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Geoscientific
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Interactive comment on "Modeling wet deposition of inorganics over Northeast Asia with MRI-PM/c and the effects of super large sea salt droplets at near-the-coast stations" *by* M. Kajino et al.

Anonymous Referee #2

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The authors present a modeling study over Northeast Asia, with focus on Japan, employing an offline-coupled regional chemistry transport model they have developed. The model contains the major processes necessary to represent gas-phase chemistry and aerosols. A comparison between model results and surface observations of selected gaseous components, aerosol components through analysis of high-volume filter pack samples and measurements of wet deposition of inorganic ions is presented. The authors distinguish between near-the-coast and stations far from the coast in their analysis and focus on Na+ ions to identify a contribution of "super large sea salt droplets" as explanation for a severe underestimation of Na+ wet deposition found when comparing the model against measurements.

Main points of concern:

1) The configuration of the model system used is not adequate to investigate the hypothesis of "super large sea salt" droplets, and no further empirical evidence is presented to support it. A simulation with a 60 km horizontal grid spacing is compared with coastal stations. This is by no means straightforward, and would require considerable sophistication in the point to grid-cell comparison methodology. This is not done, but rather a physical phenomenon is picked as explanation instead. No further evidence is given, except that far from coast stations (again, point against 60 km resolution grid-cell comparison) agree better (which can be an averaging artifact as well).

2) The model system is insufficiently / confusingly described and scattered across introduction and methods section, which hinders e.g. the understanding of specific reasons for shortcomings found. Especially, a discussion of the sea salt parameterization is missing (coastal surf zone included?), which is a crucial step stone in the argumentation. I suggest to revise the model description section and move parts from the introduction in there. See also comments below.

Specific comments:

Throughout the manuscript: it should be checked by an English native speaker to remove language errors.

Abstract

p. 1342 l. 1-5: This is a very long and technical first sentence for a manuscript abstract. It is common scientific practice to force a regional simulation by a global model, so the details are not important at this point and only confuse the reader – rephrase.

p. 1342 l. 6: present -> presented (happens several times in the manuscript)

p. 1342 l. 11: You do not show that your model is able to represent all processes. Rather you hypothesize based on good correlations with wet deposition measurements that all the other processes must be correct, which is not the same. Further, what do you mean by “successfully”? Quantitative information would be much better (mean fractional biases, rmse, slopes, correlations for major components investigated), as “successfully” can be anything.

Introduction

p. 1343 l. 7-8: “to simulate quality of Asian air and precipitation”. You do not explain why precipitation is difficult to model especially in Asia (it is difficult all over the world). Rephrase.

p. 1343 l. 26: You did not develop this model system in this work, but before. So I suggest rephrasing: “To accurately simulate the fate of Asian air pollutants, a model system has been developed with the following components:”. Then list the major components of RAQM2 (e.g. what gas-phase chemistry do you use? How do you treat organic matter? SOA? Bin or modal modules? Which bin ranges modes do you have...?). Aqueous-phase chemistry? Further, do you do cloud processing of aerosols? If so, state. Most important: explain the sea salt parameterization.

p. 1343 l. 27 ff: this is a methods description, so it should be moved from the introduction to the methods section. (p. 1343 l. 27 - p. 1344 l. 16)

p. 1344 l. 8: avoid “successfully” and give quantitative figure instead

p. 1344 l. 10: What do you mean by comparing “mixing types” with observations. What is a mixing type in this context? Do you mean mass contributions? Reformulate.

p. 1344 l. 13: Rephrase the sentence about hemispheric and stratospheric O3, and rather state that you added time-varying lateral and upper boundary conditions.

p. 1344 l. 16: present -> presented

p. 1345 l 1. - 11.: This part is confusing. I understood it that way that these are the inputs the model systems can use, and then you say what you really used. I suggest removing the possible inputs, and only (concisely) state the input for the regional models.

p. 1345 l. 12: add here the section from the introduction.

p. 1345: note explicitly that your chemistry model is offline driven by meteorology.

p. 1345 l. 28: replace Ox by NO2 and O3 (or do you really prescribe Ox and give a fixed split?)

p. 1346. l. 8-9: you should explain why you use different inventories (especially why NH3 from previous EDGAR version)

p. 1346. l. 12: which MEGAN version?

p. 1347. l. 13: There are several current studies showing that neglecting evaporating precipitation leads to considerable errors in pollutant concentrations (Gong, 2011; Saide et al, 2012).

p. 1348. l. 11-12: “is not discussed in this study”

p. 1349 l. 1: this is the crucial point in you manuscript: your model grid cell is completely inhomogenenous where these coastal stations are located. Do you have any other measurements, like e.g. ship measurements, which show the agreement there? The sea salt parameterization in your model is not built for the coastal case (breaking waves, obstacles), but rather for (average) high sea (or is it?). So these stations are not representative when comparing against a model with such coarse grid and without measures to account for this point-to-grid-cell comparison problem.

p. 1349: Section 3.1 is unrelated to the topic of the paper, so it can be compressed in 1 sentence (e.g. “We added time varying lateral and upper boundary conditions for O3 and found good comparison with observations”), and this sentence should be added to the methods section -> model description, plus a note which model is used for BC.

p. 1351 l. 20: I do not understand this sentence. What is the connection between RMSE and averages that makes them being equal a figure of merit? Further, an Rˆ2 of

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0.52 is not good (50 % of variance explained, if you can assume normal distribution). How do other models compare?

p. 1351 l. 23: Does stating “These aerosols are natural and difficult to simulate.” implicate that anthropogenic aerosols are easy to simulate? If not, rephrase.

For the remainder of the manuscript: it is especially confusing for people working on longer timescales when you use the word “trend” in the sense of “slope of the regression line” or “annual cycle”, because “trend” has a completely different significance for them. You should consider using a different term.

p. 1354 l. 4: replace “quantities” with “values”

p. 1354 l. 10: delete “rather”

p. 1355: what about SO2 emission uncertainity?

p. 1356 l. 10: the coastal stations are, for Na+, not representative (see comment above) as they contain sea salt contributions from breaking waves etc. which is not considered in the model parameterization. Hence this analogy with reasonable simulation of other components does not hold.

p. 1354 l. 17: you overestimate SO2, so your SO2 in liquid-phase (and the oxidation products) will be too high, unless your in-cloud sulfur oxidation is too slow (e.g. because you lack oxidation by trace metals). You come to the conclusion that it is right purely by wet deposition measurements – this does not mean your process is right.

p. 1357 l. 10: You say it yourself here: these stations are not representative (and should not be considered – if not correct for biases!)

p. 1357 l. 16 ff:. You find a strong overestimation in T-NO3, so you will overestimate W-NO3 if your wet deposition mechanism is correct. However, you find good agreement, so this suggests it is too low. How does this compare to your other findings? This seems inconsistent.

p. 1358 l. 7-8: what does “to encounter from the bottom of the pack” mean? That they do not reach the pack? Reformulate.

p. 1358 l. 15ff: these motivation sentences come very abrupt at the end of a section discussing another topic, and it does not become clear why you do pH now. Add 1-2 sentences to explain the relations, and move the paragraph to the next section.

p. 1359 l. 2-6.: either explain what this sentence has to do with what follows, or remove it and simply state that you use that parameterisation.

p. 1359: This section reads like it is meant to ensure that coastal stations can still be deemed representative to measure precipitation pH. You should state clearly at the beginning of the section that this is what you intend to do. Also, add this important finding (i.e. that you can indeed still use them) as result in the abstract.

Conclusions

p. 1360 l. 6-9: Do not restate the details of your lateral and boundary forcing setup. p. 1360 l. 13: You did not investigate the transformation, and only very little of the transport, so the conclusion is rather “that the model reproduced the wet deposition of major" p. 1360 l. 15: “within a factor of two to five for (x / y) percent of observed values”. You do have values outside this range!

Figures

Figure 1: Han et al. and Clarke are process models, not emission inventories. They would need their own box, or at least a mentioning of the process (dust, sea salt). (Here or in text): State whether you prescribe aerosols at the boundaries, and if yes, if all species are given, or only a subset.

Figure 4: can be removed, together with section 3.1

Figure 5: Make a single headline (“FC”,”NC”) on top, and add the species name at the right hand side of each row. This saves space and removes visual clutter.

Figure 6 and 7: the timelines of wet deposition do not reveal a lot of information, as one can neither identify individual peaks, nor see differences for low values due to the scaling. I suggest averaging it to monthly values, as done for the aerosol plots on the right hand side.

Figure 8: Using a line to connect categorial variables (stations) does not make sense! Use points or bars instead. Further, I suggest adding a vertical (dashed) line between FC and NC stations to better see the distinction.

Figure 9: Either rewrite your discussion to better explain what is seen, or remove.

Tables

Table 1: x and y indizes are not important for the reader, remove. Remove dash from “character-istics”

Table 2: You cannot have 2 times the “c” index – clarify. Does it really belong in the headline as well (“concentrations (ugm/3)c”)?

Table 3: can be converted into a sentence and then removed.

Table 4: did you average pH by exponentiating, averaging and taking the logarithm again, or simple averaging?

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