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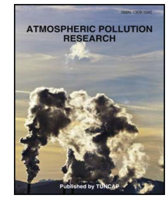

The Chernobyl nuclear accident ^{137}Cs cumulative depositions simulated by means of the CALMET/CALPUFF modelling system

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A B S T R A C T

The widely used dispersion modelling system CALMET/CALPUFF has been applied in order to evaluate its ability to simulate dry and wet depositions at regional scales (up to 1000 km from a source) in the specific case of radionuclides released in the atmosphere, during the 1986 Chernobyl Nuclear Power Plant accident. The ^{137}Cs cumulative deposition data sampled at 410 sites on the entire territory of Ukraine after the accident have been used for model verification. As meteorological input for feeding the CALMET pre-processor, we used a dataset of time series recorded in 211 surface stations, 194 precipitation stations and 14 upper air stations. Two different schemes for the emissions source have been adopted both available from scientific literature on pollutants release during the Chernobyl accident. This work shows that the CALMET/CALPUFF system is able to reproduce the large-scale features of the measured ^{137}Cs deposition pattern, which are the main traces on the territory of Ukraine. However, the fine structure of depositions, which are mainly due to precipitations, are poorly caught. The simulated deposition pattern appears excessively smoothed and an explanation for that is provided. Besides, we have found that the resistant model for dry deposition velocity of ^{137}Cs aerosol particles significantly underestimates depositions and the closest agreement with measurements is achieved with constant deposition velocity of 0.005 m/s. Finally, the strong dependence of the simulated contamination pattern on the emission source parameterization is confirmed.

1. Introduction

Atmospheric dispersion models are powerful tools to give answers to many scientific and socio-ecological questions. The most important of them are: (1) the quantification of impacts due to accidental releases of hazardous (radioactive or toxic) substances into the atmosphere, (2) the assessment of influence of routine anthropic emissions on human beans and the environment. There exists a large variety of dispersion models, which differ in terms of their areas of focus, general level of sophistication, spatial scales of application, etc. (EPA, 2003; Leelossy et al., 2014). Before these models could be used in decision making processes it is necessary to ensure that they generate reliable simulations.

Among the most commonly used models for both local and regional scales there is CALPUFF (California Puff) (e.g. Carizi et al., 2000; Levy et al., 2002; Elbir, 2003; Zhou et al., 2003; Grogan et al., 2007; Rood et al., 2008; Giaiotti and Stel, 2011; Escoffier, 2013; Ivančič and Vončina, 2014; Lee et al., 2014; Schramm et al., 2016; Kovalets et al., 2017). CALPUFF is a multilayer, multispecies, non-steady-state Lagrangian puff modelling system that simulates the effects of time- and space-varying meteorological driving forces on pollutant transport, dispersion, transformation and deposition. It consists of three main modules: CALMET (meteorological preprocessor), CALPUFF (dispersion model) and CALPOST (postprocessor) (Scire et al., 2000a, 2000b). This modelling system is recommended by the US Environmental Protection Agency (EPA) for long-range transport and some other specialized regulatory applications (EPA, 2017).

CALPUFF simulations have been extensively compared against sets of field experiments or air quality monitoring data, assuming sources of different types (surface/elevated, point/line/area/volume, instantaneous/finite/constant), at different temporal (short/long-term) and spatial (near/far-field) scales, over various topographies (simple/complex) and land use/land cover characteristics (natural/urban). However, the comparisons have been mainly focused on pollutant concentration in the air and rarely on pollutants depositions at the ground.

There is no unique and standard way to assess the reliability of a dispersion models due to the complexity of the pollutants dispersion,

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transformation and removal processes. Furthermore, the variety of static boundary conditions, namely orography and land use, the meteorological driving forces and the pollutant source features make the evaluation multidimensional. Practically, the goodness of model is established by means of case studies focusing the attention on closeness between measurements and simulation of specific fields. It is the blend of all the evaluations that gives the overall quality of the model. This work aims to add a further piece to the puzzle of CAMET/CALPUFF evaluation in particular with respect to the ability of the code to reproduce depositions.

In literature, there are many information useful to assess the CALMET/CALPUFF performances, but only a small fraction of this rich set concerns the depositions; usually only concentrations are considered. Near-field good performances to reproduce pollutant concentration, released from instantaneous point and line sources over flat areas with surrounding mountains have been reported by Chang et al. (2003). Protonotariou et al. (2005) evaluated CALPUFF's reliability performance in an urban domain with complex topography. The comparison of simulated NO_2 and PM_{10} with the measurements is considered satisfactory, particularly in the case of unstable atmospheric conditions. The evaluation of CALPUFF in complex topographic conditions, by Cui et al. (2011), shows that the modelling system performs reasonably well in terms of predicting the concentration of pollutants released from a point source. Dresser and Huizer (2011) obtained better performances of the CALPUFF model compared to AERMOD when evaluating the simulations of SO_2 concentrations. Rood (2014) compared the performance of four dispersion models, including CALPUFF, in a domain with complex terrain. According to the author, none of the models out-performed the others in reproducing the tracer concentration released from a point source. Tartakovsky et al. (2013) examined AERMOD and CALPUFF's predictions of particulate matter (PM) concentration released from area sources (a quarry) located in complex topography. The authors found that AERMOD gives better agreement with the measured data. Somewhat better performance of AERMOD compared to CALPUFF was also reported in (Jittra et al., 2015), where emissions of NO_2 and SO_2 from numerous point sources in an urban area were modeled and analyzed. Acceptable performance of the CALPUFF model based on the measurements of NO_2 and SO_2 concentrations released from a point source in an urban area was carried out by Affum et al. (2016). Holnicki et al. (2016) applied CALPUFF in an urban domain with available air quality measurements demonstrating a good agreement between simulation and reality for long-term averages, while the reproduction of the short-term (1-h average concentrations) was much less accurate, particularly for the low-wind meteorological episodes.

Concerning far-field (long-range) evaluation studies. Irwin (1997) compared the CALPUFF model concentration against field experiment data showing a relatively good agreement between the predicted and measured concentrations at large distances from the source, namely 48 km and 90 km, and larger differences near the source, 3.2 km by the source. An important evaluation of the CALPUFF system was reported in EPA documents (EPA, 1998; EPA, 2012). In the first study, CALPUFF's tracer concentration field was compared with two datasets, measurements at 100 and 600 km from a source. The authors found most of the modelling results in agreement with the observations. However, the second study, which contains evaluations of CALPUFF by means of data collected during four long-range dispersion field experiments, including those considered in (EPA, 1998), showed inaccuracies of the model outputs. The authors found that the CALMET/CALPUFF's concentration are highly variable depending on CALMET input options. Anyway some evaluations of (EPA, 2012) are discussed also in (Scire et al., 2012) pointing out that (EPA, 2012) work contains flaws, which significantly affect conclusions regarding CALPUFF's performance.

In summary, it can be concluded that a majority of the evaluation studies reported a good agreement between the CALMET/CALPUFF simulations and the measured pollutant concentrations in the air.

Apart from the prediction of airborne tracer concentrations, the CALPUFF model is also often used to calculate deposition fluxes of various chemical compounds to the ground (e.g. Pfender et al., 2006; Poor et al., 2006; Scorgie and Kornelius, 2009; Tartakovsky et al., 2013). However, the number of studies, evaluating the CALPUFF system against field deposition measurements is very limited in comparison to those focusing on concentrations. Some results can be found in (Macintosh et al., 2010; Mangia and Cervino, 2012), where the model's predictions were compared with deposition measurements in a near-field, complex terrain setting and a good agreement was reported. At the same time, the capability of the CALPUFF system to predict deposition processes properly at long-range or regional scales has not been studied at all, to the best of our knowledge. Therefore, it is highly desirable to test the simulating performances of CALPUFF in terms of deposition fluxes at such scales.

Thus, the main objective of this work is to evaluate the capability of the CALMET/CALPUFF modelling system to reproduce both wet and dry deposition processes at regional scales, that is up to 1000 km from the source. The Chernobyl Nuclear Power Plant (CNPP) accident releases were considered as an appropriate "field experiment" and ^{137}Cs cumulative total (dry + wet) deposition data on the territory of Ukraine after the accident were used as an evaluation database.

Furthermore, it is important to note, that in spite many years have already passed since the catastrophe, the simulation of radionuclides transport, dispersion and deposition after the CNPP accident still remains a challenging problem (e.g. Brandt et al., 2002; Talerko, 2005; Evangeliou et al., 2013; Simsek et al., 2014). The reasons for this are the uncertainties in the emitting source, according to (Kasparov, 2016) a completely satisfactory model of the Chernobyl source term has not been proposed so far, and the complex mesoscale meteorological conditions during the releases.

Besides the modelling aspects, it is out of doubt that the great amount of radioactivity that contaminated large areas all over Europe, continues to be a topic of interest.

After the CNPP accident, several long-range air dispersion models have been used to simulate the event and they were compared with the field measurements (e.g. Klug et al., 1992). Anyway, according to the above considerations, an application of the CALMET/CALPUFF modelling system to the long-range depositions of this widely known complex dispersion case, by means of a rich meteorological set of synoptic and mesoscale measurements and suitable description of the emission source, is still worth. Additionally, given that the source uncertainty is significant, it is important to check how various commonly used parameterizations of the source affect the predicted final contamination pattern of ^{137}Cs on the territory of Ukraine. It is expected that the comparison of these modelling results with the observed contamination pattern can help improve the source description.

The next section describes data and methods used in this study. The results and discussion are presented in Section 3. Lastly, Section 4 contains our conclusions and our outlook on the CALMET/CALPUFF modelling software.

2. Materials and methods

2.1. Evaluation data

We used data of ^{137}Cs ground depositions over the territory of Ukraine. The data were collected by the Ukrainian authorities in the early 1990s using a combination of a soil sampling method and airborne gamma spectrometry. We recall that the CNPP accident occurred on April 26, 1986. Soil samples were mainly taken in population aggregates, villages and towns, because the aim was to assess the impact of radioactivity on residents and depending on the size of the population aggregates, several measurements were done in each of them. In our study, we used the averages over each village or town.

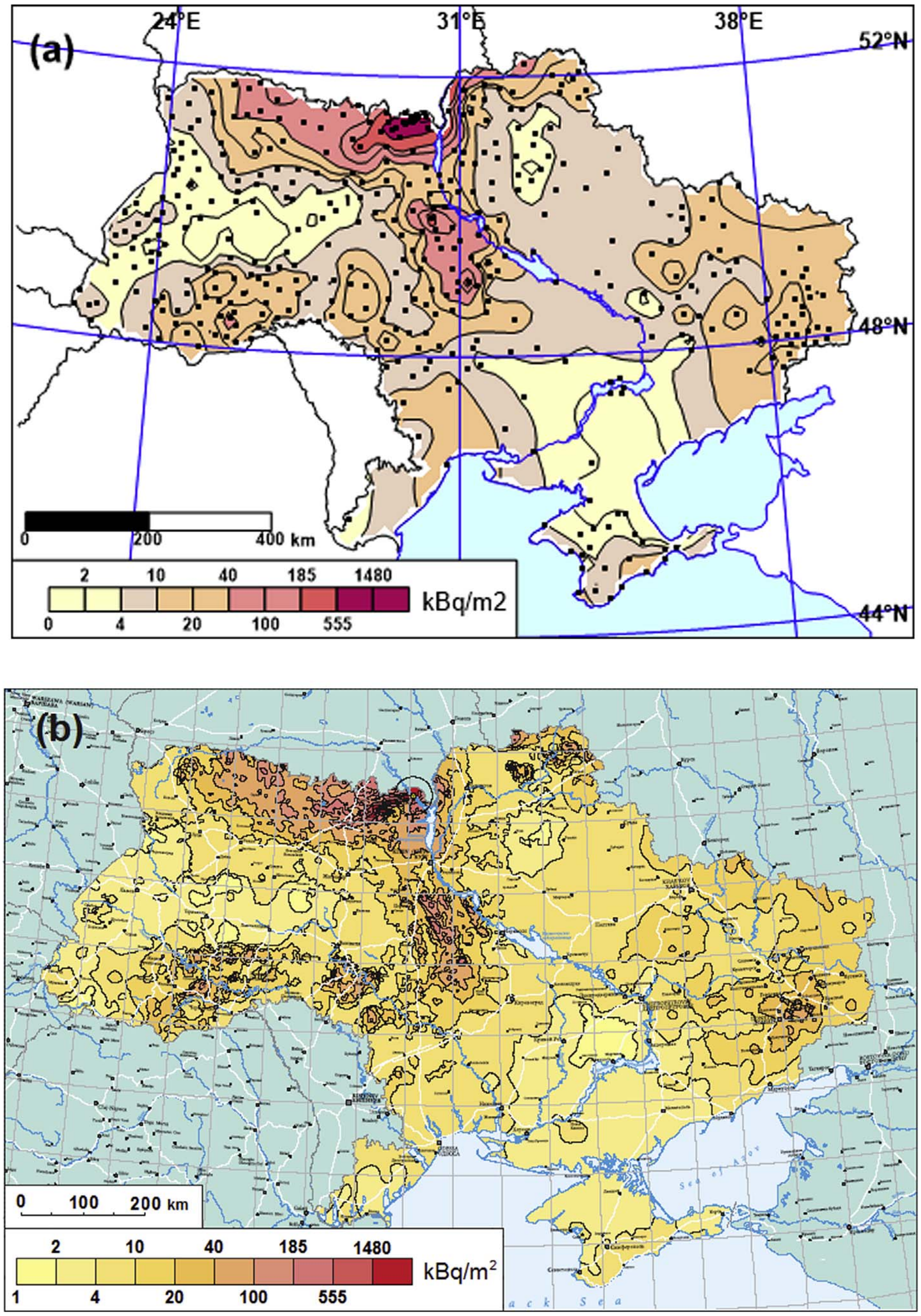

Fig. 1. (a) Location of 410 evaluation points (black dots) on the territory of Ukraine and the ^{137}Cs contamination pattern built on their base; for comparison (b) the ^{137}Cs contamination of Ukraine adapted from (De Cort et al., 1998). All maps in the manuscript with exception of Fig. 1 (b) were built by means of Surfer 10 software.

The deposition data are provided by the Central Geophysical Observatory (CGO), which is the observation institution of the Ukrainian Weather Service. In particular, these data were used to generate the Ukrainian part of the well-known atlas of De Cort et al. (1998). The deposition data have been reduced to the 10th of May 1986 applying a formula of radioactive decay, as in the De Cort et al. (1998) atlas.

We used 410 measurement sites and their locations are shown in Fig. 1 (a). In this figure, the interpolation of the deposition data into a regular grid, which is identical to meteorological/computational grid of CALMET/CALPUFF simulations, is shown as well. The interpolation was performed by means of the ordinary kriging method. In Fig. 1 (b), the map of ^{137}Cs contamination of the Ukrainian territory adapted from (De Cort et al., 1998) is presented for comparison. As it can be seen from Fig. 1 (a) and (b) the number of evaluation points used in our study capture the main features of the deposition pattern and the local scale maxima too.

According the aim of this work, it would be valuable to use daily or subdaily deposition measurements but unfortunately there are no such data for the Ukraine area, apart those presented in graphical format by (Izrael et al., 1990) at two meteorological station, Baryshivka, and Kyiv. Those data have been considered insufficient for any reliable evaluation of the model at those time scales. Therefore, this work does not consider the daily evolution of the depositions.

2.2. The emission source

One of the most important input for dispersion models is the emission source. The CNPP accident releases are considered extremely powerful and dangerous with respect to international health and environmental standards. Overall ~14 EBq of radioactivity (including ~85 PBq of ^{137}Cs) were emitted into the atmosphere according to (Izrael et al., 1990). Therefore, the emission rate and other source characteristics were modeled.

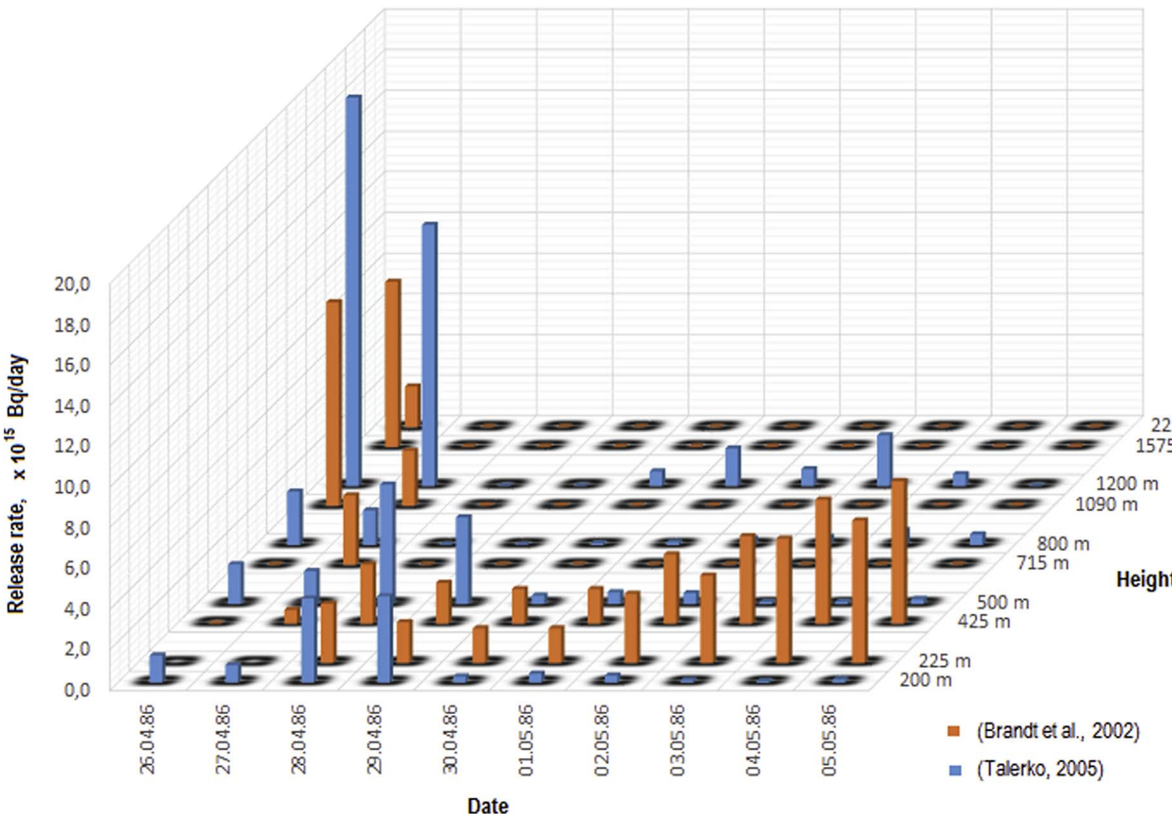

Fig. 2. Time evolution and vertical distribution of ^{137}Cs releases during the CNPP accident.

In our simulations, we used two different schemes of temporal evolution and vertical distribution of ^{137}Cs emissions. The first scheme was taken from (Brandt et al., 2002), the second one from (Talerko, 2005). In (Brandt et al., 2002) source model there are 6 emission points at highs of 225, 425, 715, 1090, 1575 and 2225 m above the ground, whereas only 4 points, at 200, 500, 800 and 1200 m, are used in (Talerko, 2005). The schematic representation of the time evolution of sources rates at a daily scale is shown in Fig. 2. As can be seen from the figure, the source models are significantly different. Both models have the bulk of the emissions at the top during the first two days since the CNPP accident, while Talerko's model (Talerko, 2005) assumes a short tail at lower levels and significant releases at 1200 m about one week after, Brandt's model (Brandt et al., 2002) describes a long lasting emissions close to the ground with daily ratios increasing up to 10 days after the initial phase of the accident.

In our study, we assumed that the ^{137}Cs releases consisted of condensed aerosol particles only, with the following characteristics: the geometric mass mean diameter of 1 μm, geometric standard deviation of 2 μm and particle density of 2.5 g cm^{-3}. The characteristics of ^{137}Cs aerosols are comparable to those used in similar simulations of accidental radioactive releases (Brandt et al., 2002; Baklanov and Sørensen, 2001; Talerko, 2005; Evangeliou et al., 2013).

2.3. CALMET/CALPUFF simulations of the Chernobyl accident releases

2.3.1. Modelling domain

In our study, we used CALMET and CALPUFF of Version 5.8.4, which are approved by the US EPA. The modelling domain is shown in Fig. 3. SRTM30 (Shuttle Radar Topography Mission, terrain elevation data) and Global/Eurasia Land Cover Characteristics Data Base (land use/land cover data) were used as geophysical inputs for the simulations. As it can be seen from the figure, the domain is quite flat with a few exceptions on its edge (the Carpathian, Crimea, and Caucasus mountains), which did not have a big influence on the dispersion and deposition processes over the territory of Ukraine.

LCC (Lambert Conformal Conic) geographical map projection was used for the CALMET meteorological grid as well as for the computational and sampling CALPUFF grid. The grid resolution is 15 km and it accounts for 98 × 75 grid cells. Besides the sampling grid, discrete receptors have been defined, which coincide with the measurement locations.

Vertical levels are set to 10, 30, 60, 120, 200, 290, 390, 500, 620, 800, 1040, 1280, 1550, 1850, 2250, 2750 and 3500 m above the ground and these 17 levels have been chosen to give a proper vertical resolution of the meteorological fields according to the emission altitudes defined in the emission sources.

The CNPP accident occurred on April 26, 1986, at 00:23, Kyiv summer time (21:23 on April 25 in UTC). Intense radioactive releases lasted until May 5 (Izrael et al., 1990; De Cort et al., 1998). Therefore, the simulated period is from April 26, 00:00 until May 10 23:00 (15 days) when most radioactive clouds left the territory of Ukraine.

2.3.2. Meteorological data

In order to calculate the CALPUFF's input meteorological fields by means of the CALMET preprocessor we used measurements from 211 surface weather stations, 194 of which are located in Ukraine and 17 in the neighboring countries, and 14 upper air stations, 9 of which are in Ukraine and 5 in the neighboring countries. Precipitation data are available only from the 194 Ukrainian surface stations. Station locations on the domain are shown in Fig. 4. All Ukrainian stations belong to the Ukrainian meteorological network that performs monitoring on a regular basis. The Ukrainian data were provided by CGO. The data from the neighboring countries were obtained from open-access Internet sources (NOAA, Wyoming University).

The Ukrainian meteorological network measurements are available with a 3 h time resolution and we performed an interpolation in time to achieve 1-hour resolution. Simple linear interpolation was applied for all parameters, except precipitations.

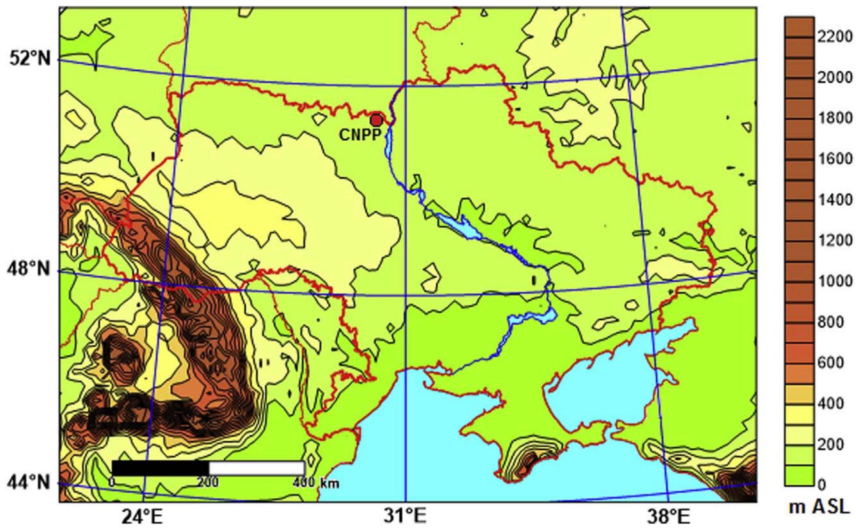

Fig. 3. The modelling domain. The map was built based on the SRTM30 terrain elevation data on a 15 km grid.

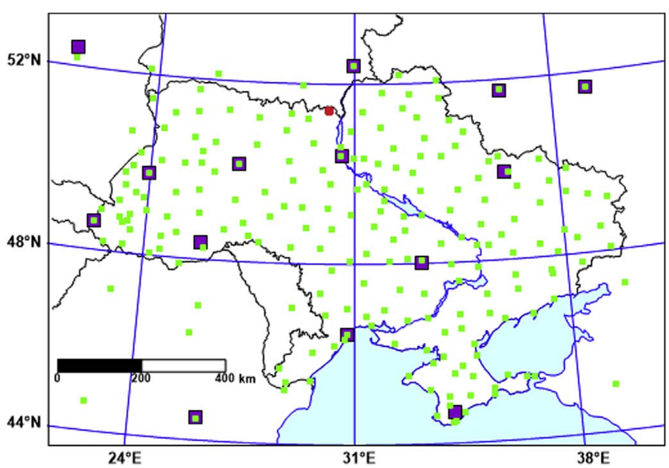

Fig. 4. Location of the CNPP (●), upper air (■) and surface (■) stations on the domain.

In Ukraine in 1986, precipitations at surface meteorological stations were measured four times a day, at 06:00, 09:00, 18:00 and 21:00 LST. In addition, the information regarding the beginning and the end of every rain/snow episode, as well as for other meteorological events, are reported in special tables. Thus, in order to interpolate in time the precipitation data, we equally distributed the amount of precipitations of each rain episode over the hours during which the rain was reported. Such simple interpolation scheme may produce hourly time series quite far from the reality. However, this approach allows the generation of hourly precipitation time series matching those of the other weather variables and, since we are interested in the integrated depositions over the whole simulated periods, it does not introduce large errors because the integrated wet removal is proportional to the total amount of precipitation.

The upper air data were collected twice a day or more, up to 4 sounding a day. They did not need any interpolation because the CALMET preprocessor does it.

All surface meteorological data were checked by METSCAN, one of the CALPUFF's preprocessors, to conduct the quality control. To avoid spikes in the meteorological inputs, checks on precipitation and upper air data were performed scanning the hourly precipitation field and the vertical profiles of all parameters for every measurement location.

2.3.3. Deposition calculations in CALPUFF

Dry deposition of particulate matter in CALPUFF is calculated on a full resistance model for the deposition velocity (Scire et al., 2000b). Besides the resistant model, a user defined constant, or varying during a day, deposition velocity can be used in the modelling system as well. Wet deposition fluxes due to precipitation scavenging are computed using an empirical scavenging coefficient approach. In our simulations, the scavenging coefficient for liquid precipitation was taken to be 10^{-4} s^{-1}.

2.3.4. CALMET/CALPUFF modelling protocol

There is a large number of control parameters in both the CALMET and CALPUFF initialization files. The default values were used for most of them. However, several controls are site/case specific and have to be defined based on the information about the domain terrain features, grid spacing, space distribution of the surface, upper air and precipitation stations used in simulations, characteristics of pollutant modeled, prevailing meteorological conditions during simulation, etc. (Barclay and Scire, 2011). In this work, to define the site/case specific controls we followed recommendations given in (Fox, 2009; Barclay and Scire, 2011; Scire et al., 2012).

As pointed out in many CALPUFF evaluation studies (e.g. EPA, 2012; Rood, 2014), the model's predictions are very sensitive to the values of the site-specific controls in CALMET. It is not a very simple task to define them properly a priori. Therefore, we performed several sensitivity tests with different values of site-specific controls within their range of reasonableness before to run the set of simulations, which we evaluated against measurements. In these adjusting iterations, in addition to CALMET control parameters we tested also several CALPUFF controls in order to avoid simulations clearly unrealistic in comparison with measured deposition patterns. We paid special attention to the controls responsible for the puff splitting, both vertically and horizontally. According to (Scire et al., 2012) the puff splitting is a very important setting for the CALPUFF model, particularly in the case of long-range applications.

Details on the CALMET/CALPUFF's site/case specific controls we used in simulations along with their short descriptions can be found in the supplemental materials (SM, Tables 1 and 2). Default values were used for all other control parameters.

2.4. Model performances evaluation

The following statistical tests and indices were employed to evaluate the accumulated total dry + wet deposition of ^{137}Cs: BIAS, FB (fractional bias), geometric mean bias (MG), normalized mean square error (NMSE), geometric mean variance (VG), Pearson's correlation coefficient (PCC), PCC for data in log-scale (PCC_{lg}), factor of excedance (FOEX), factor of 2 (FA2) and factor of 5 (FA5). Their comprehensive

Table 1
Evaluation statistics for the simulations.

Run	BIAS (kBq/m^2) Best 0	FB (%) Best 0	MG – Best 1	NMSE – Best 0	VG – Best 1	PCC – Best 1	PCC$_{lg}$ – Best 1	FOEX (%) Best 0	FA2 (%) Best 100	FA5 (%) Best 100
1	−52.90	−149	0.19	49.99	208.94	0.36	0.49	−33	28	49
2	−53.06	−138	0.51	43.12	17.89	0.50	0.34	−12	43	78
3	−21.00	−41	0.67	13.53	4.06	0.72	0.72	−12	44	84
4	−21.92	−41	1.42	9.62	10.36	0.72	0.43	10	33	70

description can be found in (Mosca et al., 1998; Chang et al., 2003). All the tests are performed using all the pairs of (M_i, C_i), $i = 1, ..., 410$, where M_i and C_i are the measured and calculated ^{137}Cs total deposition respectively at the i-th evaluation point. The need for several tests comes from the fact that different statistical indices can evaluate the model performance in different aspects. It should be noted that BIAS, FB, NMSE and PCC are more strongly influenced by infrequently occurring extreme values, whereas MG, VG and PCC$_{lg}$ provide a more balanced treatment of both high and low values (Chang et al., 2003). All the measured and calculated data in our study were greater than zero with the minimal value of $\sim 10^{-2}$ kBq/m^2. This is important because MG, VG and PCC$_{lg}$ cannot be run for zero values. In general, as it was pointed out in (Chang et al., 2003), for a dataset in which both the measured and calculated data vary by several orders of magnitude, MG, VG and PCC$_{lg}$ are more suitable.

A scatter plot and a Figure of Merit in Space (FMS) were also included in the evaluation procedure. However, we calculated FMS with a slightly modified formula. Usually, FMS is computed for a certain significant level using the expression (e.g. Mosca et al., 1998)

$$FMS = \frac{A_C \cap A_M}{A_C \cup A_M} \cdot 100\%, \tag{1}$$

where A_C and A_M are the simulated and measured areas respectively with contamination above the significant level. In our study, instead of a single significant level, we used more than one according to the map as in (De Cort et al., 1998). Their consecutive pairs define ten contamination intervals, and the right limit of the last 10-th interval is not bounded from above. We computed FMS for each of those significant levels as follows

$$FMS_j = \frac{A_{C\,j} \cap A_{M\,j}}{A_{C\,j} \cup A_{M\,j}} \cdot 100\%, \quad j = 1, ..., 10, \tag{2}$$

where $A_{C\,j}$ and $A_{M\,j}$ are the calculated and measured areas with the contamination in the j-th interval. Then the generalized or weighted estimate of FMS, which characterizes all significant levels (the entire contamination pattern), is expressed as

$$FMS_g = \sum_{j=1}^{10} FMS_j \frac{A_{M\,j}}{A}, \tag{3}$$

where $A = A_{M1} \cup ...\cup A_{M10}$. The best score for FMS$_g$ is 100%. It will occur when the calculated areas of all contamination intervals coincide completely with the corresponding measured areas. FMS$_g$ is a weighted average of FMS$_j$ where each weight is the fraction of the total area considered for the comparison, which refers to the j-th significance interval, so it is given more importance to the matching of the model outputs with measurements in extended areas than to the reproduction of small-scale deposition patterns.

To compute FMS$_g$ we used the total depositions simulated in discrete receptors coinciding with the evaluation points. Both the measured and simulated deposition data were interpolated on a regular grid using the same interpolation method. After that, all $A_{C\,j}$ and $A_{M\,j}$ were defined on the Ukrainian territory only (not in the entire modelling domain that includes neighboring countries).

3. Results and discussion

The results of four different simulations are presented here. In the first simulation, referred as Run 1, we used the emission source term proposed in (Talerko, 2005) while in the second simulation, Run 2, the source proposed in (Brandt et al., 2002) was employed. In both simulations, the full resistant model for the deposition velocity available in CALPUFF for dry deposition calculations was used. Two other simulations were performed with the same source terms, Run 3, by means of the same source as in Run 1 (Talerko, 2005), and Run 4, with the adopted in Run 2 (Brandt et al., 2002), but for both these simulations we used the constant deposition velocity of 0.005 m/s. The constant value was taken as an alternative to the resistant model. Such value for the deposition velocity of ^{137}Cs aerosol particles was taken from literature (Izrael et al., 1990) and it comes from the assessments made during the CNPP accident. The same value is used in the (Talerko, 2005) work.

The evaluation statistics for every Run are summarized in Tables 1 and 2. The scatter diagrams and the predicted final total deposition fields are presented in Figs. 5 and 6 respectively. The animations showing the calculated results, namely the wind field and ^{137}Cs concentration at the lowest vertical level, precipitation field, dry, wet and total (dry + wet) ^{137}Cs cumulative deposition fluxes to the ground, with the time resolution of 1 h can be found in the supplemental materials (SM, run1.mp4 – run4.mp4). Such animations were created for every simulation (including the initial iterations/sensitivity tests) in order to facilitate the analysis of the dispersion and deposition processes over the domain.

Supplementary video related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.11.007.

The best agreement between the simulated and measured depositions is obtained in Run 3, that is using Talerko's source (Talerko, 2005) and the constant deposition velocity of 0.005 m/s. In this run, all the

Table 2
Figure of merit in space (%) for the simulations.

Run	Limiting significant levels of total deposition (kBq/m^2)										FMS$_g$
	0–2	2–4	4–10	10–20	20–40	40–100	100–185	185–555	555–1480	> 1485	Best 100
1	7	7	24	18	3	1	0	0	0	0	15
2	12	21	29	19	7	4	2	0	9	0	20
3	12	14	29	26	8	15	5	24	20	100	22
4	13	15	28	11	6	12	1	15	30	100	18

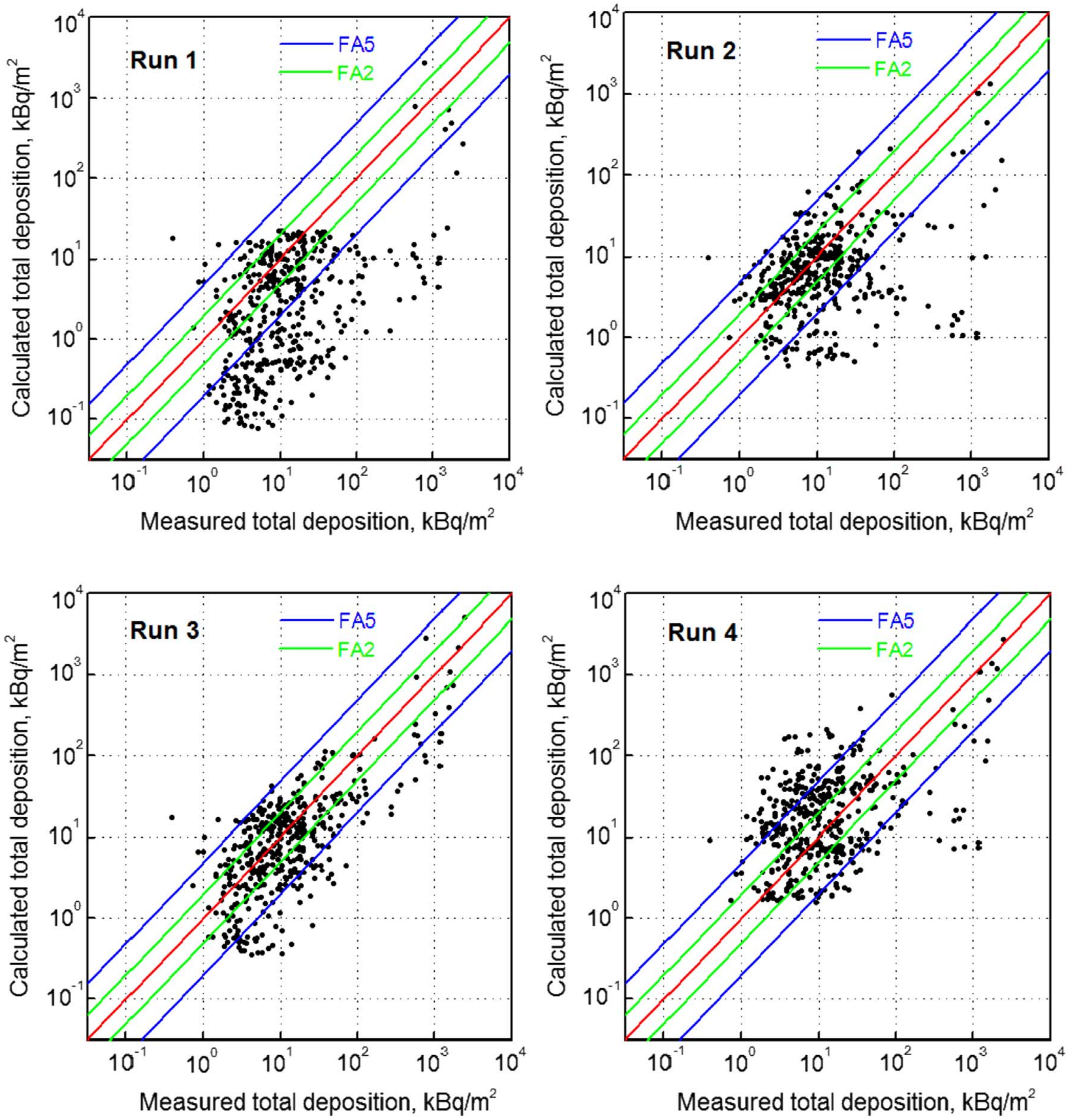

Fig. 5. Scatter diagrams for every simulation.

main traces, including the western and southern ones and large-scale local maxima in the eastern part of Ukraine, have been predicted with acceptable evaluation statistics. 44% of the predictions are within the factor of 2 and 84% are within the factor of 5. High correlation between the calculated and measured deposition data is also observed (PCC = 0.72). MG and VG have the values of 0.67 and 4.06 respectively, which are relatively close to their best estimates. According to Table 2, approximately 22% of the contamination pattern on the territory of Ukraine are modeled correctly in this run. Taking into account high sensitivity of FMS_g to wrong predictions (displacement) of the deposition pattern, this can be considered as a good result. Other evaluation statistics, with the exception of BIAS and NMSE show a rather good agreement between the predictions and measurements (see Table 1).

Run 4 also reproduces relatively good values for a majority of the evaluation statistics. However, the final deposition patterns is not realistic (compare Fig. 6, Run 4 and Fig. 1b). Both Runs with the resistant model for the deposition velocity, Run 2 and particularly Run 1, underestimate the depositions with respect to the measured data, although Run 2 produced quite high values of FA2 and FA5.

It should be noted that not all maxima have been caught in Run 3 as well as in other runs, in fact comparing Figs. 6 and 1b there are many discrepancies between the calculated and observed deposition patterns. The most problematic features of the deposition pattern are relatively small-scale local maxima, which are related mainly to wet depositions due to precipitations. As a result, the calculated final deposition patterns appear excessively smoothed with respect to the reality. We believe that one of the possible reasons for oversmoothing is the CALPUFF's computational algorithm for wet depositions. In the following sections, we discuss it along with other possible reasons for the discrepancies.

3.1. Reliability of wet deposition simulations

According to our experience in simulating the CNPP pollutants dispersion, we conclude that it is very difficult to define a set of CALPUFF control parameters suitable to reproduce wet depositions in the far-field dispersion cases. Our simulations show that CALPUFF is not capable to reproduce the fine structure of the deposition pattern in the areas located far from the source. Such a fine structure occurs

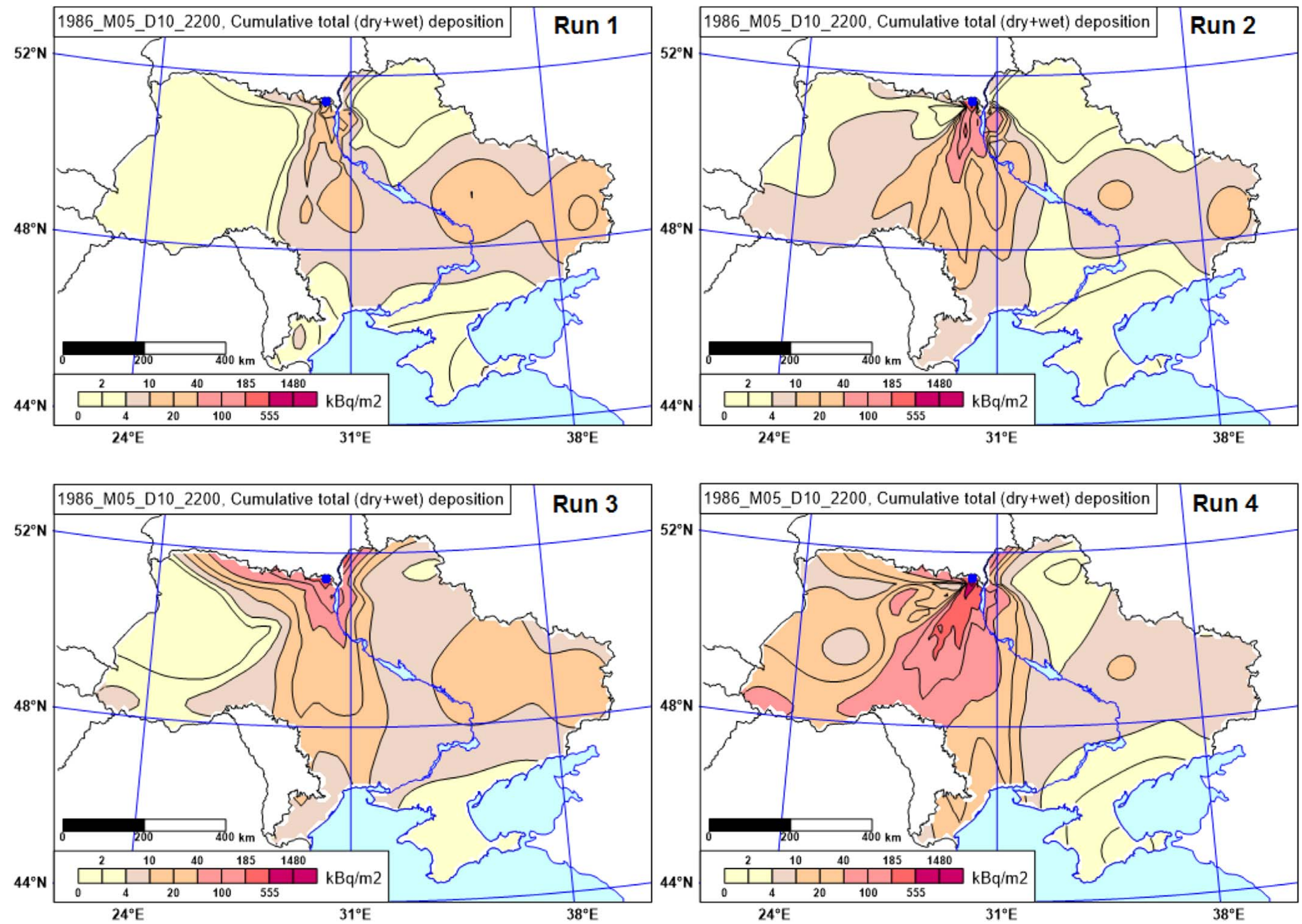

Fig. 6. The predicted final cumulative total (dry + wet) deposition of ^{137}Cs (at 22:00 on May 10, 1986). The results obtained with the source term of (Talerko, 2005) and (Brandt et al., 2002) are in the left/right panels respectively. The results obtained with the resistant deposition model and with the constant deposition velocity of 0.005 m/s are in the top/bottom panels respectively.

primarily because of precipitations, that is wet depositions.

In CALPUFF, wet fluxes from a puff towards the ground are calculated, at every computational step, using the precipitation amount at a single meteorological/computational grid point only, namely the closest one to the puff center. Then this calculated value is extended over the whole puff and it is described by the following formula (according to the CALPUFF Fortran code)

$$F_{wet}(i, j) = \frac{1}{\Delta t} M(i, j)\left(1 - e^{-\lambda \frac{R\left(i_{pc}, j_{pc}\right)}{R_1} \Delta t}\right), \tag{4}$$

where $F_{wet}(i, j)$ is the wet flux at any of the ground grid points (i, j) that are covered by the puff, Δt is the time step, $M(i, j)$ is the mass of the puff in a vertical column with 1 m^2 cross-section near point (i, j), λ is the scavenging coefficient, $R(i_{pc}, j_{pc})$ is the precipitation rate at the grid point that is the closest to the puff center (i_{pc}, j_{pc}), and R_1 is the reference precipitation rate of 1 $mm\ hr^{-1}$.

In our opinion, expression (4) is acceptable only for puffs which sizes are comparable with typical precipitation patterns. For rains dominated by meteorological mesoscale and convective phenomena, as in the CNPP case, such condition is true for near-field applications of the dispersion model. In the case of long-range dispersion, when puffs can become quite large in comparison with the precipitation area, it may not necessarily work well. When the amount of precipitations is highly variable in the horizontal directions, it is obvious that for quite large puffs, the algorithm may either overestimate and underestimate the wet fluxes quite significantly. According to (4), overestimation occurs when the precipitations interest the puff center only, whereas underestimation is expected when the precipitations overlap large parts of the puff but not the center. It is clear that in both cases the simulated deposition pattern will be smoother that the real one and the polluted area is unrealistic. By means of this work both situations can be clearly identified comparing the concentration, precipitation, and cumulative wet deposition fields at 16:00 on the May 1, 1986 for Run 3 (the animation run3.mp4 of SM). It is likely that the replacement of $R(i_{pc}, j_{pc})$ with $R(i, j)$ in formula (4), will provide a more realistic deposition pattern removing both such undesired effects. Of course, this assumes that precipitation field is defined in each (i, j) grid point. Such approach is shared by other puff/Lagrangian dispersion models (see e.g. (Brandt et al., 2002)).

Without any change in the CALPUFF code, to workaround the problem the split option, which is available in the CALPUFF model, can be used. Such code option activates the splitting of the puff into several pieces, when it becomes enough large. Anyway, this workaround has shortcomings.

3.2. Limiting action of the horizontal puff splitting

Radioactive materials are very specific pollutants. Even small amounts of radioactivity in the air can cause significant contamination of the ground when intense precipitations occur. Thus, in order to avoid the difficulties with the calculation of wet depositions mentioned above and to reproduce a realistic fine structure of the deposition pattern, we activated the puff split option of CALPUFF acting on almost every puff that becomes larger than the grid spacing (15 km in our simulations). Consequently, CALPUFF control parameters, which regulate the puff splitting were set. We acted on the CNSPLITH parameter, that is the minimal averaged concentration in a puff to be split, and on SYSPLITH, the minimal radius of a puff to be split (see Table 2 in SM for details on the values used for CNSPLITH and SYSPLITH). Two other control parameters for the horizontal splitting, namely NSPLITH and SHSPLITH (see Table 1 in SM for definitions), have to be also set to their minimal values. However, we found that they do not influence significantly the results.

Given that the ^{137}Cs emissions during the CNPP accident were very large and lasted 10 days, the use of the split option with small values of the horizontal splitting control parameters resulted in a significant growth of the number of puffs. Due to the complex meteorological conditions (e.g. change of wind direction) during the simulation period, many puffs were traveling several days over the domain and they had to be split several times. Consequently, the computational cost of the simulations increased dramatically. Moreover, aborts of CALPUFF's runs due to exceeding the maximal number of puffs (2000000 in our case) and running out of computer memory was frequently observed in the initial adjusting iterations.

The set of values for the horizontal splitting control parameters adopted in our final simulations, which are reported in Table 2 of SM, was an optimal balance between the computational cost and ability of the model to reproduce the observed deposition patterns.

3.3. Limiting action of the vertical puff splitting

Another cause of discrepancy between simulated and measured deposition pattern, might be the vertical puff splitting algorithm adopted in CALPUFF. In fact, in the model, one of the vertical splitting requirements is that puffs have to cross the mixed layer (puffs have to be previously below the height of the mixed layer and after above it, and they have to be uniform in the vertical direction). This condition is wired in the CALPUFF code and it is not manageable by means of initialization file. Such approach can lead to situations in which puffs emitted at night above the nocturnal boundary layer split only the next evening. In our application this generates inconsistencies in the first day of the CNNP accident simulation when the radioactive contamination interested the western territory of Ukraine. On that day (April 26, 1986), a very sharp rotation of horizontal wind component was observed in the CNPP area. At 08:30 Local Time (06:00 UTC) 110° clockwise rotation was measured in the layer from the surface to 1500 m, whereas at 14:30 Local Time (12:00 UTC) 70° clockwise occurred in the layer from the surface to 3000 m. (see Fig. 7, where the vertical profiles of the horizontal wind components observed in Kyiv radiosounding are shown). We conclude that the vertical splitting condition described above in the frame of the wind rotation is the reason for the poor reproduction of the western deposition pattern (compare Fig. 6, Run 3 and Fig. 1b).

3.4. Aspects limiting the quality of simulated depositions

Our simulations show that the resistant model for the dry deposition velocity of ^{137}Cs aerosol particles gives a substantial underestimation of the contamination with respect to the measurements (see Figs. 5 and 6, Run 1 and 2). A significant underestimation of the particular matter (PM) dry deposition, simulated by means of CALPUFF with the resistance model and compared with that simulated by means of AERMOD model, was reported in (Tartakovsky et al., 2016). The conclusion drawn in that work was that the difference in PM deposition fractions probably results from distinct estimation of the plume height above the ground utilized in the models.

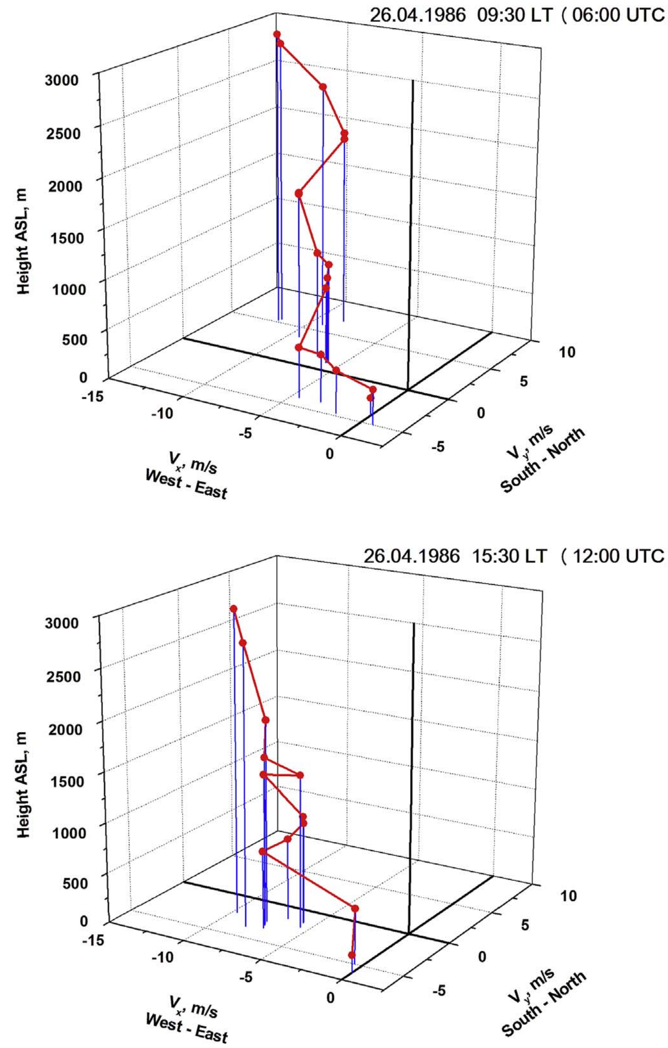

Fig. 7. Vertical profiles of the horizontal wind components on the first day of the CNPP catastrophe near the source (Kyiv radiosounding, ~100 km from Chernobyl).

The order of magnitude of the ^{137}Cs dry deposition velocity calculated with the resistant model, analysing the results of our simulations for runs 1 and 2, is approximately 10^{-4} m/s. This value is smaller than other previously published assessments by a factor of 10 (see e.g. Izrael et al., 1990). As it was pointed out above, the constant deposition velocity of 0.005 m/s gives much better results. However, the constant deposition rate does not take into account many features of the deposition processes (e.g. a diurnal cycle) that can have a profound effect on the contamination pattern.

Besides the reason mentioned in (Tartakovsky et al., 2016) that it is likely also for our case study, we consider worth other two explanations of the low simulated depositions. The first one is an incorrect representation of the physical properties of ^{137}Cs aerosol particles used in the simulations, while the second is an ineffective description of the source (height or/and emission rate). Additional modelling exercises, in particular such that include a comparison with the results of other Lagrangian models as it was done in (Tartakovsky et al., 2016), would help to clarify this problem.

In the scatter diagrams and in Table 1 it can be seen that not only simulations with the applied resistant deposition model, but also other simulations produce results that underestimate the measured values with quite large magnitudes of BIAS, FB and NMSE. Such values of the statistical indices are yielded mainly due to large negative differences between the calculated and measured depositions in the range of the highest values of contamination, that is the three last intervals of the contamination scale. All sampling sites for these high contamination values are located in the area closest than 60 km from the source and mainly the western trace. The location of the considerable difference can be attributed to the fact that the ^{137}Cs contamination in this area was caused not by means of condensed ^{137}Cs aerosol particles only, but also because of nuclear reactor fuel particles, which have significantly larger density (up to 10 g/cm^3) (Kuriny et al., 1993; Kasparov, 2016). According to the direct measurements reported in (Kuriny et al., 1993) from 25 to 75% of ^{137}Cs contamination in 60 km zone is due to the fuel particles, which were injected in the atmosphere during the accident. Such contribution to the deposition was found also far away from the source (Pöllänen et al., 1997). Therefore, the source term should include fuel particles. Otherwise, since the effects of fuel particles are relevant mainly 60-km by the source that zone should be excluded from the domain and modeled separately.

There are two more reasons for the underestimation of the predicted ^{137}Cs depositions. The first one is that we do not take into account the ^{137}Cs depositions due to the nuclear weapons tests that happened before April 26, 1986. In the Atlas (De Cort et al., 1998), the common contamination (after the CNPP accident and other sources that had happened earlier) is presented. The second reason that could cause the deviation of the simulation from the measurements is that we do not take into account those parts of the radioactive clouds that left the modelling domain and came back later. Such situation happened during the first 5 days after the accident, according to the modelling results in a larger domain (Brandt et al., 2002; Evangeliou et al., 2013; Simsek et al., 2014).

3.5. *The source model for the CNPP accident releases of ^{137}Cs*

When comparing results of Run 1/3 with Run 2/4, it becomes evident that the main uncertainties in the predicted final deposition pattern were due to the emission source data. The contamination patterns obtained with different sources are significantly different. The predictions that are based on the source data presented in (Talerko, 2005) appear to be more reliable. Such a conclusion is hardly surprising because Talerko's source was fitted according to the contamination of the territory of Ukraine (Talerko, 2005). However, an adjustment was also applied to the dispersion model (namely, LEDI) and the source term has not been tested with any other dispersion model and in a larger (European/continental) domain, to the best of our knowledge.

On the other hand, the simulations based on the source data presented in (Brandt et al., 2002), have been compared only with the measurements taken far away from the source and neither the Ukrainian nor the Belorussian ^{137}Cs deposition data have been never used in such studies (e.g. Brandt et al., 2002; Evangeliou et al., 2013; Simsek et al., 2014) apart form the recently published work by Evangeliou et al. (2016). In our opinion, the source term published in (Brandt et al., 2002) should be modified. The release at height below 500 m on the first day of the catastrophe should be added, otherwise it is impossible to reproduce the western (south-western) trace on the territory of Ukraine due to the sharp shift of wind direction near the source. Furthermore, the emission rate on the last days appears to be too large, causing overestimates in the southern trace on the territory of Ukraine.

As it was pointed out in (Kasparov, 2016) and (Evangeliou et al., 2016), so far there has been no consensus on the dynamics and magnitudes of the radionuclide releases during the CNPP accident, especially for volatile radionuclides. We believe it is worthwhile investigating an improved ^{137}Cs source model, that includes the strengths of the both source schemes considered above and the fraction of fuel particles.

4. Conclusions

This paper presents the evaluation of the CALMET/CALPUFF modelling system against the Chernobyl ^{137}Cs deposition data measured on the territory of Ukraine after the CNPP catastrophe. The modelling system has been mainly tested for its ability to predict properly dry and wet deposition processes at regional scales (up to 1000 km from the source). The satisfactory performance of the CALPUFF model has been obtained setting the constant deposition velocity to 0.005 m/s and using the source emission term, presented in (Talerko, 2005). In this simulation, the main large-scale features of the ^{137}Cs deposition pattern and the local maxima on the Ukrainian territory have been satisfactory reproduced according to the evaluation statistics.

However, the calculated contamination pattern is smoother than the reality because the fine structure of the depositions, that is the relatively small-scale local maxima, which were created mainly by means of precipitation, are beyond the limits of CALPUFF modelling system. The analysis of the wet deposition process representation in the model code suggests that the formulas used to describe it are not enough performant, so they can lead to either large overestimation or underestimation of the deposition. When the dispersion is modeled at regional scales and the puffs become rather large compared to the precipitation area oversmoothing occurs. To mitigate this effect without making changes in the code, the horizontal puff split option has been used, but the problem still persists. Moreover, the puff splitting causes a significant growth of the computational cost. An improvement of the CALPUFF's wet deposition algorithm has been proposed involving precipitation rates of all grid points falling in the puffs. We believe that algorithm also leads to the decrease of the computational time.

Our simulations with the resistant model for the dry deposition velocity of ^{137}Cs aerosol particles have shown substantial underestimation in comparison with the measured data. The magnitude of the deposition velocity for ^{137}Cs aerosol particles, calculated in CALPUFF with the resistant model, is an order of magnitude smaller than in other previously published assessments. The possible reasons for the decrease of the deposition rate might be attributed to the incorrect particle properties or to the wrong specifications of the source term. However, further investigation of this problem is necessary.

It has been also shown that the predicted contamination pattern depends strongly on the source term employed in the simulation. The uncertainties in the CALPUFF's results mainly come from using insufficiently accurate source models. The need for an improved source term of the CNPP accident releases that includes fuel particles has been pointed out.

In spite of the fact that a complete and suitable set of meteorological information, including surface, upper air and precipitation data, was used as driving forces inputs for the simulations, it is not excluded they could add uncertainties to the results.

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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.apr.2017.11.007.

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