

Interactive comment on “Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition” by A. Stohl et al.

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Received and published: 23 November 2011

Dear Horst-Michael Prasser,

We would like to thank you for your constructive comments on our paper. We particularly appreciate the suggestion to mark the time of the earthquake and tsunami (on this time scale, the two events can hardly be separated graphically) in Fig. 4, which we will try to accommodate in the revised version of our paper.

Regarding our finding of an early start of Xe-133 emissions, we would like to point out

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the following to clarify the uncertainties with respect to timing errors:

1. The time resolution of the emissions obtained from our inversion is 3 hours. Therefore, it is impossible to judge from our results whether emissions took place during the 45 min time interval between the earthquake and the tsunami arrival with the associated station blackout, as it was erroneously said in some of the media reports on our study. Notice the word “possibly” in our statement in the abstract “There is strong evidence that the first strong Xe-133 release started very early, possibly immediately after the earthquake and the emergency shutdown on 11 March at 06:00 UTC.” We will rephrase this to make clear that essentially we mean that we have strong evidence that the emissions started before the first venting event.
2. The time resolution of most of the Xe-133 measurements we use is 12 h, and the time resolution of reconstructed emissions is 3 h. One might wonder whether or how this is possible. The answer is that each measurement, regardless of its duration, has a well-defined time period (periods) during which it can be affected by the sources. If no concentration is detected, this is an indication of zero emissions during the corresponding time period of the source. Overlay of many measurements, as done by the inversion algorithm, thus allows in principle a time resolution of the source that is better than that of the receptors.
3. We would also like to draw attention to our statement on Page 28345 (line 19) that “Individual 3-hourly emission fluxes are more uncertain” than the 20% we have estimated for the total emissions and the explicit a posteriori uncertainties as contained in the supplemental material.
4. In order to clarify our results for the initial phase of the accidents, we have plotted the a priori and a posteriori emissions together with two different estimates of the lower end of the confidence interval (with 1 and 2 sigma, where sigma is the

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- a posteriori uncertainty) with high time resolution and for the sum of the three vertical emission levels (Fig. 1 of this response). From this figure, it becomes evident that
- a. Any emissions before March 11, 03 UTC are not significantly different from zero even though the algorithm gives a value of about 8 TB/3h.
 - b. With the next 3 h interval (the earthquake occurred towards the end of this interval), they rise by a factor of 50, remaining roughly constant for 9 h, and they become highly significant. In these 9 h between 03 and 12 UTC of March 11, before the next jump in the emissions, a total emission of about 1 PBq (or, taking the lowest estimate, maybe 0.2 PBq) is obtained. We do not think that this amount is unrealistic, taking into account also the fact that after the loss of the AC power with the tsunami, there was no cooling to unit 1 core at all. The too early onset of the increase might indicate an error in the implicit transport velocity between Japan and the west coast of North America of about 3 h or about 3% of the total transport time of just less than 5 d. This is quite possible.
5. Reconstructed emission rates suddenly jump by more than 2 orders of magnitude at March 11, 12 UTC, with little uncertainty according to the inversion algorithm, and being already close to the maximum release rate of the first emission peak. This time coincides with the time given by the MELCOR calculations as presented in the Japanese Government Report for the failure of the primary containment vessel. While the a priori would gradually start to increase the release rate at this time, the inversion shows a sudden increase and then a further slow increase until the time of the deliberate venting. This is an interesting finding, and it is compatible with other evidence as given in the Report. According to INPO (2011, p. 16), 1.2 mSv/h dose rate occurred outside the reactor building personnel air lock door at 14 UTC. Such a dose rate appears to be associated with a major release, thus we conclude that the onset of the major release in the interval 12–15 UTC obtained by the inversion is confirmed by observations. This enhances

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- also the confidence for the inversion results in the hours before.
- The inversion result that substantial emissions have occurred before the first venting operation is not only found for the first peak, associated with unit 1, rather, this feature appears to repeat itself also at units 2 and 3, probably indicating leaks to the containment before its failure, for example through damaged secondary piping.
 - Notice that Xe-133 measurements started to rise very early at the North American west coast, e.g., at Richland on 16 March between 1:52-13:52 UTC (Fig. 8). The model broadly reproduces the arrival time, but tends to underestimate the very first enhanced Xe-133 concentrations, even with the higher and earlier Xe-133 emissions. Thus, to reproduce this early concentration increase would, with given meteorological conditions, in principle require an even earlier or faster onset of emissions.
 - It is difficult to quantify meteorological uncertainty and, admittedly, this introduces some timing uncertainty. The usual way to at least roughly quantify this is to use meteorological data sets from different sources. We have used data from two meteorological centers and we obtain a very similar early emission start both with GFS and ECMWF meteorological data. This agreement, together with the fact that we successfully simulate the arrival time of the plume at the North American west coast, lends confidence into our estimate although, as discussed above, timing errors on the order of one to three source time intervals (3-9 h) have to be considered possible. Certainly, greater timing accuracy could be achieved if measurement data closer to the accident site were available. To clarify the emission onset, however, Japanese data would not have helped since this part of the plume was transported out into the Pacific Ocean.
 - Finally, what is the evidence that emissions did not start before the first venting? Increased radiation was observed in the turbine hall at 11 March on 14 UTC.

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At 18:45 UTC on 11 March, workers trying to measure the radiation dose in the reactor building opened an air lock and found a white “cloud” coming out, causing them to immediately shut the air lock again (see <http://spectrum.ieee.org/energy/nuclear/24-hours-at-fukushima/0>). The Report (p. IV-34) says that on March 12 (without clear time, but obviously before the venting) “TEPCO began preparations for PCV venting because the PCV pressure was high, but the work ran into trouble because the radiation level in the reactor building was already high”. In Table IV-5-1, p. IV-50, it is stated for 11/3 18:18 (9:18 UTC) “Opening operation was performed on IC(A) system supplying piping isolation valve MO-2A and return piping isolation valve MO-3A / steam generation was observed”. 7 min later MO-3A is reported closed. At 21:30 JST (12:30 UTC) again it is reported “IC 3A valve was opened / steam generation was observed.” These operations alone could have released a considerable amount of noble gases, and even minor leaks of the containment would have released more and probably earlier.

More or less the same arguments apply to the drop of the Cs-137 emissions on 20 March. Again, we cannot determine the exact hour of the drop. However, in this case Japanese data were available to constrain the emissions, and we see consistency in our emission estimates when using Japanese deposition data only, Japanese concentration data only, and concentration data from outside Japan only (Fig. 6). As can be seen in Fig. 6, the exact time of the emission drop varies between the different inversion experiments but all experiments do suggest such a drop.

From our study we cannot distinguish between emissions from different reactor units. Our suggestion that emissions from the spent-fuel pool of unit 4 are likely to be responsible comes from the temporal coincidence of the decrease of emissions with the spraying in combination with the fact that this pool had the highest loading. Other explanations for the continued emissions for the period 16-19 March may be possible. It is beyond the scope of this paper to explore these possibilities.

Regarding your argument that there is no visible damage to the fuel elements in the spent fuel pool of unit 4, it has also been suggested that the photographs published by TEPCO provide no proof that no emissions have occurred (Christoph Müller, personal communication; see <http://www.tec-sim.de/images/stories/spf-fa3.pdf>, pages 57-60).

Kind regards,

The authors

Reference:

INPO (2011): Special Report on the Nuclear Accident at the Fukushima Dai-ichi Nuclear Power Station. Special Report INPO 11-005, November 2011, http://www.nei.org/filefolder/11_005_Special_Report_on_Fukushima_Daiichi_MASTER_11_08_11_1.pdf, (last accessed 23 November 2011).

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 28319, 2011.

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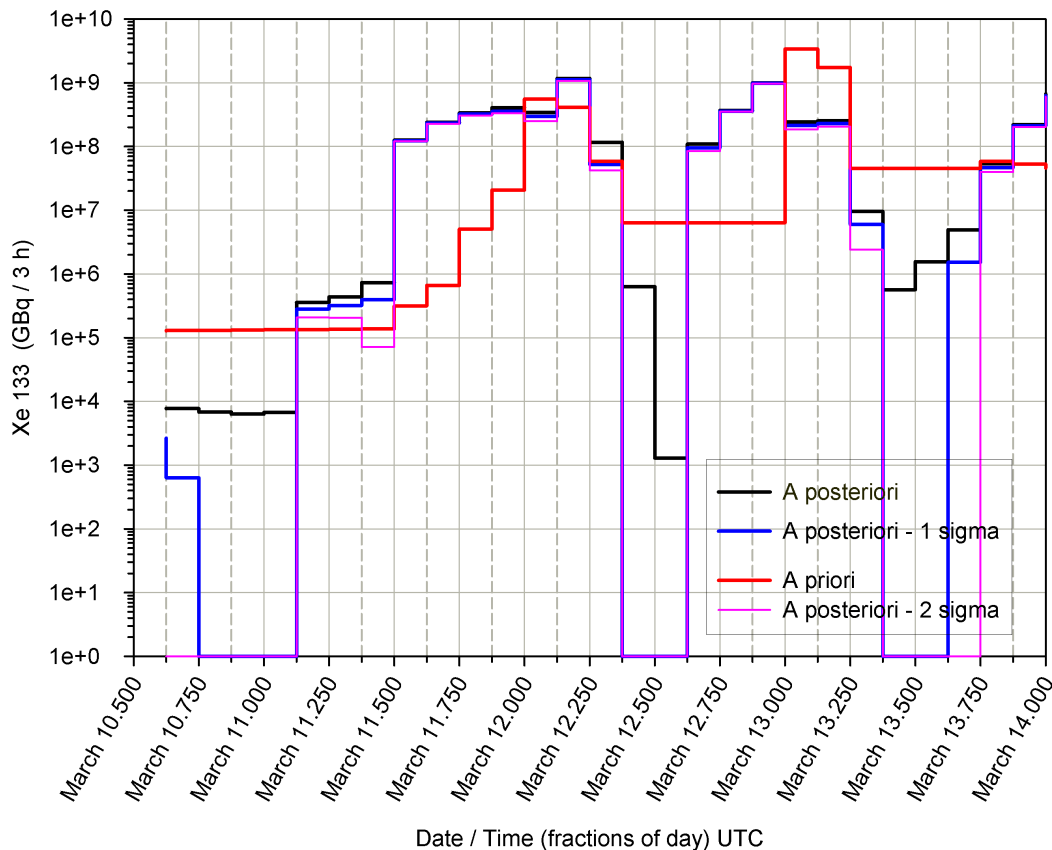


Fig. 1. A detailed view of a priori and a posteriori emissions, including also 1- and 2-sigma lower limits of the a posteriori emissions, for the first three days of the accident.