Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of atmospheric dispersion model with improved deposition scheme and oceanic dispersion model" *by* G. Katata et al.

Anonymous Referee #4

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1   General remarks

This paper presents a very detailed analysis of the complex release and dispersion situation for the Fukushima reactor accident performed mainly by the Japan Atomic

Energy Agency JAEA. It provides both a better methodological background and an update of the preliminary source term of Chino et al. (2011) which has found widespread use. Therefore, this work is a useful addition to the body of literature on the meteorological aspects of the accident. However, there are a number of issues to be addressed for a revised version.

2   Major comments

1. The paper is very long and not easy to read, even though certain important aspects are still not sufficiently covered. I would recommend that the paper is revised in a way that it would be more systematic, focus on the most important aspects, and would defer minor aspects to the Supplement. For example, the discussion of the single phases on different days of the accident could be trimmed down, moving a systematic description to the Supplement. However, this description should then be really systematic, best in the form of a table with standard information for each phase, possibly also related figures.

The authors give now more insight into their method of source determination. This is really important as previous publications have not been very explicit on this topic. However, the presentation should still be improved, and be better placed in the context other similar work, as is detailed below.

2. The authors use both the terms "reverse estimation" and "inverse estimation". It seems that the latter is reserved for the part of the source reconstruction using concentration data in ocean water. The authors should explain what they mean by these terms and consider established technical terms. In applied mathematics and related sciences, the term "inverse problem" and derived from it "inverse

method / modelling / ..." is the standard language (however, as will be explained below, the method does not correspond to a formal inversion).

3. The authors explain that for the land data, they proceed as follows, with my questions in brackets:

   (a) *Divide the time axis into intervals* [Obviously, this is largely subjective. What is the role in this division of steadiness of meteorological conditions, steadiness of the plant state, steadiness and/or availability of measurements ?]

   (b) *Only a single measurement available for the respective release period: pair maximum of both measurement and model to determine source.* [Does it really occur often that only a single measurement is affected? This seems unlikely. Deposition data are always available, even though they were not used here. And how can one ascertain that only a single release phase impacts this measurement? What if this is not the case? What is the time and space window considered for matching the maxima? Even though this method should increase the robustness of the result, it could also be a major source of error if something is paired which does not match.]

   (c) *If multiple measurements are affected, average both measurements and model.* [Will again only the peaks be considered, or will whole time series over some interval be averaged? What is the justification of this approach – should one not rather average the resulting ratios?]

4. Usage of dose rates: is it true that only 5 nuclides are considered? Which fraction of the total dose rate can they explain? How does this fraction depend on time and distance?

5. The paper is quite obscure concerning how decay is considered, which is relevant for iodine and other short-lived nuclides. This pertains for example to Figure 2 and specifically 2c as well as the adopted I-131/Cs-137 ratio. It must be

clearly stated, for all the data (measured, modelled, release) whether they are decay-corrected (and to which time) or not. If not, how can the ratio be assumed constant over days? Is the ratio only used when the source estimate is based on dose rates, or throughout? I tried to analyse this nuclide ratio from the release data in the supplement and found that after ca. 8 days, it suddenly drops from 10 to 5. It may be just coincidental, but this looks like decay has been ignored for 8 days, then some I-131 measurements come in and the ratio drops corresponding to the decay. Soon after, the ratio jumps by a factor of almost 100 within very short time. Unless a convincing explanation is given for that, I don't think that these ratios and thus the iodine releases can be considered reliable, even though I admit that the comparison with the airborne survey is a good support for the source term. However, as also the deposition parameterisation has been strongly modified, and as the reconstructed source term has such a large temporal variability, this evidence is not totally conclusive. I am also surprised by the large scatter of the ratios presented in Figure 2, they need a proper explanation, including uncertainties of the measurements.

6. The releases tabulated in the supplement contain three physical species of iodine. It should be explained in the paper how they were derived, which measurements were available, and what the uncertainties are for this subdivision.

7. The explanation in Section 2.3 is difficult to follow. It should be more formalised, but with proper notation. It is not helpful how variable names Ci, Co, Mo are used. It would be better to to use a standard notation such as c for concentration, with a subscript such as, e.g., m and o for model and observation, and not to use the letter C for something which isn't concentration – better call the correction factor r or f or similar. Also, if the index j is occurring everywhere, it can just be dropped. Why is a log function used for averaging? Also, some variables are written upright, others in italics, etc.

8. As visible in the supplement, various assumptions for the source geometry are made for different release periods. Some explanation on that is needed in the the methods sections.

9. A discussion of the method for source estimation is required. It should include reference to less subjective, formal methods for solving inverse problems related to atmospheric dispersion which are available and partly have already been applied to the Fukushima accident. It should then be explain why the authors believe that their own method, which requires a lot of subjective decisions and manual intervention, is preferable. I do understand that in this situation, very complex both with respect to the source processes and the meteorological phenomena, with data of different type and degrees of quality, a selection guided by all the available knowledge can be very useful in obtaining a robust result, and agencies tasked with this emergency have certainly accumulated a lot of such knowledge. On the other hand, there is also the risk of mistakes in the process and of underusing available information through the lack of a comprehensive method. Personally, I would believe that it should also be possible to use a more comprehensive, formal inverse method together with such knowledge. In any case, the impression should be avoided that the approach used here would be the best or only one for dealing with unknown source terms in a nuclear emergency. It would be good if the authors would frankly explain their experiences and rationale for their approach, and to document their knowledge so that others can also use it (see my suggestion above, for a systematic coverage of the release in the Supplement).

10. The comparisons based on the regional-scale WMO calculations show, all in all, lower scores when the new source term is used compared to Terada 2012. Interestingly, NAME with ECMWF data performed best, even though ECMWF is said to have precipitation not well reproduced in this case. There is no discussion of these findings, however, they may indicate a kind of overfitting – or were WMO models tuned to the Terada 2012 source term?

11. Global HYSPLIT simulations and comparison with measurements: I consider this part a candidate for removal from this paper. Global simulations and comparisons with distant sites are not that useful for verifying the fine details of the source with which the present paper is mainly concerned with. There are some aspects of the results that would need to be addressed in more detail, such as the underprediction of particulate iodine and the lack of correlation for caesium. Note that the authors speculate that the correlation for iodine is caused only by the co-factor of age and associated decay – see my comments above on decay correction! I was also wondering why the time series plots are almost all clipped so that they don't show the arrival of the plume properly. Better work on a separate study than reporting preliminary results without proper explanation.

12. Discussion of source terms (Section 4.1): I don't feel so much convinced with these arguments. The increased wet scavenging might be too high, and estimates of dose rates at nearby monitoring stations would depend on effective source heights, which are not so well known and which appear to have been set to some predetermined values based on expert judgement only (the paper is not explicit on this). The emissions are partly attributed, for example, to wet venting of unit 3, "wet venting" or "dry venting" is also referred to in other places. Even though the emergency staff tried to conduct such operations, often it is also reported that "success is not clear" or that operations failed. Furthermore, it is still not well known which release paths to the environment were created during the course of the accident. Thus, one should not be certain that venting operations reported represent properly the release paths. All in all, results therefore may be not as robust as they are appear in the paper.

13. Supplement with release rates: when I open the xlsx file, I am told that a link to an external file exists (which of course is not present). In view of long-term archival, I don't thing xlsx is a good format to include the data – a simple text file could be more useful (including both might be an attractive option). Also, give the units for

all the values (it is not stated that releases are given as Bq/h). Do not mix text and data, or source geometry that cannot be read automatically, but provide a format allowing data to be easily ingested by programmes.

14. Measurement data: The paper refers to various sources of measurements, usually web resources. However, many are in Japanese, and usually in the form of PDF files. This kind of sources is not very suitable for use in further studies. I would like to encourage the authors to do their best to contribute to a collection of the relevant environmental data in an accessible, machine-readable format, and provide the most useful sources in their references.

3 Other comments

1. I think Eq. 2 is trivial enough to be skipped (and writing whole phrases as subscripts is not very appropriate).

2. Reference CTBTO (2011) is missing.

3. Figures 12 and 15 should match better (same order of variables shown, domain)

4. Figure 25: It would be better to separate this important overview into a Cs-137 and I-131 part.

5. Figure 17: It is much too small. Showing the Cs-137 results is sufficient, the others are very similar.

6. I found Figure S3 quite interesting and was wondering why this Figure was moved to the supplement. Concerning the way it is referenced in the text, this type of figure is not suitable to show that for the majority of monitoring sites the model was within a factor of 2 of the observation. There is a number of sites with nice

agreement, but also sites which don't agree and where the new version did not improve much. It would also be of interest to know whether the sites are a subset, or include available ones.

7. Figure 18: Not needed and unreadable. Just give a score such as FA5 as a small table.

8. Figure 19: Why is the colour scale totally different than in the observation plot (Fig. 12)? Also it is very difficult to find geographical features in both figures for comparison. Remove the province border lines and show a well-readable geographic grid, same in both figures. Restrict the area in this Figure to the one in Figs. 12 and 15.

9. Tables 6–8: This presentation is not sufficiently clear. All comparisons should be done with the same metrics and be shown in the same layout (a clear one!). My preference is FA2/FA5/FA10 (maybe not all of them are needed), correlation coefficent and bias. FAx is the best metric for these quasi-lognormal data. Bias should not be normalised by mean of obs and model (FB) – gives better score to overpredicting model. As all comparisons in one class have the same obs data, no need for normalisation. Or if you want, normalise with the mean observation only. Add units for all columns which contain dimensional data.

10. Many of the figures (also some tables) are to small to be legible when printed, only on the screen with 200-400% magnification they can we well read (but some are blurred at high resolution, even worse). Please make sure in the final version that all figures included are readable in print format.

11. Several captions of figures with model–observation comparisons lack information about the observation data set used.

12. I don't think having an appendix and a supplement is needed. I would suggest to move the description of the deposition schemes to the supplement.

13. Language proofreading would be a plus, especially as native English speakers are on the list of authors. (Just a single observation: don't use the word 'trend' for general temporal variation.)

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