in a database implemented for research purposes. The data are direct measurements carried out by local authorities. The main goal is “data-rescue” considering that 30 years after Chernobyl no updates have been established into a single database. Furthermore, a gridded database resulted after interpolation of the original measurements now becomes freely available at a resolution of  $0.01^\circ \times 0.01^\circ$  ( $\approx 1$  km) in the CEZ ( $25^\circ\text{E}–40^\circ\text{E}$ ,  $47^\circ\text{N}–55^\circ\text{N}$ ) and at  $0.1^\circ \times 0.1^\circ$  ( $\approx 10$  km) in the rest of Europe for <sup>137</sup>Cs and at  $0.5^\circ \times 0.5^\circ$  ( $\approx 50$  km) for <sup>134</sup>Cs. The database may be used for any kind of research purposes (e.g., model validation or health assessment for humans and biota etc.). Finally, researchers needing the original observations for their work may request direct access to the point observation data, which is currently under restricted access.

## 2. Sample origin and analytical methodology

### 2.1. Atmospheric measurements

The database consists of surface air activity concentration measurements (in Bq m<sup>-3</sup>) of <sup>134</sup>Cs (1927 observations), <sup>137</sup>Cs (1601) and <sup>131</sup>I (2041) and deposition density observations (in kBq m<sup>-2</sup>) of <sup>134</sup>Cs (2966) and <sup>137</sup>Cs (11,334). The most important contribution of the present work is the updated database of deposition observations. As mentioned above, air concentrations could be only detected until 2 months after the accident, hence the database of air activity concentrations could not be further updated, except from observations based on our literature search (Supplementary Data).

### 2.2. Deposition measurements

On the contrary, the final receptors of the aerosol components are the terrestrial, freshwater and marine ecosystems since the aerosols are removed from the atmosphere by wet and dry deposition (Seinfeld and Pandis, 1998). Once radionuclides are deposited onto the soil, their surface concentrations decrease due to radioactive decay, vertical migration and washout to any kind of reservoir in an environmental cycle. The whole process is characterized by the so-called effective half-life, which combines physical and ecological half-lives of <sup>137</sup>Cs. The ecological half-life of <sup>137</sup>Cs is defined as the period of time it takes for <sup>137</sup>Cs to decrease by half, affected by processes others than its radioactive decay (e.g., vertical migration, runoff etc ...). It is directly linked to the effective half-life, which also includes radioactive decay of <sup>137</sup>Cs (see also, Evangeliou et al., 2013). For terrestrial <sup>137</sup>Cs, the effective half-life can be highly variable being rapid in erosional places and places where decontamination practices, such as stripping soil surface, have been conducted or up to a half-life in undisturbed places (Bergan, 2000; Evangeliou et al., 2015). This means that deposition measurements taken the following years could be incorporated into the dataset. The associated uncertainty strongly depends on how many years after the actual accident the samples were collected and analyzed and how prone to migration or erosion soils can be.

Nevertheless, in the terrestrial environment of Chernobyl, ecological loss of radionuclides through vertical migration and/or runoff are very slow and most of the radionuclides remain in the top 10–20 cm of the soil (Kashparov, personal communications; Ivanov et al., 1996). For this reason, all the corrections were made using the radioactive decay half-life of <sup>137</sup>Cs. The database only includes deposition measurements from the terrestrial ecosystem,

because the last constitutes a more stable environment, whereas radionuclides in the ocean, for instance, are quickly dispersed further (Buesseler et al., 2011). From the 11,334 deposition observations for  $^{137}\text{Cs}$ , 4077 were adopted from the public REM dataset, 5280 were made available from Talerko (2005a, 2005b) and 1324 from Kashparov et al. (2001, 2003). The remainder (509) were adopted from numerous publications included in the Supplementary Data.

The data of  $^{137}\text{Cs}$  deposition over the FSU countries were collected using the standardized method adopted previously in the former USSR (Tsaturov et al., 1996). The samples were collected within national framework programs for the determination of radioactive deposition in settlements. Specifically, samples were collected at a depth of not less than 10 cm and without any disturbance to avoid mixing of soil layers. The samples were then measured with gamma-spectrometry. The total number of soil samples for every settlement depended on its size and varied from 2 to 3 for small villages to several hundreds for large towns or towns that were highly contaminated. The obtained data were included into the database used for the creation of the Atlas. However, as stated before, they are not included in the public REM database.

Sampling and analysis in the rest of the European countries followed the standard procedures of the IAEA (International Atomic Energy Agency) (e.g., De Cort et al., 1998). Air concentrations were estimated using airborne gamma spectrometers mounted on aircraft or helicopters capable of flying at low altitudes (25–100 m) during the initial period after the accident. In countries where concentrations were lower, surface air was sucked through filters using high-volume samplers for a long time (e.g., hours to days depending on the relevant detection limits and the air concentrations). Then the filters were measured by gamma spectrometry using High Purity Germanium Detectors or High Volume Scintillation Detectors (e.g., NaI(Tl)). As regards to the levels of deposition, they are based on (i) *in situ* measurements of the ground by gamma spectrometry and (ii) sampling and measurements of surface soil samples. The latter has been determined by following the respective guidelines of the IAEA (IAEA, 1989). According to the guidelines, stones, and biological residues are removed from the soil sample and the weight of the remainder is recorded. Then the sample is homogenized and dried ( $\sim 100^\circ\text{C}$ ) until a constant weight is reached. The sample is then sieved through a 2-mm sieve, and it is finally transferred to the optimal counting geometry for gamma spectrometry measurements.

Given that each research group has used a different measurement technique (different detection systems), it is evident that the uncertainty of the results is different for each sample. For example, laboratory analyses of surface soil samples, in general, characterize the deposition at a location better than what is possible with *in situ* gamma spectrometry. On the other hand, *in situ* gamma spectrometry provides a rapid and spatially representative result, but it is sensitive to the distribution of activity in the environment.

### 3. Inverse Distance Weighting (IDW) method

The deposition data (both for  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) have been compiled to a basic map that is compared to the map presented in the Atlas (De Cort et al., 1998). For this reason, the collected observations have been corrected for decay to May 1st, 1986 (similar to the Atlas) and then interpolated onto a grid of  $0.01^\circ \times 0.01^\circ$  ( $\approx 1$  km) inside the CEZ and of  $0.1^\circ \times 0.1^\circ$  ( $\approx 50$  km) in the rest of Europe for  $^{137}\text{Cs}$  and onto a grid of  $0.5^\circ \times 0.5^\circ$  ( $\approx 50$  km) for  $^{134}\text{Cs}$  using a modified Inverse Distance Weighting (IDW) method described by Renka (1988). The different grids for  $^{137}\text{Cs}$  were chosen because of the high density of the measurements in the CEZ

(45% of the observations are located there). The interpolation method was chosen in order to be able to compare the obtained map directly with the map presented in the Atlas for the quality assessment of the presented dataset. This method is preferred due to its ease of use and to its high quality of interpolation.

The IDW interpolation is defined by:

$$\hat{v}(x, y) = \frac{\sum_{i=1}^n w_i v_i}{\sum_{i=1}^n w_i}$$

where  $\hat{v}(x, y)$  is the interpolated value at point  $(x, y)$ ,  $w_1, \dots, w_n$  are the relative weights and  $v_1, \dots, v_n$  are the observation values. The weights are defined by the inverse distance functions:

$$w_i = \left( \frac{r_w - d_i}{r_w d_i} \right)^2 \quad \text{for } (r_w - d_i) = \begin{cases} r_w - d_i & \text{if } d_k < r_w, \\ 0 & \text{if } d_k \geq r_w. \end{cases}$$

where  $r_w$  denotes the radius of influence about the point  $(x_i, y_i)$  and  $d_i$  the Euclidean distance between point  $(x, y)$  and  $(x_i, y_i)$  and  $d_k$  is the threshold distance. For  $^{134}\text{Cs}$  we used a threshold distance ( $d_k$ ) of 100 km all over Europe, while for  $^{137}\text{Cs}$  the threshold distance ( $d_k$ ) was defined as 10 km close to the CEZ ( $25^\circ\text{E}$ – $40^\circ\text{E}$ ,  $47^\circ\text{N}$ – $55^\circ\text{N}$ ) and 60 km in the rest of Europe. Again, a different threshold distance of  $^{137}\text{Cs}$  observations was selected due to the high density of measurements in the CEZ. The Euclidean distance is calculated using Vincenty's formulae (Vincenty, 1975).

## 4. Results

### 4.1. Data records

The Chernobyl database presented in the paper is archived at the Norwegian Institute for Air Research's (NILU) Data Center website <http://radio.nilu.no>. NILU has a long history in hosting large databases, as it maintains aerosol and gas long-term monitoring data from stations all over the world that are continuously updated (<http://ebas.nilu.no>). The original database contains five different records; three for air activity concentrations of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  formatted as below:

- 1) SITE: station code;
- 2) LON: longitude of sampling station;
- 3) LAT: latitude of sampling station;
- 4) STARTDATETIME: beginning date and time of sampling in a format year-month-day hour:minuteZ;
- 5) ENDDATETIME: ending date and time of sampling under the same format;
- 6) VALUE: surface air activity concentrations (in Cs134\_con, Cs137\_con and I131\_con) or deposition density (in Cs134\_dep and Cs137\_dep);
- 7) UNIT: the respective unit ( $\text{Bq m}^{-3}$  for air concentration,  $\text{Bq m}^{-2}$  for deposition).

It also contains two records for the total deposition of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  under the same format. For each record, there is one self-describing ascii-file that includes extensive metadata and describes the measurements (Cs134\_con, Cs137\_con and I131\_con for air activity concentration data and Cs134\_dep and Cs137\_dep for deposition density).

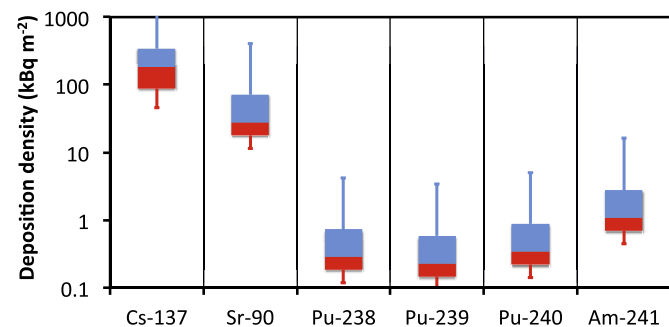
Due to restrictions imposed by data providers, the original observations are currently under restricted access but may be available upon request (details can be found in the relevant website). On the other hand, the gridded database of deposition of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  resulted after IDW interpolation of the original database is freely available, it can be easily and directly accessed and it may be

used for scientific studies. The format is LON, LAT, VALUE (Cs134 or Cs137 followed by the unit) and the name of the ascii (American Standard Code for Information Interchange) files starts with “gridded\_” followed by the name as before (e.g., Cs134\_dep) and ends with the resolution (e.g.,  $0.01 \times 0.01$ ).

The database also includes deposition observations of  $^{137}\text{Cs}$  ( $t_{1/2} = 30.2$  y),  $^{90}\text{Sr}$  ( $t_{1/2} = 28.8$  y),  $^{238}\text{Pu}$  ( $t_{1/2} = 87.7$  y),  $^{239}\text{Pu}$  ( $t_{1/2} = 24,100$  y),  $^{240}\text{Pu}$  ( $t_{1/2} = 6563$  y) and  $^{241}\text{Am}$  ( $t_{1/2} = 432.6$  y) from the CEZ from year 2015. The data ( $N = 48,781$ ) were made available to us from the Ukrainian authorities, in order to study the fires in contaminated forests of Chernobyl in April and August 2015. The data can be freely obtained from the website <http://radio.nilu.no> upon request. The format is identical to the aforementioned deposition observations of the Chernobyl database. The data cover a small area ( $29.2^\circ$ – $30.0^\circ\text{E}$  and  $51.2^\circ$ – $51.6^\circ\text{N}$ ) that suffered from the fires in 2015. They were not included in the original Chernobyl database because (a) they correspond to the year 2015, and (b) the density of the observations in the area is already high enough that additional measurements would not give more information about the deposition of  $^{137}\text{Cs}$ . The deposition levels of the six radionuclides in the area are depicted in Fig. 1. As expected, the highest values were obtained for  $^{137}\text{Cs}$ , which ranged from 3.3 to 24,044  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $482 \pm 1021$ , median: 184), whereas  $^{90}\text{Sr}$  values were found between 1.4 and 13,446  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $144 \pm 446$ , median: 27.5). Transuranium elements were found to be much lower, varying within 0.01–138  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $1.5 \pm 4.6$ , median: 0.28) for  $^{238}\text{Pu}$ , 0.01–111  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $1.2 \pm 3.7$ , median: 0.22) for  $^{239}\text{Pu}$ , 0.02–166  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $1.8 \pm 5.5$ , median: 0.33) for  $^{240}\text{Pu}$  and 0.06–527  $\text{kBq m}^{-2}$  (average  $\pm$  SD:  $5.6 \pm 17.5$ , median: 1.1) for  $^{241}\text{Am}$ .

#### 4.2. Technical validation

Observations of surface air activity concentrations of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are only available from the largest European cities within the database. This is primarily a result of the relatively poor monitoring before and during the first days of the accident in April 1986. Considering that atmospheric activity concentrations of aerosol-bound radioactive substances were the largest during the very first few days after the CNPP accident (Asmolov et al., 1988; Drozdovitch et al., 2013; Horvna and Wilhelmova, 1988; Lepel et al., 1988; Raes et al., 1988), no further improvement of these datasets could be made during the years after the accident, as the aerosol-bound radiocesium is removed from the atmosphere by wet and dry deposition, and  $^{131}\text{I}$ , in addition, has a very short



**Fig. 1.** Box & Whisker plots of deposition measurements of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Am}$  from samples collected and analyzed in an area between  $29.2^\circ$  and  $30.0^\circ\text{E}$  and  $51.2^\circ$ – $51.6^\circ\text{N}$  in 2015. The plot shows the minimum value, the 25th percentile (in red), the median, the 75th percentile (in blue) and the maximum. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

radioactive half-life ( $t_{1/2} = 8.01$  d) and is rapidly lost via radioactive decay.

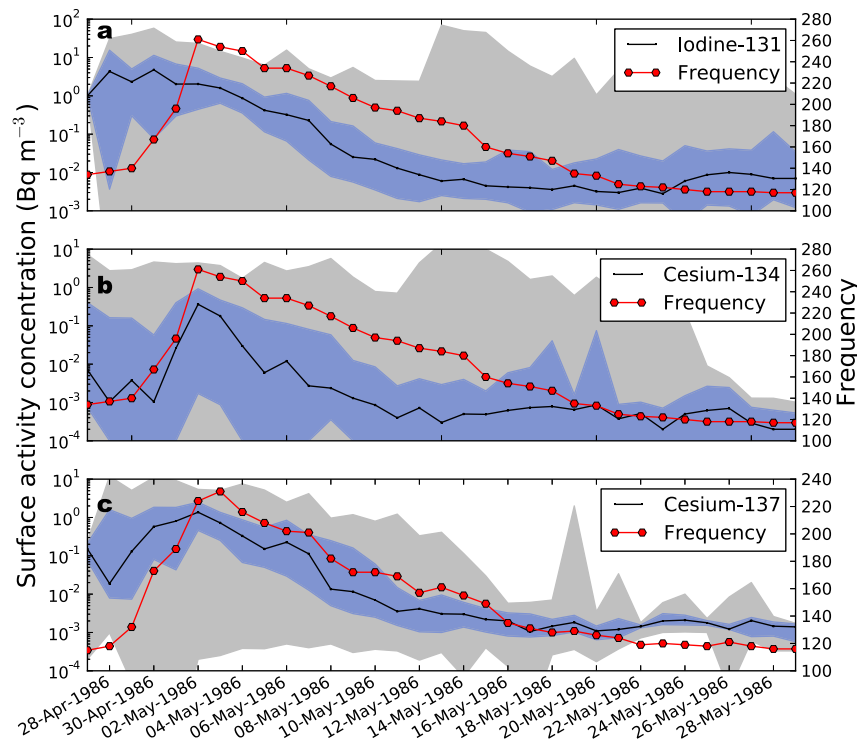
Fig. 2(a), (b) and (c) depict the time-series of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  activity concentrations from stations all over Europe as Box & Whisker plots. It appears that  $^{131}\text{I}$  showed up to one order of magnitude higher surface concentrations than radiocesium. This is because the core inventory of  $^{131}\text{I}$  was up to 11 times higher than that of  $^{137}\text{Cs}$  (De Cort et al., 1998), which in turn resulted in total emissions of  $^{131}\text{I}$  after the CNPP accident of about 6–20 times larger than those of  $^{137}\text{Cs}$  (Abagyan et al., 1986a; Borzilov and Klepikova, 1993; Haas et al., 1990; Izrael et al., 1990; Talerko, 2005a; UNSCEAR, 2000). Iodine-131 ( $^{131}\text{I}$ ) is important for thyroid cancer. Apart from Belarus, Ukraine and Russia, Austria had the second highest average  $^{131}\text{I}$  deposition density. It is noticeable  $^{131}\text{I}$  dispersion differs considerably from  $^{137}\text{Cs}$ . The (presumably very small) particles containing iodine in various forms would have been suspended in aerosols and spread throughout Europe. Radioiodines would also have been distributed as iodine gas,  $\text{I}_2$ . UNSCEAR (2008) reported that the main  $^{131}\text{I}$  uptake was ingestion via the grass pasture-cow-milk pathway. Presumably some radioiodine would also have been inhaled, as aerosol and gas and a smaller amount would have been absorbed through skin (Fairlie, 2016).

As regards to radiocesium, the highest concentrations over Europe were found in the beginning of May 1986 (2–3 May 1986), when the measurement frequency was also highest (Fig. 2(b) and (c)). Between April 26 and May 6, 1986, Chernobyl's fallout was very widely dispersed over Europe and the northern hemisphere (Brandt et al., 2002; Evangeliou et al., 2013 and others). For example, Chernobyl fallout concentrations were measured in Japan, over 8000 km from Chernobyl (e.g., Aoyama et al., 1991). The largest concentrations of radiocesium (as well as of volatile nuclides and fuel particles) occurred in Belarus, Russia and Ukraine. However, more than half of the total quantity of Chernobyl's volatile inventory was deposited outside these countries. Russia, Belarus and Ukraine received the highest amounts of fallout while former Yugoslavia, Finland, Sweden, Bulgaria, Norway, Rumania, Germany, Austria and Poland each received more than 1 PBq of  $^{137}\text{Cs}$ .

Fig. 3(a) depicts the sampling points where deposition measurements of  $^{137}\text{Cs}$  are available in the public REM database from the CNPP accident (JRC, 2015), in contrast to our updated database (lower panel). In addition to  $^{137}\text{Cs}$  deposition observations, the database also contains deposition measurements of  $^{134}\text{Cs}$ . However, due to its short-lived nature ( $t_{1/2} = 2.01$  y),  $^{134}\text{Cs}$  levels declined significantly the years following the CNPP accident and presently no meaningful measurements can be made.

Deposition data are available in the form of discrete (point) measurements. These data can be part of a regular sampling (e.g., points along a terrestrial sampling campaign) or of a random survey (values from research stations, weather stations, etc ...). Such data may be presented in different ways. For instance, presenting data point-by-point colored according to the measured levels of deposition is indicative of how thoroughly an area has been investigated (e.g., Fig. 4(a) for  $^{134}\text{Cs}$ ). The deposition of  $^{134}\text{Cs}$  was interpolated onto a  $0.5^\circ \times 0.5^\circ$  ( $\approx 50$  km) grid applying a threshold distance between observations of 100 km. Cesium-134 was more intensely deposited close to the CNPP (Ukraine and Belarus), in Romania, Austria and the Nordic countries (Norway, Sweden and Finland), as well as in parts of Italy, France, Germany and Greece. As expected, deposition is high close to the CNPP, in parts of Romania and in Scandinavian countries, where a significant amount of the radioactive fallout was deposited (Evangeliou et al., 2013).

Fig. 5(a) depicts the deposition of  $^{137}\text{Cs}$  from the Atlas map (De Cort et al., 1998), and after the IDW interpolation of the 11,334 observations presented in NILU's repository (lower panel). The data have been interpolated onto a grid of  $0.01^\circ \times 0.01^\circ$  ( $\approx 1$  km) inside



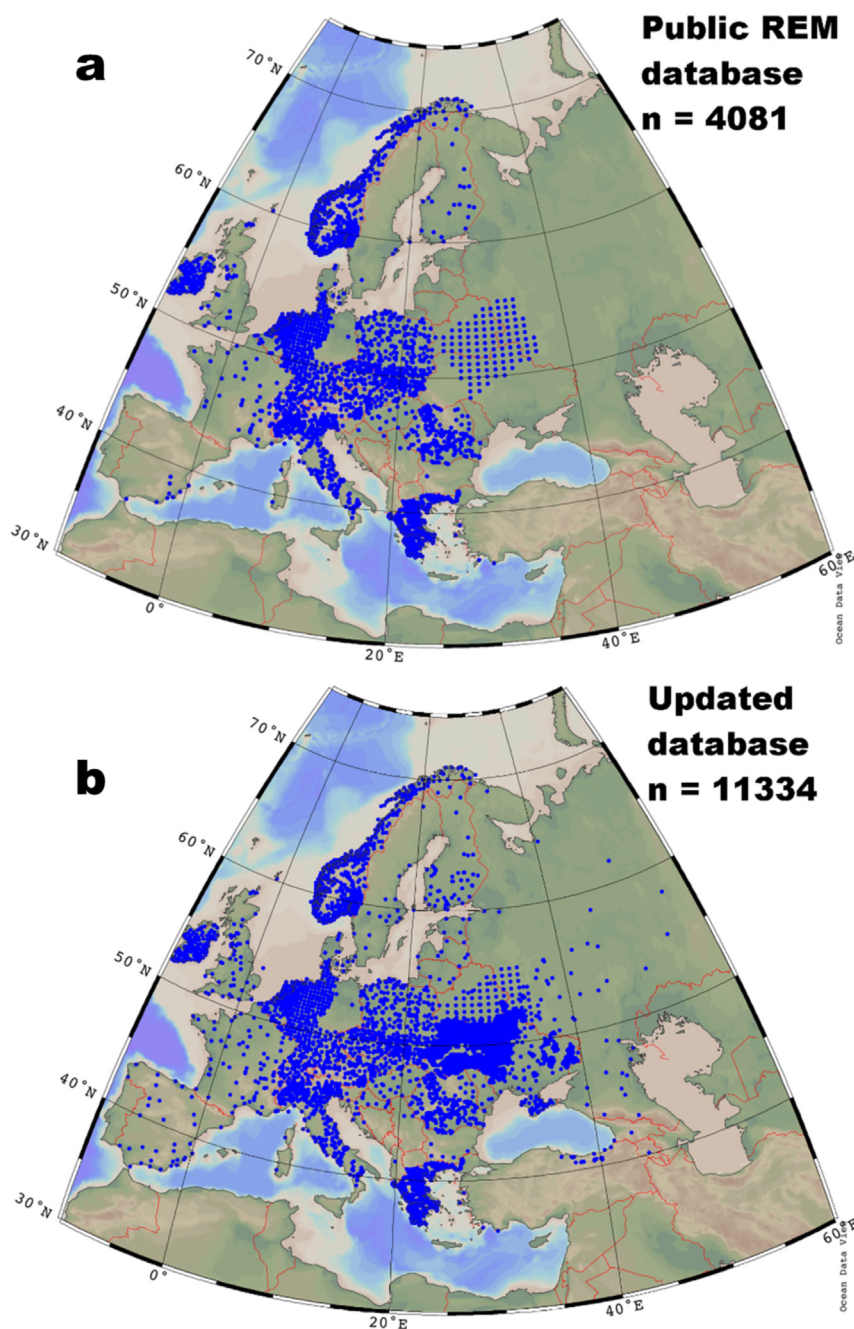
**Fig. 2.** Time-series of surface air activity concentrations of (a) <sup>131</sup>I, (b) <sup>134</sup>Cs and (c) <sup>137</sup>Cs all over Europe following the Chernobyl Nuclear Power Plant (CNPP) accident. The series of observations are given in shaded areas between minimum and maximum (grey) and between 25th and 75th percentile (blue) with the median plotted as a black line due to the high variability of the concentrations close and far from the source (CNPP). The frequency of the observations is displayed on the secondary axis (red line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the CEZ (25°E–40°E, 47°N–55°N) and  $0.1^\circ \times 0.1^\circ$  ( $\approx 10$  km) in the rest of Europe applying a threshold distance between observations of 10 km inside the CEZ and 60 km in the rest of Europe. The same scale and palette as in the Atlas have been kept. The data with the highest deposition densities were collected from areas within the CEZ, in East Belarus (Gomel) and East Russia (Bryansk Oblast), although contaminated regions ( $>40$  kBq m<sup>-2</sup>) were also observed in Scandinavia, in Central Europe (North Italy, Austria, Slovenia) and Greece. All data have been corrected for radioactive decay to May 1st, 1986 similar to the data presented in the Atlas. However, one problem of this type of data presentation is that areas with low data density (e.g., in North and Western Russia) cannot be mapped (shown as grey areas in Figs. 4 and 5(b)), despite the possibility of contamination. In the present map (Fig. 5(b)), <sup>137</sup>Cs deposition levels represent total deposition, irrespective of the source (Chernobyl accident, nuclear weapon testing or any other type of release). Given that the reported values of <sup>137</sup>Cs deposition over Europe from nuclear weapon tests have been reported to be up to 3–5 kBq m<sup>-2</sup> at maximum in Central Europe (De Cort et al., 1998) and several years have passed until our reference date (May 1st, 1986), a relatively small contribution from the nuclear tests can be assumed. However, the deposition level corresponding to 0–2 kBq m<sup>-2</sup> is typical of the deposition values expected from the weapon tests and may be used, in a simplistic way, to divide areas unaffected from areas affected by the Chernobyl accident. Considering that for the total <sup>137</sup>Cs deposition map in the Atlas (De Cort et al., 1998) it was claimed that a total number of 399,533 observations (most of which are unavailable to the public and many may have been lost entirely) were processed against 11,334 here, the map produced from our <sup>137</sup>Cs deposition database seems very similar.

More specifically, high and low deposition values of <sup>137</sup>Cs are

well captured. For instance, the deposition levels in Scandinavia, where contaminated areas have been confirmed to occur between Gladheim and Dombas villages (area 1), as well as near Nordli, Ankarede, Fatmomack and Arksjo (area 2) in Norway, along the coasts of Sweden in the Gulf of Bothnia (near Örnköldsvik, area 3) and in the southern part of Finland (near Tampere city, area 4), are all captured. Moreover, deposition levels in areas far from Ukraine, such as in Scotland (area 5), where they have been reported to be between 10 and 40 kBq m<sup>-2</sup>, in Central Italy (area 6) and in Northwestern Greece (10–40 kBq m<sup>-2</sup>, area 7) are reproduced in our map as well. Another good example of the efficiency of the present database can be seen in Central Europe, where deposition patterns of 10–40 kBq m<sup>-2</sup> are well-captured in Switzerland (area 8), Austria (area 9), Slovenia (area 10), in the borders of ex-Czechoslovakia with Poland (area 11) together with a contaminated area in Austria (deposition  $>40$  kBq m<sup>-2</sup>, area 9).

Near the CNPP, the map has been generated with  $0.01^\circ \times 0.01^\circ$  grid resolution ( $\approx 1$  km) due to the high frequency of the observations, as around 45% of the observations used to construct Fig. 5(b) were from this area. The deposition levels of <sup>137</sup>Cs in the CEZ (area 12), Belarus (mostly near Gomel and Vetka, area 13) and Russia (in Bryansk Oblast and nearby areas, area 14), as well as in areas of Southern Ukraine (area 15) are well-reproduced, despite that some areas of reported deposition above 40 kBq m<sup>-2</sup> are missing (e.g., in Russia – east of area 14) due to the lack of available observations. In addition, reported high levels of <sup>137</sup>Cs in the northern and southern borders of Romania are well represented in the map (area 16). Our deposition finds some higher values in Southern Italy (area 18), but it agrees with the Atlas deposition showing some high values in Northeastern Russia (area 17). These values are about an order of magnitude higher than in the Atlas map mostly covering areas, where observations are very sparse

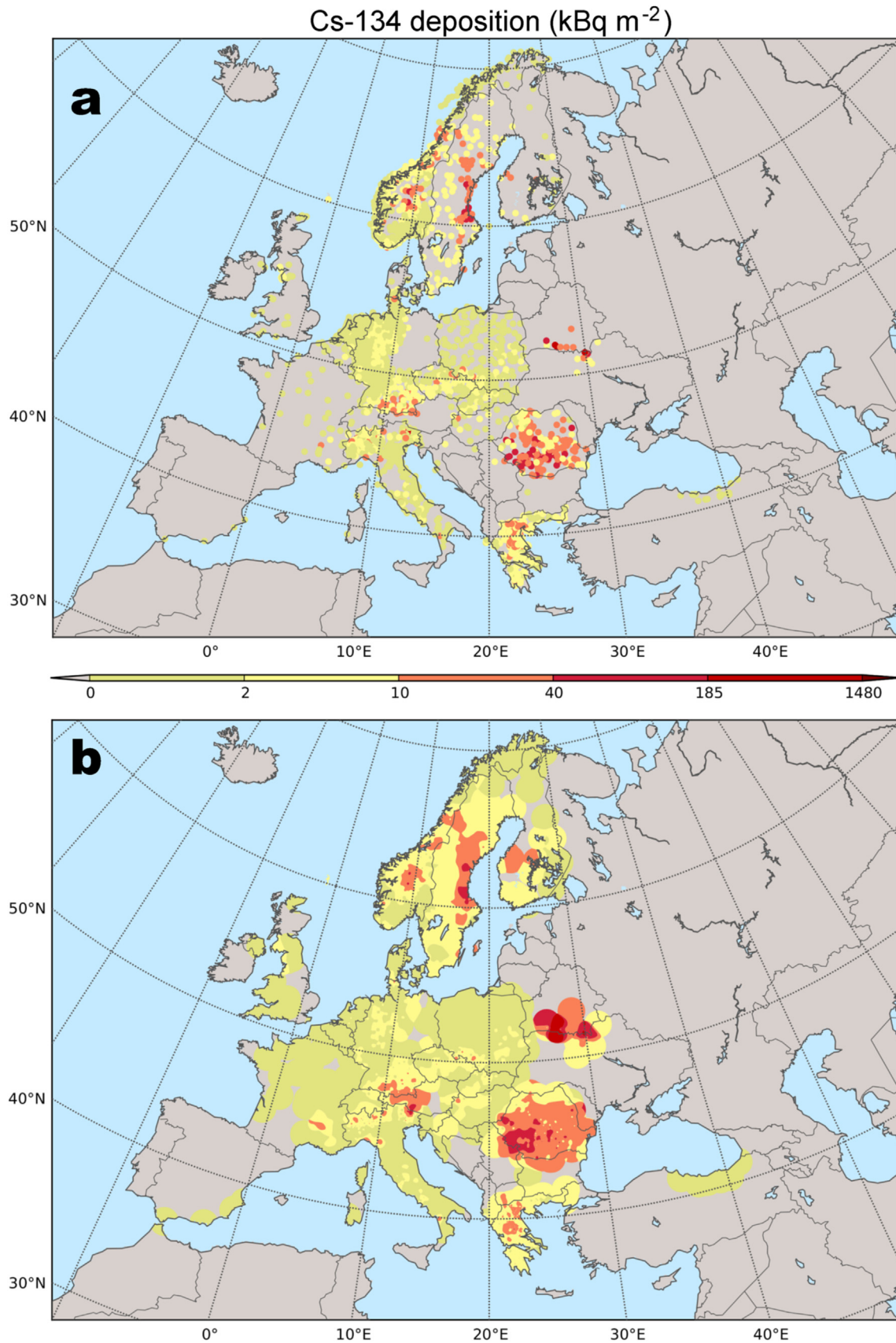


**Fig. 3.** (a) Sampling points of deposition observations of  $^{137}\text{Cs}$  all over Europe from the public Radioactivity Environmental Monitoring database of the Joint Research Center. It is noteworthy that the database lacks observations from the Former Soviet Union countries where the actual deposition of the radioactive fallout took place following the initial releases of Chernobyl. (b) The updated database, which now covers all the highly contaminated areas in Ukraine, Belarus and Russia.

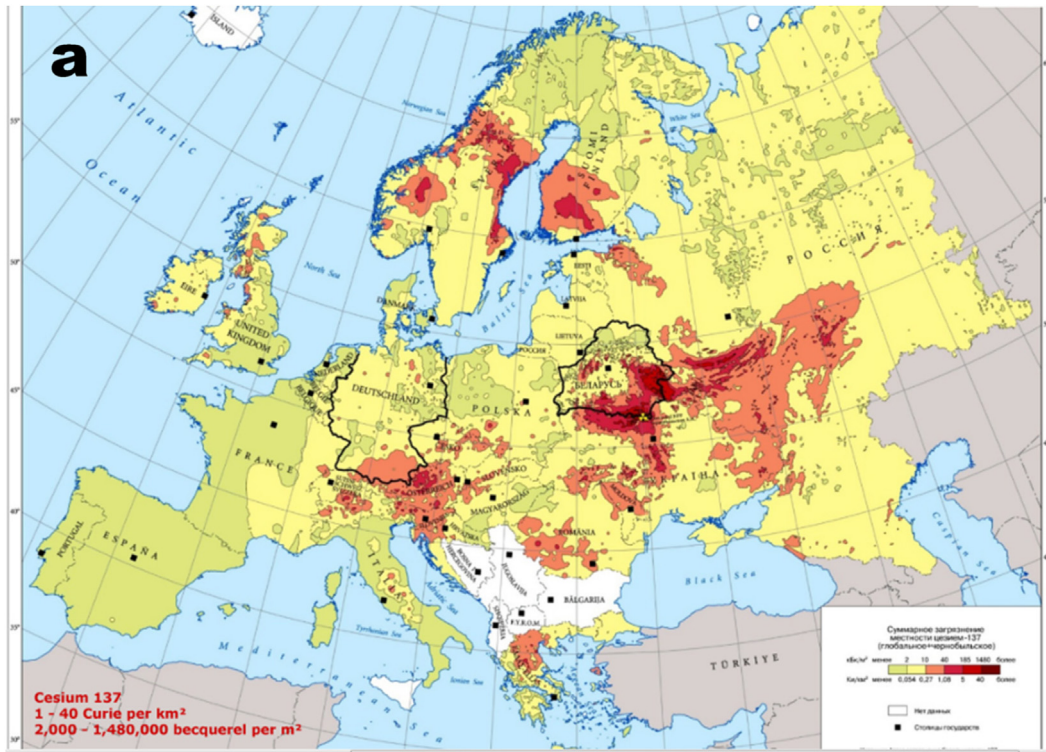
(e.g., Fig. 3(b) all values east of  $40^\circ\text{E}$ ). For these reasons, they should be interpreted carefully. Finally, the present deposition from our database defines a contamination spot at the eastern part of the Black Sea (area 19), for which data do not exist in the Atlas map. Nevertheless, the area has been confirmed as contaminated by the Chernobyl fallout showing values of up to  $150\text{ kBq m}^{-2}$  (Kose et al., 1994; Varinlioglu et al., 1994). After excluding sparse values from the database (all values east of  $40^\circ\text{E}$ ), the deposition of  $^{137}\text{Cs}$  deposited over Europe is estimated to be around 23 kg equivalent to 75 PBq, which is in the same order as what has been reported until now (Abagyan et al., 1986b; Talerko, 2005b: 39 PBq, Brandt et al., 2002 and Persson et al., 1987: 85 PBq, Izrael et al., 1990:

75 PBq). On the other hand,  $^{134}\text{Cs}$  observations are relatively sparse close to CNPP, allowing only a lower estimate of the total deposition over Europe (27 PBq, versus 54 PBq reported by Brandt et al., 2002; Persson et al., 1987).

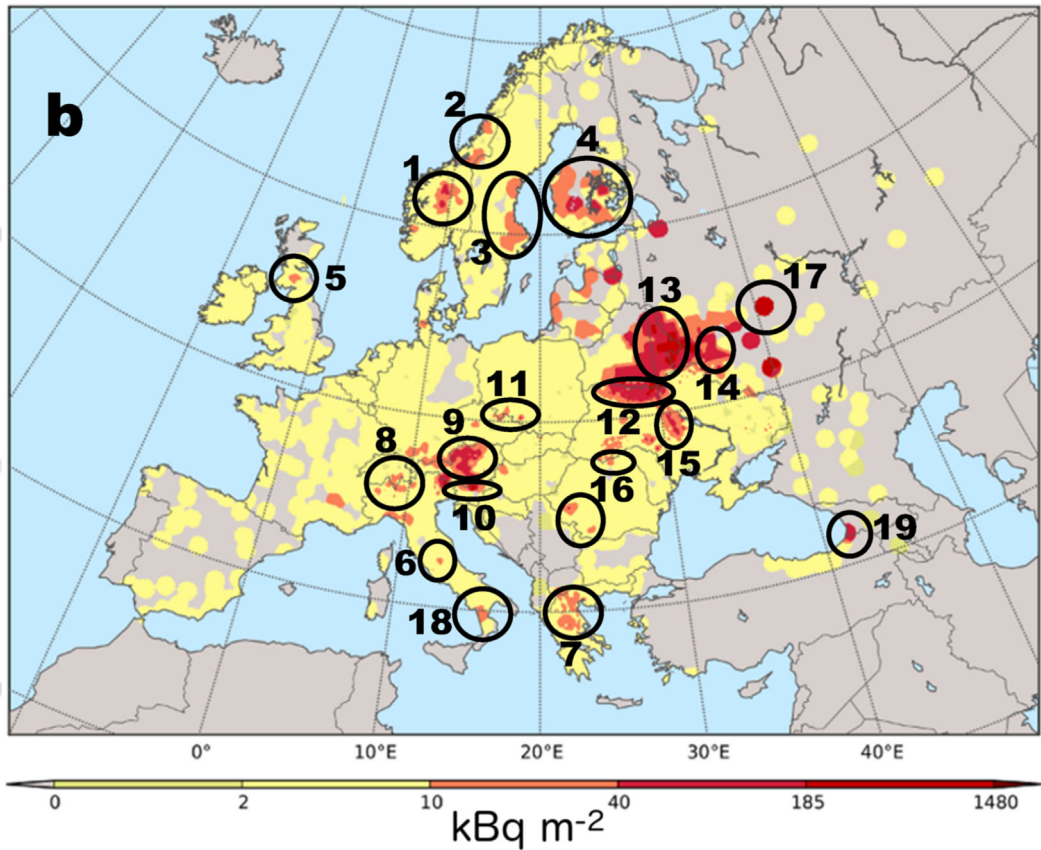
Another way to validate the effectiveness of the present database is to estimate country-by-country total deposition of  $^{137}\text{Cs}$  and compare it with the same calculation made for the Atlas. For this reason, we have masked every European country that provided measurements, specifically Austria (AT), Belarus (BY), Belgium (BE), Croatia (HR), Czech Republic (CZ), Denmark (DK), Estonia (EE), Finland (FI), France (FR), Germany (DE), Greece (GR), Hungary (HU), Ireland (IE), Italy (IT), Latvia (LV), Lithuania (LT), Luxembourg (LU),



**Fig. 4.** (a) Sampling stations and levels of <sup>134</sup>Cs deposition over Europe before and (b) after Inverse Distance Weighting interpolation onto a 0.5° × 0.5° (≈ 50 km) grid applying a threshold distance between observations of 100 km. Although <sup>134</sup>Cs is mapped for the first time and major contaminated areas are captured, it appears that the coverage is relatively poor in Europe.

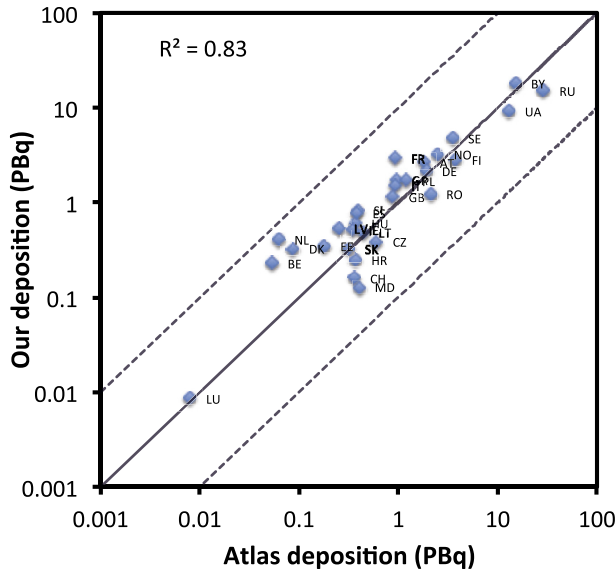


Cs-137 deposition (IDW d=60km)



**Fig. 5.** (a) Deposition of <sup>137</sup>Cs all over Europe from the Atlas (De Cort et al., 1998) based on 399,533 measurements adopted from the IRS Belrad (2015). (b) Deposition of <sup>137</sup>Cs all over Europe based on 11,334 measurements presented in our database. The data were interpolated onto a grid of 0.01° × 0.01° (≈ 1 km) close to the Chernobyl Exclusion Zone (CEZ) (25°E–40°E, 47°N–55°N) and of 0.1° × 0.1° (≈ 10 km) in the rest of Europe applying a threshold distance between observations of 10 km close to the CEZ and 60 km in the rest of Europe. Grey areas depict missing values.





**Fig. 6.** Country-by-country deposition of  $^{137}\text{Cs}$  in the Atlas versus in the database presented here. In most cases, the data are close to 1 by 1 line showing a correlation coefficient of 0.8.

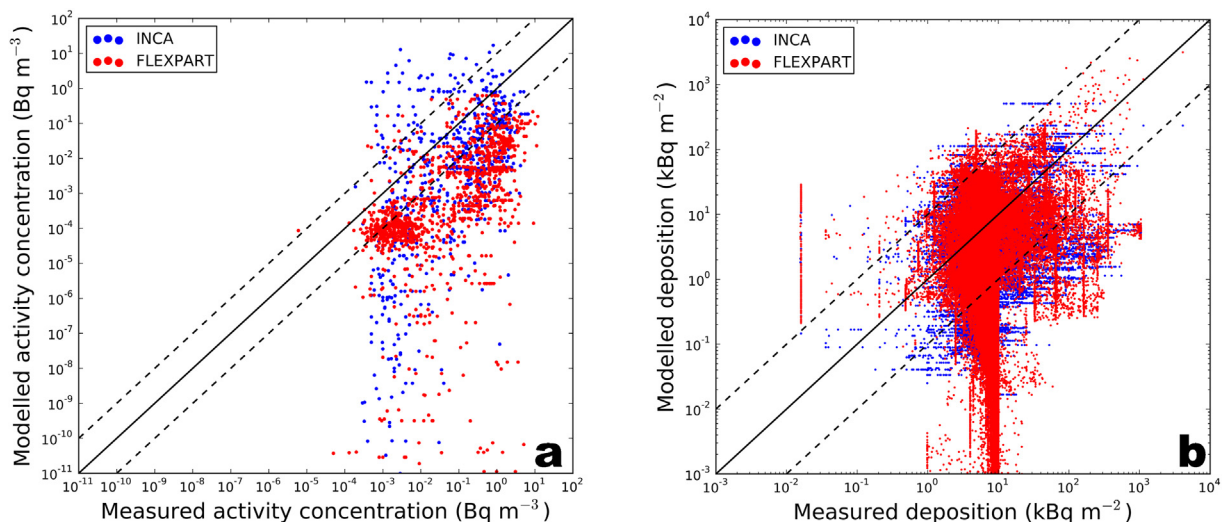
Moldavia (MD), Netherlands (NL), Norway (NO), Poland (PL), Rumania (RU), Russia (RU, European part), Slovak Republic (SK), Slovenia (SL), Spain (ES), Sweden (SE), Switzerland (CH), Ukraine (UA) and United Kingdom (GB). The results are shown in Fig. 6 as a scatter plot of  $^{137}\text{Cs}$  total deposition in any country included in the Atlas versus in the dataset presented here and the respective countries are labeled accordingly. In most cases, the country-by-country total depositions of  $^{137}\text{Cs}$  were very close to what was reported in the Atlas, based on more than 399,533 measurements, showing a very large correlation coefficient ( $R^2 = 0.83$ ). Over-estimations of our dataset were found in Netherlands (0.41 versus 0.062 PBq in the Atlas), Belgium (0.23 versus 0.053 PBq in the Atlas), Denmark (0.33 versus 0.087 PBq in the Atlas) and France (2.9 versus 0.93 PBq in the Atlas). Our database mainly underestimates the Atlas deposition in the Czech Republic (0.39 versus 0.6 PBq in the Atlas) and in Moldavia (0.13 versus 0.4 PBq in the Atlas), as well as in Russia (17.2 versus 29 PBq in the Atlas) due to the lack of

available observation in most of its territory. In total, we find that 75.02 PBq of  $^{137}\text{Cs}$  were deposited over Europe relative to 85 PBq reported in the Atlas (De Cort et al., 1998).

A large part of the present database was used to validate the Eulerian atmospheric transport model LMDZORINCA (INCA) at a resolution  $0.45^\circ \times 0.51^\circ$  over 19 vertical levels (see also: Evangeliou et al., 2013) and the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005) at a resolution of  $0.1^\circ \times 0.1^\circ$  over 10 vertical levels (Fig. 7(a) and (b)). The comparison is presented here for the first time. Both models managed to predict the radioactive contamination in most of the regions similar to the Atlas showing high correlations for atmospheric activity concentrations, cumulative deposition of  $^{137}\text{Cs}$  and arrival times of the radioactive fallout. Furthermore, a map similar to the Atlas was obtained (Supplementary Data – Fig. S1). Biases both in the activity concentrations and deposition are probably attributed to the uncertainties of the source term, the meteorological data used in the simulations and the deposition scheme used within the model. In addition, the prevailing environmental processes (such as vertical migration and/or runoff), and the background  $^{137}\text{Cs}$  deposition from releases that had occurred prior to the accident (e.g., from nuclear bomb testing) and the model did not account for may also play a major role.

## 5. Discussion and conclusions

We present a database with measurements of surface air activity concentration and deposition for the radionuclides  $^{134}\text{Cs}$  (air concentration and deposition),  $^{137}\text{Cs}$  (air concentration and deposition) and  $^{131}\text{I}$  (air concentration). Considering that  $^{131}\text{I}$  is a very short-lived radionuclide, hardly any further improvement in the database is expected, while airborne  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  are only significant due to resuspension events or redistribution after fires close to the CEZ (e.g., Strode et al., 2012). Therefore, the main improvement in the present database refers to the deposition of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ . The data originate from random surveys and monitoring stations (REM) organized by national authorities in Europe, where samples were collected and analyzed at the time of the accident and the very first years thereafter. In addition, observations from FSU countries have been included, which were taken into account for the construction of the Atlas, but they are missing since then and they have not been included in the public REM database for the Chernobyl data. The



**Fig. 7.** Validation of a Lagrangian (FLEXPART) and a Eulerian (INCA) model using (a) surface concentration observations and (b) the enhanced gridded database of deposition of  $^{137}\text{Cs}$ . About 72–81% of the modeled deposition values were within a factor of 10 from the observations and 55–77% of the modeled concentrations fell within the same factor.

two deposition databases are stored (<http://radio.nilu.no>) in the form of restricted data (original measurements) and gridded data (after applying IDW interpolation, similar to the Atlas). The first can be accessed upon request simply explaining the purpose of its use, while the gridded data are directly available in the link <http://radio.nilu.no/Chernobyl/Griddeddata.aspx>.

The main goal for gathering all the observations of deposition and surface air concentration of radionuclides over Europe into a unique database is multifold: (i) First priority is to rescue the data since measurements that have been used for mapping radionuclide distribution after Chernobyl might have been lost (e.g., from the 399,533 observations supposedly used for the creation of the  $^{137}\text{Cs}$  deposition map of Atlas, only 4081 are publicly available), (ii) to make data available for model validations, and/or (iii) for health assessments for humans that suffered from the accident and biota that live and reproduce in highly contaminated zones (e.g., in the CEZ), given that such observations have not been included in any easily accessible database since 1998. Using such data to conduct health assessments leads to an additional ability to assess the performance of dose-response models used in radiation protection of humans (e.g., linear non threshold model, hormesis model etc.) (EPA, 1999; ICRP, 2005; NRC, 2006; Tubiana et al., 2009), as well as radioecological models for biota (e.g., ERICA tool, RESRAD biota etc.) (Larsson, 2008; Yu et al., 1993). The main limitation of these datasets is the lack of data in Northern and Eastern Russia, despite the deposition of  $^{137}\text{Cs}$  in these regions is expected to be far below  $40 \text{ kBq m}^{-2}$  (according to model calculations and the Atlas map), which corresponds to non-contaminated areas. However, the authors aim at further enriching the database in the future and we welcome additions from any research group willing to provide observations of surface air activity concentrations and deposition densities of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{131}\text{I}$ .

We evaluate the database by comparing the deposition map of  $^{137}\text{Cs}$  created using the gridded observations, which resulted from IDW interpolation of the original measurements, similar to the Atlas (De Cort et al., 1998). The results are very similar at capturing very well all the highly contaminated zones over Europe as defined in the Atlas. We estimate that around  $75 \text{ PBq}$  of  $^{137}\text{Cs}$  were deposited over Europe as of May 1st, 1986, which is very similar to the total estimated emissions of  $^{137}\text{Cs}$  from the Chernobyl accident. The difference is clearly attributed to the lack of observation over a large territory (Russia). Furthermore, country-by-country deposition of  $^{137}\text{Cs}$  was also compared to reported values in the Atlas showing that the depositional patterns in the two datasets are similar. Finally, the same database presented here for  $^{137}\text{Cs}$  has been also used to assess the validity of a Eulerian chemistry transport model showing good correlations in air concentrations and deposition all over Europe with relatively small biases.

We further enriched the current dataset by adding 48,781 deposition measurements of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Am}$  from an area inside the CEZ corrected for decay to the year 2015. The aim of using these measurements was to study the recent fires in the CEZ in April and August 2015. They are kept under the same policy as the original observations from the Chernobyl accident and can be accessed upon request. According to them, the highest values are observed for  $^{137}\text{Cs}$  (average:  $482 \pm 1021 \text{ kBq m}^{-2}$ ) and  $^{90}\text{Sr}$  (average:  $144 \pm 446 \text{ kBq m}^{-2}$ ). Transuranium elements were found to be much lower with an average for  $^{238}\text{Pu}$  to be  $1.5 \pm 4.6 \text{ kBq m}^{-2}$ ,  $1.2 \pm 3.7$  for  $^{239}\text{Pu}$ ,  $1.8 \pm 5.5 \text{ kBq m}^{-2}$  for  $^{240}\text{Pu}$  and  $5.6 \pm 17.5 \text{ kBq m}^{-2}$  for  $^{241}\text{Am}$ .

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2016.05.030>.

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