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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling the global atmospheric transport and deposition of radionuclides from the Fukushima Dai-ichi nuclear accident" *by* T. Christoudias and J. Lelieveld

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Dear Editor,

We would like to thank the referees for their careful reading of our manuscript, "Modelling the global atmospheric transport and deposition of radionuclides from the Fukushima Dai-ichi nuclear accident".

We are most grateful for the comments, constructive criticism and very useful suggestions received on how to improve the paper.

Please find attached our detailed answers to the questions (indicated by ++) and a new

version of the paper, which we hope satisfactorily address the points raised during the discussion.

Anonymous Referee #1:

The paper provides new knowledge and inovative results. Therefore, it would deserve publication in the journal ACP. Before that, there are several corrections and/or clarifications that are essential to be done by the authors. After that the manuscript should reconsidered.

The major comments on the manuscript are the following:

-Page 2 - Session 2.1: "These factors and a number of other assumptions ... of five.": The paragraph starting from the previous sentence needs more work in order to be more comprehensible. For example, do you report the factor of five based on this work or you just explain the paper of Chino et al. (2011)? Please explain.

++ The factor of five is based on the uncertainty range indicated by Chino et al. (2011). It is now clarified in the text.

-Page 3 - Session 3.1: "Only 20-50% of ... unattenuated." Please provide evidence for the fraction of Iodine being scavenged by filter papers (1-2 publications) in order you to prevent speculative characterisation of your statement.

++ This portion of the text was reworked and a reference to the "Detection of radionuclides emitted during the Fukushima nuclear accident with the CTBT radionuclide network" by Stoehlker et al. (2011) was added.

-Page 3 - Session 3.1: "The highest concentration of Xe ... to be accurate." Please provide more analysis here. What do you mean by dynamic range and why detections larger that 100 Bq m-3 are inaccurate? It is not clear.

++ We added a reference on the "Detection of radionuclides emitted during the Fukushima nuclear accident with the CTBT radionuclide network" by Stoehlker et al.

(2011) with more information on the detector operational characteristics.

-Page 3 - Session 3.1: "It is also known ... after the accident." Please provide references or other evidence abou that station's condition. Please explain what you mean by dead time. You need to do the same for the next sentence: "The Japanese particulate ... 2011." Also, for the stations JPP38 and USP71. The details you provide are not known and it would be useful to have a reference that certifies them.

++ All relevant details are found in the added reference to Stoehlker et al. (2011).

-Page 4 - Session 3.1: "The greater extend to which deposition ... transport." Please explain the sentence in order to be more comprehensive.

++ We added explanatory remarks: "Removal processes take place on a sub-grid scale and are therefore parameterized and thus less explicitly resolved by the model than atmospheric dynamical and transport processes."

-Page 4 - Session 3.1: "Furthermore, ... Chino et al. (2011)." I recommend you to do the same as in the 2nd comment.

++ This portion of the text was reworked and references added as in the 2nd comment.

-Page 5 - Session 3.2: "We estimate that the land area ... 46 million people". In this part it would be very important to work on a better mapping. For example, you mention several cities (Sendai, Yokohama, Chiba, Tokyo), which are not shown in the figures of deposition as they should. Please change.

++ We removed references to specific cities that are not shown on the map.

-Page 5 - Session 3.3: LAST PARAGRAPH: You calculate the 50 year ground deposition doses for Cs-137 and I-131. Normally, your main assumption in the calculations would be a stable environment that does not change due to vertical migration of radionuclides or other processes (e.g. washout, runoff etc...). And I agree with you that it is more or less an accurate estimate for cesium, which has a 30.2 years halflife. How-

ever, how do you account for the decay of I-131 in the formula you used for the dose calculations? According to my estimations, given that the halflife of I-131 is 8 days, you may have overestimated your doses up to 50%. Please explain or remove doses based on Cs and I.

++ The method for the calulation of 50-year integrated effective doses from ground deposition, including consideration of resuspension, weathering and ground roughness can be found in INES 2009, Appendix I). The integrated dose conversion factors, per unit ground deposition of each radionuclide, include the external dose and committed dose from inhalation (resuspension) resulting from remaining on contaminated ground for lifetime (50 years) (IAEA-TECDOC-1162).

-Fig.5, Fig.6 and Fig.7: Please re-create these figures in order the units to be in kBq/m2 since these depositional units you mention in the manuscript. This is crucial for the coherence of your manuscript.

++ The figures were re-created with units in kBq.

Anonymous Referee #2

The manuscript presents the global atmospheric transport and deposition of Xe-133, I-131 and Cs-137 released into the atmosphere from the Fukushima Dai-ichi nuclear power plant (FD-NPP) in Japan by the global transport model. Also, the manuscript discusses the total deposition of I-131 and Cs-137 in the Western Pacific and Japan. 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. However, the author's work is fundamentally based on the transport model of global scale, which has large uncertainty inthe regional scale. Hence the conclusions should be discussed carefully based on the observed data especially in Japan. In particular, the results related to the Japanese

situation give the large impacts to the Japanese 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. Model results in Japan: The horizontal resolution of the author's global model (T255 resolution) is approximately 0.5 by 0.5 degrees. The resolution is too coarse to resolve the complex topography around the FD-NPP. As a result, the modeled spatial distribution of deposition of the sum of Cs-137 and I-131 may be different from the actual situation. The authors need to evaluate the modeled results in Japan based on the observation data. For example, the deposition map of Cs-137 based on the aircraft measurement released by MEXT (Ministry of Education, Culture, Sports, Science and Technology) of Japan (http://radioactivity.mext.go.jp/en/contents/4000/3179/24/1270_1216.pdf) is now available. It is strongly recommended that the authors compare the modeled deposition with the measurements (e.g. http://radioactivity.mext.go.jp/en/). And then, the section 3.2 (4-13 lines of page 24542) and section 3.3 should be revised.

++ Section 3.3 was substiantially revised and Figure 8 adjusted to highlight the model grid resolution. Since the Xe-133 simulations have shown that the transport is modeled accurately, there is no reason to assume that transport over Japan would not be adequately simulated. Nevertheless, we agree that the 50km resolution is too coarse to simulate the conditions directly around the nuclear power plant, in particular the consequences of complex terrain for deposition processes. This is mentioned in the text.

2. Chemical characteristics and wet deposition of I-131: Are there any proofs or references of "iodine largely remains in the gas phase" (Page 24533, lines 2-3) and "I-131 in not removed by wet deposition"(Page 24540, line 9)? In the previous studies, it is

usually assumed that iodine is either bound to particle or in gaseous phase (Sportisse, 2007; Kristiansen et al., 2012). Actually, the measurements at the sites around FD-NPP after the accident shows that the I-131 consists of gas and particulate phase though the gaseous fraction has a big temporal and spatial variation (private communication with Japanese scientists). Additionally, for the wet deposition of I-131, the deposition rate for gaseous I-131 is reported in the previous study (Sportisse, 2007). If the wet depositions of particulate I-131 are considered in the global simulation, the author's modeled results may change drastically. (Ref.) Sportisse: A review of parameterizations for modelling dry deposition and scav- enging of radionuclides, Atmospheric Environment, 41, 2683-2698, 2007. Kristiansen et al.: Atmospheric removal times of the aerosol-bound radionuclides 137Cs and 131I during the months after the Fukushima Dai-ichi nuclear power plant accident – a con- straint for air quality and climate models, ACPD, 12, 12331-12356, 2012.

++ We added considerations, discussion and references on the characteristics and wet deposition of particulate I-131 (Stoehlker et al. (2011), Hoeve and Jacobson (2012)). A sensitivity test with a 1 to 4 particulate to gaseous I-131 ratio was performed and was added to the paper.

< Individual comments > 1) Page 24532, line 24: "Stohl et al., 2012" and "Chino et al., 2011" should be reversed in order.

++ Reversed order of citations.

2) Introduction As a previous work in the global simulation of radionuclides from the FD-NPP, the following reference which is a first publication in the global simulation should be added. (Ref.) Takemura et al.: A Numerical Simulation of Global Transport of Atmospheric Particles Emitted from the Fukushima Daiichi Nuclear Power Plant, SOLA, 7, 101-104, 2011. https://www.jstage.jst.go.jp/article/sola/7/0/7_0_101/_pdf

++ We added the reference to Takemura et al. (2011).

3) Page 24535, line 9: Are there any proofs or references of “mean radius 0.25um”?

++ The mean radius used is representative of the distribution of atmospheric aerosol in the accumulation mode and most influenced by washout and rainout effects. It is consistent with the measurements of radioactivity after Chernobyl (IAEA, 2006) and modeling studies by other groups. The removal of accumulation mode particles by wet and dry deposition (the latter being much less efficient) is not sensitive to this assumption as the scavenging efficiency of accumulation mode particles in our model is not size dependent.

4) Page 24535, line 18: Recently, the Chino’s group revised the emission data (Terada et al., 2012). Some comments are needed. (Ref.) Terada, H., G. Katata, M. Chino, and H. Nagai: Atmospheric discharge and dispersion of radionuclides during the Fukushima Dai-ichi Nuclear Power Plant acci- dent. Part II: verification of the source term and analysis of regional-scale atmospheric dispersion, J. Environ. Radioact., 141-154, 112, 2012.

++ We added comments adnd discussion of the revision by Katata et al. (2012) and validation by Terada et al. (2012) of the emission data by Chino et al. (2012).

5) Page 24540, lines 3-4: The modeled results based on the emission inventory by Chino et al. should be shown in Figure and/or Table.

++ Added reference to modeled results by Chino et al. (Fig. 4).

6) Figs. 2(b), 2(c), 3(b) and 3(c): The size of each figure is too small. They need to be improved.

++ The figure size was enlarged for the final print version.

7) Fig. 6 (bottom) The location of “Tokyo Metropolitan Area” is a small mistake. The rectangle should be shifted to the Northeast.

++ The rectangle has been shifted accordingly.

8) Fig. 7, line 1 of the legend “Eastern Pacific” should be changed to “Western Pacific”.

++ Changed to "Western Pacific".

Anonymous Referee #3

The authors modeled the global atmospheric dispersion and deposition of the Fukushima emissions by using the EMAC model. They compared the simulations at two spatial resolutions: one simulation was carried out with a spectral truncation of T255 and the other one with T106. Their source term was composed by 3 isotopes: I-131, Cs-137 and the noble gas Xe-133. They used the Xe-133 release rate assessed by Stohl et al 2012; the I-131 release rate estimated by Chino et al 2011 and they considered the Cs-137 release rate proposed by Stohl et al as well as the one from Chino et al. Simulated air concentration near the ground are compared to CTBTO measurements and results are discussed. Finally, they modeled the consequences of the Fukushima accident due to the deposition and to the inhalation of radioactive materials. The paper is very close from other work like the Ten Hoeve and Jacobson (2012 Energy and Environmental Science) paper. The work need to be enhanced and expanded before publication.

1- p24533 The authors wrote “both radionuclides are released as gases, caesium has a low volatility and partitions into ambient aerosol particles, whereas iodine largely remains in the gas phase.” They wrote it again p24535 . . . This statement amazes me; do the authors have any reference to support this asser- tion? My understanding was that a large part of the Iodine and Caesium were rejected as CsI aerosol. Moreover, non negligible amount of iodine as aerosol was observed. It means that wet deposition of iodine occurred and has to be considered to assess the consequences of the accident and the doses.

++ The Iodine and Caesium radionuclides are emitted as gases and partition into ambient aerosol particles at the lower atmospheric temperatures, which influences the volatility of the gases. Based on the literature (Stoehlker et al. (2012), a factor of

4 gaseous to particulate fraction of I-131 is appropriate. We added a more detailed discussion added to the paper, as requested.

2- About the Stohl et al and Chino et al source terms - The authors describe in detail the method used by Chino et al to estimate the source term. They discuss the uncertainties and show that their source term is highly uncer- tain. They should do the same and highlight as well the high uncertainties of the Stohl et al source term. One of their main concerns about the Chino et al source term is that the assessment is "limited by the use of Japanese station data only and a regional sim- ulation domain". I do not understand their argument since the use of a large amount of Japanese data is the best way to retrieve the release events dispersed above the Japan land. On the other hand, one of the difficulties encountered by Stohl et al was the small number of Japanese station data available to be used with their method. Most of the data were far away from the Fukushima power plant and they could not efficiently help to improve the source term (observed concentration were too low to have any impact). Therefore the Stohl et al source term is also highly uncertain.

++ We agree and added appropriate discussion of the method emplyed by Stohl et al. and associated uncertaities. The discussion of the Chino et al. method was rephrased and expanded.

- The source term proposed by Chino et al was assessed just after the Fukushima accident. They improved it in Terada et al 2012 (Journal of Environmental Radioactivity). The authors should use it instead of the Chino et al one.

++ We added the reference to Terada et al. and discussed the associated differences. Unfortunately, due to limitations of our computational resources, repeating the model simulations is not possible.

3- About the description of the simulations The authors should give the references of the parameterisation they use for the vertical diffusion. What is the horizontal diffusion coefficient they use?

++ We added a reference to Roeckner et al. (2003) where these details about the model can be found.

4- About the model to data comparisons - The simulations are compared to the CTBTO measurements. The authors should recall that the CTBTO measurements have been used to assess the Stohl et al source term. It certainly helps to obtain a good agreement for the model to data comparison. To validate their simulations, the authors should compare their results to measurements not used to assess the source terms. If not possible, they need to enlarge their compar- ison to other data and especially to Japanese data: air concentration and daily deposit measurements wherever data are available, airborne deposition. Unfortunately, the authors can not use dose rate measurements because of the lack of source terms for key isotopes like I-132, Te-132, Cs-136, Cs-134 . . . They should also discuss how their model fit with the observations compared to other model to data comparisons (Terada et al; Stohl et al; Hoeve and Jacobson, 2012 Energy and Environmental Science. . .).

++ We added a comparison of results to those of the other studies mentioned above.

- Do the authors think that a better resolution may improve their results in the vicinity of the source and even at the Japan scale?

++ That is indeed the case, especially in the vicinity of the accident site. The results for Japan have been reworked, also based on comments by other referees. Nevertheless, the model transport was shown to be quite accurate, which will also apply over Japan. Though we agree with the reviewer that the details around the reactors (within 50km radius) cannot be resolved by the model. For that reason, Fig.8 was altered to convey this limitation.

- Figures 2-3-4 are too little and should be enlarged. A solution would be to shorter the time scale and to zoom in on the period where radioactive materials were measured. They should use a time scale with dates. I do not think that the logarithm scale used for the scatter plots is suitable.

++ We followed the suggestions and enlarged the figures for the final print version. The time scale was changed to days and zoomed in to focus on the relevant period. The plots also benefit from higher resolution and more precise (to the nearest hour) time association between measurements and observations.

- The discussion on the CTBTO data is very interesting but requires references.

++ We added reference to the "Detection of radionuclides emitted during the Fukushima nuclear accident with the CTBT radionuclide network" by Stoehlker et al. (2011).

- The authors should precise which source term has been used especially for the Cs-137 simulations. The simulations done with the Chino et al source term should also be plotted and results discussed.

++ Results using Cs-137 inventory by Chino et al. added to the plots.

5- About the deposition assessment - p 25541 the authors wrote “131I, which has a low solubility, so that convection effec- tively redistributes this gas into the free troposphere where the wind speed is typically higher and transport distances larger”. Do they think that it is coherent with the physical behaviour of iodine in the atmosphere? If so, they need to add a reference to support this statement. Do they think that this behaviour may explain the underestimation of the iodine concentration they show Fig.4? If so, they need to discuss such hypothesis. I wonder when and when convection events occurred? - The authors can not neglect iodine wet deposition. - The authors should discuss the validity of their meteorological fields and in particular the validity of the rain fields. - The authors should compare their deposition assessment to other studies, for in- stance to the one from Morino et al. . .

++ Added discussion on I-131 wet deposition. Added comparison of deposition assesment to results by Morino et al.

The low solubility of iodine is based on measurements reported e.g. in the CRC Hand-

book of Chemistry and Physics and is freely available to the public. It is directly reflected in our model results and does not lead to ambiguity. For a description of the processes we refer to Tost et al. (2006, 2007), which are refrenced in the paper. We added discussion on the I-131 wet deposition. We also added a comparison of the deposition assessment to results by Morino et al.

About the validity of meteorological/rain fields: Unlike regional models that are typically driven by boundary conditions and produce their own dynamics, our model dynamics are nudged towards and follow throughout the atmosphere the ECMWF ERA-Interim reanalysis data. We have checked total precipitation in the week of 11-18 March and the period 11 March - 31 May and found good agreement between the model and the ERA-Interim reanalysis data.

6- About the doses assessment - The doses are assessed by considering 2 isotopes only. The impact of other isotopes should be taken into account to assess realistic doses (i.e. Te, other iodine isotopes, other caesium isotopes. . .). I understand that the authors have no source terms for those isotopes but they should at least discuss their potential impact. - The authors assess the doses “around the Fukushima” power plant. What is the dis- tance from the plant they consider? Do they think that the spatial resolution of their sim- ulation is sufficient to allow an analysis of the consequences “around the Fukushima” power plant? The highly contaminated zone in the North West of the plant covers an area of about sixty kilometres long which is less than 2 meshes in their simulations. In the model simulations, emissions are diluted in the release mesh so the concentra- tions computed in this release mesh are usually not used. Several meshes around the release mesh should also be removed from the analysis. What do the authors do to assess the doses within the 150-200 km around the plant? If they use the simulated concentrations they should at least discuss this point and show by using a model to data comparison that the computed concentration are realistic enough to be used in the doses assessment. - I wonder what would be the impact of the given order of magnitude of the doses assessment on the Japan population. The authors should at

least highlight the un- certainties of their study. When reading the paper I understand that the assessment minimizes the doses. In what proportion is that so? - The authors should compare their results with similar studies already published like the one from Ten Hoeve and Jacobson (2012 Energy and Environmental Science).

++ We added consideration of the isotope Cs-134 and discussion of other isotopes – in particular Te-132 for the dose assesement. We also added comments on the limitations of model resolution for the geographic projection of dose distributions within the land area of Japan. We changed Fig. 8 to raster plots from smoothed contours to better show model resolution. Furthermore, we added a comparison of results to those published by Ten Hoeve and Jacobson (2012 Energy and Environmental Science). The population data results are not very sensitive to the resolution issue nearby the reactors because most people live at greater distances.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C10948/2012/acpd-12-C10948-2012-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24531, 2012.