

## Point-by-point responses to the comments of anonymous referee #2

(Referee's comment in blue, author's reply in black)

### *Overview:*

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<sup>+</sup> ions to identify a contribution of “super large sea salt droplets” as explanation for a severe underestimation of Na<sup>+</sup> wet deposition found when comparing the model against measurements.

### *Main points of concern and replies:*

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).

We admit that your statement is convincing. We removed the statements highlighting the super large sea salt droplets (SLSD) from the major part of the manuscript (such as title and abstract).

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.

We didn't include the coastal surface zone emissions. We substantially revised the model description part according to your specific comments.

*Specific comments:*

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

I am sorry for this. The manuscript was really sent to the grammar correction by native speakers before submission. In some cases, I found the reason how mistakes happened is because they changed our original meaning but I simply missed it before submission. I don't have time and budget to send it again to the grammar correction before re-submission this time, but I will certainly send it to the correction again by the final publication.

*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.

We rephrased it to “We conducted a regional-scale simulation over Northeast Asia for the entire year of 2006 by using an aerosol chemical transport model, with the hourly lateral and upper boundary concentrations predicted by a global stratospheric and tropospheric chemistry-climate model.”

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

I asked one native (American) English speaker about this and she said “present” sounds more natural in the cases.

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.

I modified the sentences to “Statistical analysis showed that about 40-50% and 70-80% of simulated concentration and wet deposition of SO<sub>4</sub>, NH<sub>4</sub>, NO<sub>3</sub> and Ca are within factors of 2 and 5 of the observations, respectively.”

*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.

I meant “quality of precipitation”. I rephrased to “to simulate concentration and deposition of atmospheric constituents in Asia.”

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.

We modified this part to “a model system has been developed with the following components: a model system has been developed with the following components: emissions of anthropogenic trace species, biomass burning, biogenic and natural (dust and sea salt) aerosol emissions, advection and turbulent diffusion, photochemistry and new particle formation, gas-to-particle conversion of inorganic and organic compounds, Brownian coagulation, CCN and IN activation and cloud microphysical processes, grid-scale liquid-phase chemistry in hydrometeors as well as in aerosol water, sub-grid-scale convection and wet scavenging, and dry deposition of gas and particles (details are given in Kajino et al., 2012, and in the current paper)”.

p. 1343 l. 27: 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)

We moved this part to Section 2.1.

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

We avoided the term “successfully” here and elsewhere, too. We changed the phrase to “Kajino et al. (2012) showed that the modeled size distributions such as  $PM_{2.5}/PM_{10}$  of total mass and  $PM_1$ /bulk ratios of chemical components were consistent with those observed.”

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.

We deleted the sentence. It is a bit difficult to explain in a few sentences. It is not an important information here in the context.

p. 1344 l. 13: Rephrase the sentence about hemispheric and stratospheric O<sub>3</sub>, and rather state that you added time-varying lateral and upper boundary conditions.

We modified it as follows: “They used the constant climatological values for boundary conditions so the hemispheric transport and the intrusion of stratospheric O<sub>3</sub> may not be well

represented. In the current study we added time-varying lateral and upper boundary concentrations of gaseous species predicted by...”

p. 1344 l. 16: present -> presented

I asked one native (American) English speaker about this and she said “present” sounds more natural.

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.

I am sorry for this. We added the sentence clarifying the combination that we really used in Figure 1 and modified the main text as well, accordingly. We are preparing to submit a series of papers by MRI-PM using the different combinations listed in the Figure 1.

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

I did it.

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

I did it.

p. 1345 l. 28: replace Ox by NO<sub>2</sub> and O<sub>3</sub> (or do you really prescribe Ox and give a fixed split?)

NO<sub>2</sub> was included in NO<sub>x</sub> so I changed it to O<sub>3</sub>.

p. 1346. l. 8-9: you should explain why you use different inventories (especially why NH<sub>3</sub> from previous EDGAR version)

Because EDGAR3.2 does not provide NH<sub>3</sub>. I added it in the main text.

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

MEGAN2.

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).

Thank you for the information. I modified the sentences as follows (I could not find the literature Gong, 2011): “Aerosols in CLD and ICE not converting to RNW, SNW, or GRW within a time step are defined to evaporate and regenerate again after the cloud microphysics operator. The chemical components in RNW, SNW, and GRW are assumed to reach the ground surface instantaneously as wet depositions. Resuspension due to rain evaporation could be an important

source of sub-cloud aerosols (Saide et al., 2012) but the current model does not consider the process.”

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

I changed it.

p. 1349 l. 1: this is the crucial point in you manuscript: your model grid cell is completely inhomogeneous 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.

No, we do not have any other measurements than we showed. Also, both our sea salt parameterization and the model grid cell are not suitable for the case. We gave up focusing on the sea-salt issues, simply stated in the abstract and conclusion that we failed to simulate Na, and further modeling study with finer grid resolution and a suitable sea salt parameterization (i.e. surf zone) is necessary.

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 O<sub>3</sub> 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.

I deleted Section 3.1 and added simple sentences in Section 2.1 as “The MRI-CCM2 well reproduced the seasonal variations in measured O<sub>3</sub> concentrations at stations near the edges of the RAQM2 domain.”

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 R<sup>2</sup> of 52 is not good (50 % of variance explained, if you can assume normal distribution). How do other models compare?

We modified the sentence to just mention the fact that there are positive correlations and the RMSE is same order as averages.

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.

We rephrased it to “Despite the large uncertainty in simulating dust emission flux...”

We used “trend” in the sense of “time variation”. We changed the word “trend” to “time variation” throughout the manuscript.

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

I changed it.

p. 1354 l. 10: delete “rather”

I deleted it.

p. 1355: what about SO<sub>2</sub> emission uncertainty?

It should be there. I added it and changed the sentences to “In addition to uncertainty in SO<sub>2</sub> emission amount, overestimation of SO<sub>2</sub> is probably due to...”.

p. 1356 l. 10: the coastal stations are, for Na<sup>+</sup>, 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.

Since the SLSD issue is no longer a main topic, Sect. 3.5 (now Sect. 3.4) is simplified. We also stated that we failed to simulate Na<sup>+</sup> due to the coarse grid resolution.

p. 1354 l. 17: you overestimate SO<sub>2</sub>, so your SO<sub>2</sub> 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.

In the revised manuscript, we tried to avoid claiming that our model succeed to simulate the whole processes and just mentioned the statistical values and degrees of consistency in the model system. Please see the revised Sect. 3.4 (now Sect. 3.3).

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

In the revised manuscript, we claimed that further simulation with finer grid resolution is necessary to simulate Na<sup>+</sup>.

p. 1357 l. 16 ff.: You find a strong overestimation in T-NO<sub>3</sub>, so you will overestimate WNO<sub>3</sub> 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.

We tried to avoid the terms “successful” or “reasonable” for the process modeling and just gave the statistical values. Please see the revised Sect. 3.4 (now Sect. 3.3).

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

Yes. I changed the sentence to “a large downward velocity to reach the bottom of the pack”.

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.

Thank you for the suggestion. I moved the paragraph to the next section with modification as follows: “EANET has been monitoring pH of precipitation in Asia since 2001. There are currently 12 stations in Japan and half of them are situated very close to the coast. The pH of sea water is about 8, much higher than that of precipitation in Japan (4~5). If our hypothesis presumed in Sect. 3.4 were true, the short-lived LSP could raise the pH values of precipitation in Japan and those values were not representing the precipitation of whole Japan (representing only the traveling distance of LSP, 1-10 km). However, because the salinity of precipitation is much lower than that of sea water (about 3 orders of magnitude, estimated from the averages of Table 2), pH of sea-salt originated components in precipitation would be almost 7 and thus, should not affect the pH of precipitation substantially. We proved this from the following calculation.”

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.

I simplified the sentences to “We assumed the chemical components of sea water as Song and Carmichael (2001), CO<sub>2</sub> mixing ratio at 360 ppm, ocean surface salinity of 35, and obtained a consistent value of sea water pH (8.2). In order to assess the applicability of the method, we compared....”

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.

Thank you for your suggestion. Please see the first paragraph of Sect. 3.5. I didn’t include it in the abstract as the discussion is no longer a major focus but mentioned it in the conclusion.

### *Conclusions*

p. 1360 l. 6-9: Do not restate the details of your lateral and boundary forcing setup.

I deleted it.

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” and 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!

I changed the sentences to “Statistical analysis showed that about 40-50% and 70-80% of simulated concentration and wet deposition of major anthropogenic and natural aerosols are within 2 and 5 of the observations, respectively. The performance in simulation wet deposition amount is still noteworthy, because there had been differences of one to two orders of magnitude among several regional scale models for simulating monthly mean values of wet deposition (Wang et al., 2008).”

### *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.

I modified the Figure 1 accordingly. As the current version of MRI-CCM2 simulates only bulk concentration of sulfate, nitrate, and ammonium (no mixing type nor size information), I didn't use the time-varying conditions for aerosols (very low constant value is applied instead). We modified the sentences e.g. we used time-varying

Figure 4: can be removed, together with section 3.1

I deleted the figure and section, and corresponding sentences.

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.

I did it (and renamed to Figure 4).

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.

I modified the figures to show 2-weekly mean values to be consistent with aerosol concentrations in the figures and with Table 2 (now renamed to Figs. 5 and 6).

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.



I did it (and renamed to Figure 7). Thank you for your suggestion.

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

We removed it as the SLSD is no longer a main topic of the revised version.

### *Tables*

Table 1: x and y indices are not important for the reader, remove. Remove dash from “characteristics”

I did it.

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

I deleted the “c” with the “ugm-3”.

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

I deleted the Table 3 and gave the values in the main text, instead.

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

As written in the main text, average pH is equated to minus the common logarithm of average of daily  $[H^+]$  concentration. I added an index “a” to explain it in Table 4 (now changed to Table 3).