Point-by-point responses to the comments of anonymous referee #1

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

Overview:

This study evaluates predictions of a regional AQ model for wet deposition, precipitation, speciated particle concentrations, and ozone. Based on differences in model performance for sodium wet deposition and concentration at near-coast and far-from-coast sites, conclusions are drawn about the impact of "super large sea salt droplets" near the coast.

General Comments and replies:

1. The reference to super large sea salt droplets should be removed from the title. The manuscript does not present direct measurements of such droplets and the modeling does not simulate the droplets. While such droplets could impact coastal sites as indicated by the authors, this study does not present solid evidence to support the existence of the droplets and their characteristics (e.g., size). In the conclusion section, the authors indicate that the underestimation of sodium at near coast sites was "probably due to the contribution of short-lived super large sea salt droplets". This type of statement is reasonable in helping to understand a model performance issue, but the lack of direct evidence of such effects is not strong enough to warrant the current title. Also, as mentioned below, the model configuration is not suitable for simulating near-coast processes, and so impacts of these processes should not be highlighted in the title.

I admit that your statement is convincing. I removed the statements highlighting the super large sea salt droplets (SLSD) from the title and the abstract.

2. There is not much detail on the approach to parameterizing sea salt emissions in the study. Was a special treatment of sea salt emissions from the coastal surf zone used? If so, how wide was the coastal surf zone assumed to be? Other modeling studies at finer resolution have found reasonable model performance for sodium predictions at coastal sites when a surf zone parameterization was used, but no mention of surf zone effects is given here. How was the impact of the coarse grid resolution (60 km) on sea salt emissions handled? For example, coastal grid cells would significantly overlap the ocean and the land. The average emissions in such a cell, which might contain a monitor, would be based on a combination of open-ocean emissions, coastal surf zone emissions, and land emissions. It is unclear how to interpret model performance results in such a situation. Why was such a coarse resolution used in this study where the focus of the evaluation is on a relatively small subset of the domain? A nested domain with finer resolution would provide a better resolution of the processes that are being evaluated.

We didn't include the coastal surf zone emissions. Only ocean surface coverage ratio in a grid (0-1) is multiplied to the sea salt emission flux. We gave up sticking on the SLSD issue, rather

focusing on Asian scale long-term simulation by a newly developed model system. We stated in the conclusion that further modeling study is necessary using finer nested grids and coastal surf zone emissions.

3. The definition of the super large droplets is vague in the manuscript. The authors define "super large" as particles with diameter greater than 10 "or even 100" microns in parts of the manuscript. This definition is unclear. Common sea salt emission parameterizations represent particles with diameters up to 20 microns (e.g., see de Leeuw et al., JGR, 2000, "Production of seas spray aerosol in the surf zone") without considering the wave-crest tearing process mentioned in defining droplets as "super large" here.

4. More details are needed on the diameter cutoff for the PM and wet deposition measurements. The idea that large sea salt particles would not be sampled with the filter packs but would be sampled with the wet deposition sampler needs to be clarified with details on the sampling efficiency for the measurements.

I changed the definition of super large droplets as droplets larger than 10 um and changed the term to LSP as large sea salt particles. As stated in the manuscript, it was found that the filter pack efficiently collect particles up to 10 μm by comparing with the Andersen sampler. However, with a flow rate of 1 L/min with filter diameter of 47mm, the average flow rate is about 0.2 cm/s ($=v/\pi d^2$; d=47 mm, v=1 L/min). This speed is in the case of pack without the filter so the actual flow rate should be lower than this. The gravitational settling velocity of particle with D=10μm is about 1 cm/s so the collection efficiency of particles larger than 10μm could be lower for the current settings of the method. As we wrote, wet deposition measurements are done by wet only sampler, so any size of droplets can enter the bottle when the water sensor detects precipitation.

Specific Comments and replies:

p.1342, line 14: The first reference to sodium on this line should be removed since it states that sodium was successfully modeled, whereas the second reference to sodium contradicts the first by indicating performance issues for sodium

I did it and wrote that the prediction of the sea-salt originated component Na^+ was not successful.

p. 1342, line 26: Should "administrative" be "regulatory" or "air quality management" purposes?

We changed it to "regulatory".

p. 1345, line 2-10. The description of the configuration is confusing. For example, on lines 6-8 it is stated that WRF "or" JMA were used to simulate meteorology, and then the next sentence indicates that WRF was used to simulate meteorology. Please clearly state the configuration for

each model run. Also, it is unclear in the upper-left box of Figure 1 which set of analysis data is being passed to WRF and which is being passed to MRI.

In this paper, WRF is selected as regional model, driven by NCEP-FNL. MRI-CCM2 is driven by JCDAS. The explanation is added in Figure 1. Also, the sentence in the main text is modified accordingly.

p. 1346, line 14: As mentioned above, please provide more details on the sea salt emission parameterization used including at least information related to the size range for particle emissions and how the surf zone was treated.

We inserted the following sentence: "As the proposed size distribution of the Clarke module is trimodal, this was modified into a unimodal distribution while preserving the number and volume concentrations with a standard deviation of σ = 2. We did not consider the surf zone emission (e.g. de Leeuw et al., JGR, 2000) as it is not resolvable for the grid resolution (Δx=60km) of the current model setting".

p. 1347, section 2.2: Please provide more details on the observations including at least information on the diameters of particles and droplets that are sampled. If there are known measurement accuracy issues for sodium (or other species) please note them.

We inserted the least information as follows: "The four-stage filter pack is composed of four filters in line with the air streams entering from the bottom with a flow rate of 1 L min^{-1}. Aerosols are collected on the bottom (thus first) pack with Teflon filter (pore size; 0.8 μm, diameter; 47mm) to attain good efficiency to collect sub-micron particles (EANET "Technical Documents for Filter Pack Method in East Asia"). By comparing with the simultaneous measurements using the Andersen sampler (a multi-stage cascade impactor), it was found that the FP method efficiently collected particles up to diameters of 10 μm."

p. 1350, Fig. 4: Why not show a full boxplot for the ozone evaluation?

We just followed previous studies and there were no reasons for not using a full boxplot. Anyway, we deleted Fig.4 and Sect. 3.1 as they are not a main topic in the manuscript (please see the referee 2's comment).

p. 1351, line 9: Please add some measure of bias, such as normalized mean bias, to table 2.

I added it. Thank you for your suggestion.

p. 1351, line 26: The evaluation presented here does not support the statement that long-range transport was simulated correctly. The evaluation would need to consider model performance at various distances from the source region to understand how well transport was simulated. For example, if dust emissions were vastly underestimated in the model or vastly overestimated in

the model, the evaluation results presented would take on different meaning. But we have no idea of model performance for dust near the source region.

We avoided the statement “simulation of transport was successful”. Please see the revised sentence as: “Desipite the large uncertainty in simulating dust emission flux, we were able to obtain a good value of R^2 (0.50) for nss-Ca^{2+}. It is because all the observation stations are situated distantly from the extensive source regions (e.g. Takla Makan and Gobi desert) and in the downwind region when long range transport of Asian dust was predominant in spring and autumn in 2006.”

p. 1352, lines 6-11: The relatively high bias at the far coast sites compared with the near coast sites suggest the model is no transporting sodium correctly. For instance, if the model underpredicted the deposition velocity of sea salt particles, the predictions would be increasingly biased high with distance from the coast as appears to be the case here.

You are right. We deleted the sentence “transport patterns were better reproduced by the regional model” and modified it to “We obtained a larger R^2 for Na^+ (0.27) at the FC stations, which were distant from the emission source.”

p.1352, lines 21-24: This logic is unclear to me. It seems to suggest that there is no relationship between complex terrain and vertical air motion. Such a statement requires more justification

Orographically-induced precipitation could be there at Yusuhara station. We deleted the sentences.

p. 1353, line 2: How do you know the size of the particles? There are no particle size measurements presented in the manuscript.

We wanted to talk about “typical sizes” of submicron particles. We deleted it, anyway.

p. 1354, line 24: How do you know that overestimation of SO2 is not due to overestimation of emissions?

We included the statement “In addition to uncertainty in SO_2 emission amount, overestimation of SO_2 is probably due to…”

p. 1357, line 5: In the definition of coarse particles, do you mean “greater than” 1 micron

I meant around one micron. I changed it to ~several μm.

p., 1359, line 26. Why is the impact of sea salt on pH so small? Does sea salt effectively contribute a balanced mixture of strong cations and anions?

This is mainly because the salinity of precipitation is about 3 orders of magnitude smaller than that of sea water. If pH of precipitation without seawater is 4 and weight fraction of seawater in

precipitation is 1%, pH of precipitation with the seawater is –log[0.99*1e-4 + 0.01*1e-8] = 4.004364 so ΔpH=0.004. We simply added it in the first paragraph of Section 3.5 as “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 the diluted sea water becomes almost 7 and thus, should not affect the pH of precipitation substantially.”

p. 1360, line 13: The conclusion that transport was simulated accurately does not account for the increasing bias of sodium with distance from the coast.

We modified the sentences. Basically we avoided the statement “simulation was successful” throughout the revised manuscript and just gave statistical values, instead.

p. 1363, line 4-5: The sentence “One must . . .” is unclear to me.

I deleted the sentence.

p. 1361, line 19-21: The coarse spatial resolution probably has a much larger impact on performance than the 1 hour time resolution.

Yes, you are very right. I changed the final paragraph as follows: “Finally, online coupling of regional-scale meteorology with a chemical transport model with finer grid resolutions will be essential for better simulation of the processes of wet deposition. Although RAQM2 successfully “diagnosed” autoconversion and accretion rates with a mixed-phase parameterization of cloud microphysics, these rates were based on environmental and microphysical parameters with 60 km of grid spacing at a time resolution of 1 h (the input/output time step of the WRF and RAQM2 models), which is too coarse and too long for the special and temporal scale of dynamic and microphysical processes in clouds.”