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Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #1

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The manuscript determine the emissions of Xe-133 and Cs-137 released into the atmosphere from the Fukushima Dai-ichi nuclear power plant (FD-NPP) in Japan by the inverse modeling, which combined the a priori emissions with the atmospheric transport model, FLEXPART, and measurement data from Japan, North America, and other regions. Also, the manuscript presents the atmospheric dispersion and deposition of two isotopes in Japan as well as in the hemispheric scale. At the present time, the multi-scale and multi-media environmental pollution caused by the massive release of radioactivity to the atmosphere from the FD-NPP is very severe natural and social issues, while the available information related to the emissions and pollution is very limited so far. In this situation, the author's work brings very valuable and timely information to the international society. Especially, the manuscript presents the following findings: (1) the first strong Xe-133 release started very early, possibly immediately after the earthquake and the emergency shutdown; (2) the Xe-133 inventory of reactor units 1–3 was set free into the atmosphere between 11 and 15 March 2011; (3) the Cs-137 emissions were estimated to a total emission of 35.8 (23.3–50.1) PBq; (4) the early emissions around the time when the first explosion occurred in unit 1 were estimated by the inversion model and were underestimated by the Japanese authorities; and (5) the Cs-137 emissions were not only coming from the damaged units 1-3, but also from the spent-fuel pool of unit 4 because the emissions suddenly dropped when spraying of water on the spent-fuel pool of unit 4 started. However, the author's work is fundamentally based on the transport model of global scale, which has large uncertainty. Hence the conclusions should be discussed carefully and to be analyzed from the multi-angle scientific aspects. The amounts of emissions and their temporal variations are most important information for the estimation of environmental damage due to the FD-NPP accidents. These results give the large impacts to the international society and will affect to the current works in progress and future planning for countermeasures for the reduction of radioactivity in Japan. Therefore, it is very important to draw a conclusion by careful discussion and consideration from objective and scientific evidences. The reviewer recommends publishing this paper with major revisions in response to the following questions and comments.

< Major comments >

1. Limitation of global model: Does the FLEXPART for the global scale used in this study have a capability of analysis for high resolution results, such as apportionment to the vertical layer below 1000 m and the hourly temporal variation of emissions? Generally, it is difficult to simulate the 3-D transport and temporal variation of radionuclides emitted from the point sources over the complex topography around the FD-NPP even

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if the regional model is used. Also, the global scale transport model has a large uncertainty in the simulation of long range transport. Thus, the authors would have required a more careful analysis and discussion about the following findings under consideration of high uncertainties of the author's global model: (1) The discussion for vertical emission distribution (page 28341, lines 20-27); (2) "The start of a posteriori emissions at 06:00UTC on 11 March, coinciding with the time of the earthquake, could be due to a noble gas release as a consequence of the emergency shutdown of the reactors which had occurred at this time, possibly enhanced by structural damage from the earthquake." (page 28340, lines 23-26); (3) "we find two separate maxima within a 15-h period" (page 28343, lines 3-4); and (4) "Spraying of water on the pool started on 19 March at 23:21UTC and at that time our a posteriori emissions decrease by orders of magnitude." (page 28343, lines 9-11).

These findings are just speculations based on the model with large uncertainty and need to be confirmed by evidences based on measurement data analysis, regional transport modeling, and nuclear reactor analysis.

2. Model results in Japan: In this paper, FLEXPART was driven with three-hours operational meteorological data from ECMWF (resolution of $0.18^{\circ}\times0.18^{\circ}$in Japan) and the GFS data (resolution of $0.5^{\circ}\times0.5^{\circ}$). As mentioned by the authors (lines 5-8 of page 28334), these resolution is too coarse to resolve the complex topography around the FD-NPP. As a result, the modeled result in section 4.4.1 is quite different with the measured result in precipitation as well as Cs-137 deposition. There are some substantial mistakes in the interpretation on the dispersion and deposition of Cs-137 in Japan. As a Japanese researcher joins as a coauthor, the authors will be able to access and use the measured data sources (such as precipitation data and deposited Cs-137 map) in Japan. The authors need to evaluate and reanalyze the modeled results in Japan based on the observation data in Japan.

(1) Reproducibility for precipitation: The author's model can't catch the measured precipitation in many cases and hence it may cause some problems in the modeled Cs-

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137. The measured precipitation data and the related data in the following web sites (in Japanese) were used in this comment.

Surface precipitation: http://www.data.jma.go.jp/obd/stats/etrn/index.php

Radar AMeDAS(Automated Meteorological Data Acquisition System)(The data are based on the hourly radar gauged precipitation data calibrated by ground gauged precipitation): http://agora.ex.nii.ac.jp/digital-typhoon/archive/radar_amedas/

Precipitation radar echo: http://agora.ex.nii.ac.jp/digital-typhoon/archive/radar/intensity/

(Page 28351 lines 4-25) During 12:00–15:00UTC on 14 March, the precipitation wasn't detected in the wide area of land around the FD-NPP in the data of surface precipitation, Radar Amedas, and precipitation radar echo. The frontal precipitation pass through the ocean of south of FD-NPP. As a result, the amount of Cs-137 deposited in the land of Japan on 14 March was much lower than the estimated value by the author's model.

(Page 28352 lines 4–9) The horizontal distribution of simulated precipitation shown in Fig.15 (lower panel) is quite different with the measured precipitation. According to the observation, it did not rain in/around the Tokyo Metropolitan area, though the rain (or snow) was observed in the mountainous areas around the Kanto plain (The Tokyo Metropolitan Area is located in the south of the plain). On the other hand, the author's model shows the precipitation in the wide area of the Kanto plain. Hence the modeled Cs-137 deposition is different with the real deposition in the spatial distribution and it is highly possibility that the model overestimates the total amount of Cs-137 fall out in Japan. Probably, the simulated results based on the regional model (see maps by Morino et al., 2011; http://www.nies.go.jp/shinsai/index.html#title04) may be useful for reference.

(Figs. 16 and 19) The authors demonstrate the regional-scale maps not only in Fig.

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15 but also in Figs. 16 and 19. However, the complex terrain around FD-NPP are not taken account in these maps, in spite that the authors recognize the complexity to the meteorological fields (see lines 5-8 of page 28334). The authors should be careful in demonstrating the regional maps based on the coarse-resolution model.

(2) Fig. 19 (upper panel): The map of total deposition of Cs-137 until 20 April 00:00UTC for Japan is quite different with the map based on the aircraft measurement, recently released by MEXT (Ministry of Education, Culture, Sports, Science and Technology) of Japan (http://radioactivity.mext.go.jp/ja/1910/2011/11/1910_1125_2.pdf). It is strongly recommended that the authors compare the modeled map with the measured map.

(3) Page 28352 line 19 – page 28353 line 7: The a posteriori modeled Cs-137 surface concentrations overestimate the concentrations at Japanese stations except Takasaki (which is influenced by the contamination). Additionally, the model overestimates the observed deposition at Tokai-mura. The authors need to explain why the model overestimates the surface concentration as well as deposition in Japanese stations. On the other hand, the model underestimates the surface concentration data from outside Japan. What is the main reason of these contradictory results? A speculated possibility is that the model tends to overestimate the deposition near the FD-NPP and hence the a posteriori emissions may overestimate for agreement with the measurement data from outside Japan. Table 6 seems to demonstrate this possibility.

3. Section 3.3: The description of inversion algorithm in the section 3.3 is insufficient. At least, the authors should add the explanation for cost function, temporal smoothing condition, and the assigned value for observation error and background error.

4. Early release of Xe-133: It is important to analysis after due consideration of the large uncertainty in the release time estimated by the FLEXPART used in the manuscript. An important problem in the author's result has been pointed out by H.-M. Prasser (C11484-C11486). It is recommended that the authors demonstrate in the section 4.2 or 4.3 whether the earlier release of Xe-133 can lead to the improvement

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of model consistency with the observed Xe-133 concentrations.

5. Cs-137 release from degraded fuel in the spent-fuel pool of Unit 4: The author's insistence, "these high emissions are related to the degraded fuel in the spent-fuel pool of unit 4, and this result would also confirm that the spraying was an effective countermeasure at least in this case." is a just speculation based on the inversion with high uncertainty. An evidence needs for being a scientific finding; on the contrary, the following facts based on the measurements are inconsistent with author's result.

(1) The environmental dose rate* measured in the FD-NPP site didn't decease around 23:21 UTC on 19 March. The radiation measurement point (Jimusho-kita) was located at the northwest of units 1-4, about 500 m from there and the surface wind was weak (1-2 m/s). If the author's finding is correct, the environmental dose rate should decrease. * http://www.tepco.co.jp/cc/press/betu11_j/images/110528d.pdf

(2) According to the measurement data* at the Japan Chemical Analysis Center located in the Tokyo Metropolitan area, the environmental dose rate of many kinds of radionuclides (including Cs-137) suddenly increased around 21:00 UTC on 20 March (06:00 JST on 21 March). The similar increasing was detected at other sites in the Kanto plane. The Cs- 137 increasing means that the Cs-137 released from FD-NPP at around 13:00 UTC (22:00 JST) on 20 March were transported to the south of FD-NPP due to the north wind, followed by the wet deposition due to the strong precipitation. It is considered that this episode formed the "hot spots" in the Tokyo Metropolitan area. If the Cs-137 release between 00:00 UTC on 20 March and 00:00 UTC on 21 March (09:00 JST on 20 March and 09:00 JST on 21 March) decreased by below 0.1 GBq s-1 due to the water spraying in unit 4 (see the upper panel of Fig. 5), there would not be the "hot spots" episode in the Tokyo Metropolitan area. *http://www.jcac.or.jp/lib/senryo_lib/nodo.pdf (in Japanese)

< Individual comments >

1) Page 28328, line 5: The "caesium" should be "Cs-137".

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2) Page 28332, lines 2-5: Are there any proofs or references to support some problems in the data from the other Japanese stations? The authors need to add an explanation. Also, the "Similar features" in line 2 is an ambiguous expression and should be changed to be more clear.

3) Page 28333, lines 10-13: Are there any proofs or references to support the author's assumption for the uncertainty of measurements?

4) Page 28333, line 12: The unit of "2 Bq m-2" should be checked.

5) Section 3.2: An explanation for the deposition modeling for Xe-133 needs to be added.

6) Section 4.2.3: The uncertainty in absolute amount as well as temporal variation of a priori emissions is very large. Thus, It is recommended that the authors conduct another sensitivity tests for the temporal variation of the emissions based on the Chino et al. (2011) and others.

7) Section 4.3: For the verification of total amount of a posteriori emissions, it is suggested that the authors make a comparison between modeled and measured average concentrations and total depositions during the targeted period.

8) Page 28353, line 24: According to the Fig. 16 (middle panel), the FD-NPP plume didn't reach to "Osaka", which is located in the western edge of the map. The location of Osaka should be checked.

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