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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inverse modelling of the Chernobyl source term using atmospheric concentration and deposition measurements" *by* Nikolaos Evangeliou et al.

Nikolaos Evangeliou et al.

nikolaos.evangeliou@nilu.no

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The manuscript under review presents an inversion analysis using the FLEXPART model of radio-isotopes of Cs and I, emitted after the Chernobyl nuclear power plant accident, using an expanded set of measurements (including deposition) to estimate the source term. The source estimates were evaluated using a Eulerian transport model, and systematic uncertainties are quantified with sensitivity tests. The authors address scientific questions within the scope of ACP, with valid scientific methods and assumptions. Overall, the description of experiments and calculations are sufficiently complete and precise. The language and presentation can be somewhat improved (see specific

comments below).

Specific Comments: ========== 1. On the choice of re-analysis dataset - isn't ERA-Interim more suitable as a primary database due to the higher resolution and 4D-Var assimilation? Also, see point 6 below on how it was decided which dataset gives better results. Can't the uncertainty be additionally quantified using an independent reanalysis dataset to drive the inverse model (e.g. NCEP)? Response: Indeed, we expected that ERA-interim would give better results since it's a newer, highly resolved assimilated dataset. However, it was clearly shown in the manuscript why it was not selected (detailed analysis in section 4.1). We agree that a better uncertainty could be quantified using an independent dataset. In fact, we tried to do what the reviewer suggested and get FNL and GFS fields from NCEP for 1986, for which we have tested flexpart in the past. However, neither the FNL data (https://rda.ucar.edu/datasets/ds083.2/) nor the GFS (https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forcast-system-gfs) were available for 1986 for use in flexpart model. Of course, there are other re-analysis datasets from NCEP that cover 1986 meteorology. Though, they do not contain all the necessary variable for a proper run with the flexpart model and they have never been tested before. Therefore, we ended up using 2 different products from ECMWF.

2. Though there is indeed uncertainty about the gas/particulate fraction of Iodine-131 emitted at Chernobyl, it's not well advised to model the radionuclide in the particulate form. Atmospheric measurements have revealed a gaseous/total atmospheric ratio of âĹij80% (Ring of Five). This could significantly impact atmospheric residence times and deposition patterns. Thus a quantification of the uncertainty impact on the final emission estimate is required. Response: The partition of gaseous and particulate iodine has been well studied in the recent accident of Fukushima and, as stated in the Ring of Five measurement network, it is close to 20% in particulate form and 80% as a gas. When performing inverse calculation of iodine, there is no point to state "it is not advised to model iodine as aerosol", because everything depends on the available

measurements that will be used to perform the inversion and improve the source-term. In the case of Chernobyl, the existing infrastructure of 1986 resulted in having available observations of the particulate fraction of iodine and, thus, this is the reason that we modeled iodine as an aerosol in flexpart. It would make no sense to model iodine as a gas and perform the inversion, while having aerosol surface activity concentration measurements. This would be out of question. Nevertheless, all the above do not, of course, imply that there was iodine only in particulate form released from Chernobyl in 1986. Of course, there should be gaseous iodine as well, which we cannot calculate with inverse modeling due to the lack of gaseous-phase measurements of iodine.

3. Section 3.3: When dealing with priors as an ensemble are they simply numerically averaged or do their individual uncertainties used to weight (as is more appropriate)? Response: As it is stated in the paper, the prior estimation of the source emissions was calculated by assessing the core inventory of the reactor and estimating what has been consumed from the last time that the fuel was updated (see references within the paper). In this way, they were able to assess what it has been produced in the nuclear reactor and, thus, what it has been released. All these happened in 1986. In all these inventories a constant relative uncertainty of 50% was assigned, which stems from this rough estimation of the source term that was rather arbitrary. This is the reason that we have not weighted the prior ensemble using individual uncertainties; simply because we know absolutely nothing about the daily uncertainties. Therefore, we thought that the best was to simply average prior emissions numerically.

4. Section 3.4: The i) magnitude, and ii) doubling of deposition over concentration uncertainties seems arbitrary, and should be better motivated. Response: We agree with the reviewer's comment and we have further given more information on the use of these uncertainty values (see page 11-Manuscript with track changes enabled).

5. Section 3.5: Doesn't nudging the dynamical model to ERA-Interim (used in the inversion) limit it's potential as an independent check of the emission inventory improvement? Response: We fully agree with this comment on the use of data from ECMWF

only (ERA-40 & ERA-Interim). However, the two available datasets from NCEP (GFS & FNL), which have been tested and found to contain all the necessary variables that flexpart needs, in order to run, are limited to more recent years. Please see a more detailed response in the first comment.

6. Section 4.1: It is not clear if comparison of ERA-40 and ERA-Interim was and selection of the "proper" using RMSE was done selectively for certain "previously inaccurate" regions only. Perhaps the text can be reworked to be made clearer? Response: We appreciate the reviewer for pointing us to section 4.1, because, indeed, it was not clear. We have tried to rework on it in order to make it clearer now (see page 13).

Technical corrections: ========== Abstract:

l.2 The present paper -> This paper Response: Corrected.

l.6 the real magnitude -> the magnitude Response: Corrected.

l.10 Remove "because radioactivity is a sensitive topic for the public and attracts a lot of attention" Response: We prefer to leave this argument present in the manuscript. This argument is true, as after the accident the official authorities stepped back in releasing measurements in public, in order to minimize the public concern of radioactivity and avoid terrifying the population.

l.17 give such kind of -> provide available Response: Corrected.

l.25 Please rephrase sentence "The results were of the present inversion were confirmed using an independent Eulerial model, for which deposition patterns were also improved when using the estimated posterior releases" to make meaning more clear Response: Corrected.

Section 2:

p.4 l.33 good estimations -> estimates Response: Corrected.

p.4 l.17 largest -> bulk of the Response: Corrected.

p.4 l.31 supposedly -> reportedly Response: Corrected.

Section 3:

p.5 l.20 bomb -> weapon Response: Corrected.

Section 4:

Rename section 4.1 to "Meteorological datasets results comparison" Response: We prefer to keep the title of the section as is, because it describes exactly what we did; we selected the proper meteorological dataset to proceed with the inversions. A title like "Meteorological datasets results comparison" seems less clear in our opinion.

p. 14 l.12: May 3rd, 4th and 5th -> May 3-5 (and similarly elsewhere...) Response: Corrected.

p. 14 l.14: Chapter 2? Response: Corrected (section).

p.15 l.8: less -> lower Response: Corrected.

p.16 l.12: is the main advantage of -> improvement by Response: Corrected.

p.18 l.23-27: No need to list all countries (and country codes) here. Please remove and reference for brevity. Response: Corrected.

p.19 l.7-9: Rephrase sentence: "Unfortunately, considering that 131I has a lifetime of only about 8 days, it was impossible to gather any observations of 131I deposition over Europe" -> "observations of 131I deposition over Europe are unavailable, due to the short half-life" Response: Corrected.

Figure 1: The uncertainty bands of the different priors are unintelligible. This Fig is not necessary and can be removed. Priors per radionuclide can be broken into individual figures and provided in a Supplement. Response: We had a long discussion about the presentation of our emissions. The majority of the coauthors believes that the prior emissions must be presented in the paper (and not in the Supplements). In this way,

the readers would be able to see right away how different the posterior emissions are, compared to the prior ones; thus, how drastic the change in the posterior emissions is, compared to the prior ones, using not only concentrations, but also deposition observations for the inversions. If there is no problem with the length and the number of Figures in the manuscript, we like to keep the Figure in the paper.

Figure 5: Why are only 3 emission heights levels plotted, and not all calculated? Response: The source-receptor relationship (see section 3.2) was calculated in 3-hourly intervals (8 per day) for 12 days of release (26 Apr 1986 at 00:00 to 8 May 1986 at 00:00) and at 6 layers (0 to 3000 m separated every 500 m). This gives 8x12x6=576 unknowns that the inversion needs to solve. If we separate between more vertical levels, then the unknowns increase dramatically and cannot be easily treated using our available computational resources. Furthermore, so far, all the previously published assessments for the source term from the Chernobyl accident reported a maximum altitude of emission of about 2.2 to 2.5 km (see relevant references in the manuscript), based on eyewitnesses. Therefore, we though it is challenging to define 1 additional layer, in case the observations drive the inversion into releases at higher altitudes. From what we saw in the obtained results, indeed emissions at 2.5-3 km occurred, although they were only 4%-13% of the total emissions (Table 1). Hence we expect emissions to be rather insignificant at higher altitudes.

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