Atmospheric Pollution Research xxx (xxxx) xxx–xxx

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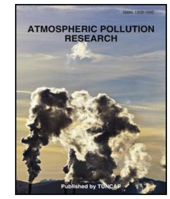
Atmospheric Pollution Research

journal homepage: www.elsevier.com/locate/apr




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

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A R T I C L E I N F O

*Keywords:*
CALMET/CALPUFF modelling system
Wet and dry deposition simulations
Model performance evaluation
Chernobyl accident
$^{137}$Cs depositions

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,

Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control.
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https://doi.org/10.1016/j.apr.2017.11.007
Received 15 August 2017; Received in revised form 7 November 2017; Accepted 8 November 2017

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.