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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol dynamics and dispersion of radioactive particles" *by* Pontus von Schoenberg et al.

Anonymous Referee #3

Received and published: 21 December 2020

Recommendation: Major revision

General comments:

The current manuscript developed a coupled detailed aerosol dynamics and cloud processes - Lagrangian trajectory model system and compared the simulation results under the hypothetical nuclear accident situations. The scope of the manuscript is well suited to Atmospheric Chemistry and Physics. Even though the results presented are not surprising (small impacts of ambient aerosol dynamics and large impacts of wet deposition scheme selections), it is worth publication as the manuscript contains some new materials (new model development). However, the current manuscript is not acceptable in its current form because their main arguments are somewhat misleading for the following aspects.

1. A detailed model may not be necessarily a better model or a model closer to truth. Detailed models need many parameters which contain uncertainties. Unless compared with observations, one cannot say which is closer to truth. Sometimes, simple models performed better. The author stated that the detailed cloud model is closer to truth but their method is still far from truth for the following reasons: (1) the in-cloud model only considers warm rain processes but many of the clouds involves cold rain processes in reality in mid-latitudes. (2) The authors used Laakso et al. (2003), but it is not a theoretical but an empirical model. The authors claimed that the first order models used in LPDM are empirical, but the authors also used an empirical model in their below cloud scavenging. How can the authors prove that their method is better? (3) The most critical point is that the cloud process involves vertical motions of air due to convection, but convection cannot be treated by the trajectory method. At least, LPDM needs to be used to say something about the cloud processes. Consequently, at this moment, to the reviewer, the current manuscript is not to show the comparison between the simple and accurate models, but just to show the comparison between two different schemes, simple and complex. (4) Additionally, there are plenty of first-order schemes with different parameters, but the authors selected only one. There could be schemes which predict more depositions than their complex model, or could be ones predict similar values with their complex one.

2. The authors claimed that aerosol dynamics is important for some cases, but compared to what? How is it important with respect to the grid resolution issues, uncertainty in real time forecast due to chaotic nature of atmosphere, and huge variations due to different selections of cloud microphysics modules and other physical modules? The reviewer fully agrees that model development is always important, but the importance varied substantially depending on the processes and properties. It is not fair to state that aerosol dynamics is important, without comparing other issues except wet deposition. (and with compared to the deposition, the impact of aerosol dynamics is negligibly small.) The reviewer also suspects that the presence of coarse mode particles, which was not considered in the model, could be as important as (or even more important

than) aerosol dynamics as presented in the manuscript.

Overall, the reviewer suggests that the context of the manuscript should be as follows: (1) detailed aerosol and cloud schemes are implemented in a trajectory model, (2) aerosol dynamics was less important compared with other processes such as wet deposition and probably meteorological simulations, (3) selection of a good wet deposition scheme is important as the different schemes caused very different results.

Because the authors did not compare their simulation results against any observations in terms of air concentration and deposition of radioactivity and because their wet deposition calculation is still far from reality, the reviewer highly recommends to remove the phrases such as “accurate” or “close to truth”. The authors may use a term such as “physically consistent” but the reviewer does not support this as well due to the reasons as listed above: (1) – (3) in general comment #1.

3. To the reviewers, the assumption of 10 days being without exchanges with surrounding air may not be realistic. The authors mainly discussed the simulated difference at 10 days but the differences at 1 day is much more important than that at 10 days. Especially for the emergency situations, pollution near the source is much more important. Probably impacts of aerosol dynamics is much less important for the shorter time integrations, i.e., for the highly contaminated areas near the source. This aspects should also be addressed.

Specific comments:

(1) P.1, ln. 22-23, The last sentence of Abstract, “e.g. in 5% of . . . to a simplified version of the model” is hard to understand. Better to be replaced by clearer statement.

(2) P.2, ln. 47: & -> and

(3) P.2, ln. 49: “deposition, and the potential to act as cloud condensation nuclei”: CCN activity may be a part of “deposition”, but why is CCN separated from “deposition”?

(4) P.3, ln. 78-79: The sentence “Therefore, the total . . . no dynamic feedbacks on

the aerosol-cloud interaction” needs additional explanation. Aerosol-cloud interaction has various feedbacks such as cloud albedo, cloud life time, and thermodynamic and microphysical invigorations. Which feedback processes can cause underestimation of total wet deposition and how?

(5) P.3, ln. 85: “these Eulerian approaches including advanced aerosol dynamics are however not easily adapted to a Lagrangian framework.” It seems not very difficult. There exists Lagrangian – Eulerian hybrid methods. Recently, Danielache et al. (2019) (https://doi.org/10.2343/geochemj.2.0542) coupled Eulerian chemical processes to a LPDM framework, although it does not seem to couple aerosol dynamics processes to the radio-Cs modeling, as done by the current manuscript.

(6) P.4, 2. Method: Horizontal and vertical grid resolutions are missing. How they considered vertical motion of trajectory in HYSPLIT is missing. Brief descriptions of CALM may also be needed such as emission inventories (anthropogenic, biomass burning, biogenic, and volcanic emissions), chemical reactions (gas, aerosol and aqueous phase reactions), and aerosol representations (modal or sectional) used. It may help to grasp how their aerosol dynamics parameters are properly set.

(7) P.5, ln. 134: “gap” -> “missing data” may be better.

(8) P.6, ln. 161-162: “super-micrometre size range is however not included in this study”. Aerosol dynamics is less important for coarse mode particles or chemical evolution may enhance the deposition efficiency of coarse mode particles. Neglecting the presence of coarse mode particles in this study may overestimate the impacts of aerosol dynamics processes, which should be noted. If the surface area of coarse mode particles is larger than that of ultrafine particles, gaseous Cs are more condensed to the surfaces of coarse mode particles.

(9) P.6, ln. 170: “Matlab function fmincon.m” Use mathematical expression, rather than a function name of a software.

(10) P. 7, Fig. 2 Units of dN/dlogDp and dS/dlogDp may be missing.

(11) P. 7, ln. 180-181: The sentence "the internal mix of the particles into three different chemical groups, ammonium bisulfate, condensable and partially water-soluble organic vapour and non-condensable insoluble compounds" is awkward. First, "vapour" is not "particles". What is "non-condensable"? Either highly volatile gas or non-volatile particles can be both "non-condensable". Where is ammonium nitrate and ammonium sulfate? Same questions for Table 1 in P. 10.

(12) P. 8, ln. 220-221: What is "column precipitation intensity"? How about the unit?

(13) P.8, ln. 225-226: "The effect of ice components in clouds is neglected, which may overestimate the scavenging efficiency". Why not "underestimate"? Do you assume that the presence of solid hydrometeors delays the scavenging of aerosols? Why?

(14) P. 11, Result -> 3. Results

(15) P. 15, Fig. 6: It is not clear to the reviewer why the difference between purple and green are very large, whereas the difference between yellow and blue are negligibly small. It looks as if "aerosol dynamics are very important when the simple deposition schemes are applied". To my understanding, the simple deposition schemes are independent on aerosol sizes, and thus the difference between purple and green should be even smaller than the difference between yellow and blue.

(16) P. 16, ln. 388: "cloud formation is buffered by newly formed particles" does not make sense. Which buffer system (Stevens and Feingold, Nature, 2009) do the authors indicate? The newly formed particles decrease the maximum supersaturation of air parcel to form cloud?

(17) P. 16, ln. 401: "Melpitz and ." and what?

(18) P. 19, Discussion -> 4. Discussion

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