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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An inverse modeling method to assess the source term of the Fukushima nuclear power plant accident using gamma dose rate observations" *by* O. Saunier et al.

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The paper introduces a method to derive a nuclear accident source term from gamma dose rate measurements, which is applied to the complicated Fukushima accident. The paper makes an important and practically very relevant contribution, using suitable a priori information, especially nuclide ratio intervals, and an appropriate variational optimisation technique for the inversion. Specifically, I appreciate the detailed comparison between on-site events and releases as reconstructed including a qualitative estimate

of the reliability of the inversion.

While the work is well done and the paper generally well written, there are still some aspects which could be improved.

Major comments

1. The abstract claims that none of the existing approaches uses dose rate measurements, and in the introduction it is stated that Astrup et al. (2004) used synthetic gamma dose rate data to improve the dose rate field. This is not correct. Astrup et al. used real dose rate data, as do Drews et al. (2004) and Duranova et al. (1999). Furthermore, there are several more works which used synthetic dose rate data (see our web site http://www.univie.ac.at/theoret-met/prepare/).

2. The authors use the term "reverse method" or "reverse approach" to designate the simple source estimation method of Chino et al. (2011) and others, whereas more elaborate methods are qualified as "inverse method". I think it is not useful to introduce this kind of terminology. "Reverse" and "inverse" don't really have a different meaning, the difference just being that "inverse" is a well-established scientific term while "reverse" may not have a specific meaning. I really ask the authors to refrain from further popularising this not very fortunate expression. It would be better to speak about "simple methods for source estimation".

3. The authors claim that, in contrast to others, their method does not make use of an a priori source term, both in the introduction and in Section 2.2. However, their assumption $\sigma_b = 0$ is an a priori, namely assuming that the source term is zero. Whereas other authors use the deviation from their nonzero a priori to stabilise the solution, Saunier et al. use the deviation from zero (i.e., the variance of the source vector). Of course, we can argue that the point here is that the method

works without having to construct a specific a priori source term. However, in principle, the other methods could as well use such a "trivial a priori" and giving the regularisation conditions sufficient weight, a solution will also be attainable. In my opinion, the point here is rather that the dose rate network is sufficiently dense and well-distributed to obtain a good solution without specific a priori. Also the statement "When no prior knowledge of the source term is used, it is still possible to solve the inverse problem but the number of measurements must be much larger than the number of unknown parameters" is not generally true – it depends on how strong the regularisation is. Regularisation has the same effect as decreasing the number of unknowns, because implicitly it reduces the resolution of the vector of unknowns. Similarly, the statement following Eq. 8, that the number of constraints (for the nuclide ratios) has to be at least equal to the number of radionuclides, is not sufficiently founded.

4. An important part of the method is the division into a step 2 which identifies potential release periods and a step 3 which then quantifies the releases during those periods. It is not clear why this step 2 must be introduced, or whether it is really beneficial. One might expect that the inversion would anyway only attribute nonzero source strength to the episodes that are identified in step 2. There is no directly visible reason why Eq. 5 should lead to different periods than applying Eq. 7 directly without previous restriction of the source time. The only obvious advantage is the reduction of the size of the problem, but this is not given as a motivation for the division into two steps.

5. What is the meaning of the statement "All of the noble gases emitted during the accident are grouped and are estimated as ^{133}Xe emissions." (Section 4.2.2, abstract and conclusions repeat this argument, should in case also be adapted.) If it is true, as said before, that Xe-133 is the only noble gas among the nuclides that contributes significantly to the dose rate, then one should not consider this as grouping and as attributing other nuclides to Xe-133. I am also wondering which

fraction of the total dose rate is caused by the selected list of 8 nuclides. Our own calculations (not yet published) showed that for achieving 98% of the total dose committed to people (external and inhalation), we would need about 20 nuclides, including Xe-135 and two Kr isotopes. One would assume that external dose only would show an even higher fraction to be attributed to noble gases and thus probably to isotopes other than Xe-133. If, however, this is not true, than the fact that other noble gas nuclides were excluded has to be introduced before the nuclide list, and it should be justified. Personally, I don't see why one would not want to include them, as their reactor inventory is known and they would thus not increase the effective number of unknowns.

6. The authors state on several occasions that they think that they overestimate the Xe release. I think that the arguments that lead to this conclusion are not sufficiently clear, and also I doubt that there is really an overestimation. It seems that the main argument why the authors believe in an overestimation is provided in Section 4.2.2, p. 15589/90. I don't understand why Te-132/I-132 should behave like a noble gas – after all, the decay of deposits of Te-132 (half-life ca. 3 d) should be clearly visible and the decay of I-132 (ca. 2 h) at least roughly visible from hourly dose rate data. Furthermore, effects of washout (which does not affect the noble gases) would also provide a means of distinguishing. I would rather think that the attribution of all noble gas isotopes as Xe-133 (see discussion above) would lead to an overestimation. Finally, the result of 12 EBq roughly agrees with the inventory of the damaged cores, and certainly isn't too high when the in-growth from I-133 is considered. The conversion between aerosol-borne and gaseous nuclides is, by the way, a potentially relevant influence factor not discussed in the paper.

7. Does the inversion method applied include the calculation of a posteriori uncertainties of the retrieved source term? If so, what is their magnitude and how variable is it?

8. At the end of the Conclusions, the authors discuss planned further improvements, among them using a nested approach for better resolving the steep gradients encountered. While this is quite appropriate, it would probably be good to touch upon this issue in the introduction section, as most other inversions rely on Lagrangian particle models which don't have this resolution problem. Furthermore, one could ask whether the smoothing effect of the Eulerian model (note that we are looking at point-to-point source-receptor relationships!) also has a regularising effect on the inversion, by broadening the sensitivity function in time.

9. It would be very useful if a supplement with the retrieved source term would be provided, similar to Stohl et al. (2012).

Minor comments

1. **Language:** While in general the paper is well written, there are numerous instances where grammar, spelling, or vocabulary is not proper English (probably influenced by the authors' mother tongue). It would be very desirable to have a language check done on the final manuscript. A few examples (not exhaustive!):

    - Page 15568, l. 24/25: *The result is that the model-measurement agreement for all of the monitoring locations is correct for 80 % of simulated dose rates that are within a factor of 2 of the observed values.* – This is not a proper sentence. A possible wording could be: It was found that for 80 % of the monitoring sites, simulated and observed dose rates agreed within a factor of 2.
    - Page 15569, l. 12: *For example, source term related . . .* – "the" is missing.
    - Page 15573, l. 21: *vector source term* – should be "source term vector" or "vector of source term elements".

    - ibid., l. 28: *humid deposition* – should be "wet deposition".
    - Page 15577, l. 19: *real accidental situation* – should be "real accident situation".
    - Page 15580, l. 8: *noble gases were rejected* – should be "noble gases were emitted" (or "released").

2. Page 15572, l. 12: The ambient dose rate is not *the linear combination of the contributions of all gamma-emitting radionuclides*, but their sum. (OK, a sum is also a linear combination, but a trivial one and it is thus misleading to use the term linear combination.)

3. Figure 1 and respective paragraph in Section 2.1: The figure caption is insufficient. The numbers 1, 2, 3 must be explained at least briefly in the caption or it has to be said that they are explained in the text. Reference belongs to caption and not to text body. The abbreviation *ex.* used in the text is not clear. It should be said that this is an idealised situation, for example assuming no wet deposition and that the plume passes over the receptor only once.

4. Page 15574, l. 5: Explain what is E. Explicitly state that off-diagonal elements are not considered (and why).

5. ibid., l. 18: Quantify which fraction of the dose rate is explained by 10 radionuclides, and also under which conditions.

6. ibid., l. 20: (The relevant radionuclides) *can be identified by . . . or by using the core inventory of the damaged facility.* – Core inventory plus release fractions for each nuclide group! Same issue after Eq. 4.

7. Eq. 6: An erroneous apostrophe appears after the last line.

8. Eq. 7: I think it is necessary to include error covariances also in the cost function contribution due to the nuclide ratio, at least in the form of a single weighting

factor. Otherwise, you are assigning implicitly a certain weight to this term in comparison to the other terms (probably, due to the shape of the function r, results aren't very sensitive to this, but that is not immediately visible, and also formally it is unsatisfactory).

9. Page 15578, l. 25: Is the output instantaneous or averaged over 1 h?

10. ibid.: Is a model top of 3400 m sufficient? Did you verify that?

11. Page 15579, l. 4: Give references and/or details for the ldX, C3X and ConsX models. How much time does it take to carry out these 381 simulations, and on which computer platform?

12. Page 15580, l. 6: I would not say that noble gases are "highly volatile", they are just gaseous (would condense only close to absolute zero). In the reactor or the environment, they never occur in solid or liquid form, so it is not so appropriate to discuss them in terms of volatility.

13. ibid., l. 9: I would think that I-132 and Te-132 are not exactly in secular equilibrium, at least not in the first hours after the shut-down. Maybe it should be made more clear how well this assumption is fulfilled at the time for which the first release is anticipated.

14. ibid., l. 15: It is not correct to say that Cs-137 has no impact if it is decaying too slowly to observe this process. Nevertheless, it is a major contributor to the dose.

15. ibid., l. 18: Who showed that the ratio of Cs-134/Cs-137 is constant over Japan (reference)? Constant within which bounds? Shouldn't there be an upper and lower threshold for the ratio in Eq. 10?

16. Eq. 11: It would be of interest to include a table which translates these nuclide ratios used as bounds into ratios of the release fractions of these nuclides, under the assumption of a known and homogeneous inventory.

17. Page 15581, l. 19: I would be more careful with respect to stating that λ_1 can be rigorously determined. That depends very much on the condition of the matrix, and even if a precise condition is formulated, this does not necessarily mean much in practice. Only if errors for all the terms are well known, the solution is truly rigorous.

18. Page 15582, l. 1: Please explain briefly how the L-BFGS-B algorithm works, whether it is publicly available in coded form, and whether additional constraints such as positive definiteness are applied.

19. Section 3.3 in general: Is there a threshold value for σ_1 being applied? How continuous/intermittent is the resulting possible release time?

20. Page 15583, l. 10: It would interesting to know which percentage of the emissions calculated by Stohl et al. (2012) occurred during periods which did not pass over Japanese dose rate monitoring sites and thus cannot be reconstructed in the framework of the present manuscript. Doing this would allow a more meaningful comparison between these two emission estimates.

21. Section 4.2: Maybe this section would better be called "Comparison with observations", as an agreement of simulated and observed dosed rates and/or activity concentration is a necessary but not a sufficient condition for the agreement of the calculated with the true source term.

22. Page 15585, Eq. 15: Note that this formulation favours overprediction as compared to underprediction.

23. Table 4: I am wondering how it comes that the biases are always positive. It is quite surprising and needs to be discussed.

24. It seems that no distinction has been made between gaseous and aerosol-bound iodine in the dispersion calculations. This should be mentioned explicitly. Could

this be responsible (at least partly) for the lower performance with respect to iodine as reported in Table 4?

25. Page 15589, l. 4: *The simulations accurately reproduce the signal's temporal evolution* – The agreement is good, but "accurately" is exaggerated.

26. Page 1550, l. 6: *The comparison is more difficult in the case of the second episode* – it is not more difficult, rather it results in more substantial deviations.

27. Various places: *fac2, fac5, fac10* used like words. Please don't use code variable names like words, better write that out, or introduce a proper symbols such as, for example, F_2, which should then be introduced at the place where it first appears.

28. Page 15591, l. 10: *The surface activity increased* – did you mean that the deposition rate increased? That is not the same. Also below, *deposition activities* – does that refer to deposition rates?

29. ibid., l. 25: *These measurements, some of which were taken after the accident,* – I thought that all of them were taken after the accident?

30. Page 15592, l. 1: I have the impression that while the pattern of the heavy contamination towards NW is fairly reproduced, but its intensity still is too low (how much?)

31. ibid., l. 13. How can it be seen that differences are due to inaccurate meteorological data?

32. Table 3: Does *Unit 2 – Unit 3* mean unit 2 *and* unit 3, or unit 2 *or* unit 3 (uncertain from where)?

33. Figures 1 and 2: It is not mentioned whether time is UTC or JST.

34. Fig. 2: Caption is too short. For example, say where the measurement is taken. Use smaller symbols for the measurements. Consider using a log scale.

35. Fig. 3. Include a demarcation of the different zones in the figure, and make the NPP site more visible.

36. Figure 4: In order to increase the readability, don't use dashed lines (maybe the vertical lines could be plotted in gray). Ticks to the outside on the time axis, and adding minor ticks for subdivision would also be useful. Indicate (somewhere – not necessarily in the Fig. 4 caption) the time shift between UTC and JST.

37. Figures 6–8: The dots are not very well readable, and it is not clear whether the width of the dots corresponds to the time interval to which it refers, or otherwise whether dots are centred or at the end of the respective time intervals. Probably horizontal lines (dashes of appropriate length) could be more suitable.

38. Figures 2, 4–8: All these figures use linear scales of the ordinate. There are of course pros and cons of linear and logarithmic scales, but I am wondering why all figures are with linear scales. I think at least Fig. 7 would gain in readability if a log scale were used.

39. Figure 9: The km-annotation on the radii and the legend to the colour bar are too small and hard to decipher. It is not clear to me whether all subfigures use the same scale, and it seems that they don't show the same area. Also, the observation map shows the outline of the provinces while this is missing in the other two maps. All of the maps lack a geographical grid. It would be best to add that and to re-plot the observation map with the same software (as far as I know, data are public). A reference for the observation map is missing.

40. It would be useful to add in a prominent location (e.g. abstract and introduction) the scale on which the method works, pointing out that it is not targeted at on-

site dose rate measurements. Please introduce the state of the art in a more complete fashion.

References

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