

***Interactive comment on* “Regional impacts of ultrafine particle emissions from the surface of the Great Lakes” by S. H. Chung et al.**

S. H. Chung et al.

vanreken@wsu.edu

Received and published: 7 November 2011

Response to Anonymous Referee #2

We thank the reviewer for his/her detailed and constructive comments. We have revised the manuscript based on the concerns outlined, and believe the paper is better as a result. A general response to the major issues raised by the reviewers is being submitted separately as a General Comment. Much of that response deals in a general way the major concerns raised by this reviewer. This direct response addresses the concerns raised specifically by this reviewer, in the order presented in the review.

Major Comments

1. Page 16211, line 10: the main trace constituent in sea-water is of course sea-salt,

C11573

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



and its concentration in the original water is on the order 35 g/kg (Pond and Pickard, Introductory Dynamical Oceanography, 2nd Edition, 1983). What were the main constituents suggested by Slade et al (2010) and their mass mixing ratio in the original water? A few lines of further description is needed here: the current paper is predicated on the possibility that significant particle formation from wave-breaking could be occurring, but the composition of those particles is a key issue. What attempts were made in the current work to simulate the composition of the fresh-water particles, as opposed to just wave breaking and assuming the particles were the same as sea-salt?

- Slade et al. (2010) measured the composition of Lake Michigan water and the results are listed in their Table S1. They measured ~ 2.1 g/kg for NaCl, and about 12.6 g/kg of total solute. Within the model we assumed that the composition of emitted dry particles follows the relative concentrations of the analyte in the lake water as measured by Slade et al (2010), except that we reclassified Mg^{2+} and Ca^{2+} as Na^{+} because the WRF-Chem speciation does not include the former two ions. As a practical matter this choice had minimal influence on number-based results presented throughout the manuscript. We ran sensitivity simulations in which we changed the composition of the emitted particles and found that the results are very similar. We have modified the manuscript to include the composition information.

More broadly, this comment and many of those that follow suggest a fundamental misunderstanding of the goals and methodology in this work. We take it as given that the size distribution and composition of particles emitted from the lake surface will be drastically different from the marine environment. We emphatically did not assume that these particles were the same as sea salt. All of section 2.3 is devoted specifically to discussing how we chose to parameterize freshwater particle emissions. In the absence of previous work on this problem, we did begin with the assumption that the mechanical production of large droplets by wave breaking and bubble bursting would be the same in freshwater and marine environments. Since each evaporating droplet would leave precisely one residual particle, this assumption implies that the number of

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

particles emitted from freshwater would be the same as in the marine environment, to first approximation. Thus we felt justified in using a marine parameterization that only predicted number emissions. This is not the same as assuming that the mass emitted, the composition, or the size distribution is the same. We placed all of those particles emitted from the lake in the smallest bin in our model, consistent with the relative purity of the parent freshwater droplets and the direct observations of Slade et al. (2010).

2. Page 16216, lines 22 to 25: While I agree with the author's assertion that the droplets produced should be similar to those produced by ocean water, the droplets are not the key issue, but rather, the amount of mass that remains once those droplets evaporate. Parameterizations such as those used by O'Dowd are for sea-salt fluxes, and make use of the knowledge that NaCl is in the bulk water in a known amount prior to that water being released into the atmosphere as waves as a result of wave action. The evaporation of those droplets then leads to the formation of aerosols (of sea-salt). This needs to be clarified in the current paper: what is the expected fractional mass of the trace compounds in the bulk lake water that, when included into the droplets that are released through wave action, result in aerosols, post-evaporation? Sea-salt has apparently been used as the model for particle formation here – what evidence is there that sea-salt is a good or appropriate model? At a minimum, I would expect a similar amount of dissolved mass in the bulk water. Better would be to have some evidence to suggest that the thermodynamic properties of the fresh-water solute would be similar to that of sea-salt (i.e. will the evaporation be similar, so that the sea-salt flux algorithms could be applied)?

- We disagree strongly that mass is the key issue, and assert instead that the number of particles emitted is the key issue. As concluded by Slade et al (2010) and discussed in the introduction of this manuscript (page 16211, lines 18-23), we already know prior to this study that the mass of the lake-emitted particles would be only a very small fraction of the mass of the total aerosol population in the region. The goal of this study was to address the potential impact of lake aerosols on total aerosol number concentration,

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

which is important for cloud formation and other atmospheric processes.

3. Page 16217, equations 1 and 2: again, are these fluxes for droplets or for dry aerosol mass, and if the latter, what was the assumed solute composition? If the former, what are the assumptions regarding droplet evaporation, etc.?

- Again, these parameterizations were derived to estimate the number of particles emitted only. Equations 1 and 2 did not make assumptions as to composition or size distribution. It is specifically for this reason that Equation 1 (based on Geever et al., 2005) was chosen as the model for this work. As the reviewer suggests, the mass and size distribution would indeed be different, but the number should be comparable- one emitted droplet leads to one residual particle. We only used the parameterization to estimate number, and then used our knowledge of the system and the observations of Slade et al. (2010) to place those particles in the appropriate size bin- the smallest one available.

4. Page 16218: Its not clear from the author's work whether the parameterization used for particle emissions here (Geever et al, 2005) compares well to the Gong et al (1997) parameterization used in WRF-CHEM, or why a different parameterization was used in the first place. Are both parameterizations originally intended to describe sea-salt particle formation from evaporating droplets?

- The Gong et al. (1997) parameterization was originally designed to describe sea-salt particle formation and was already in the public-released version of WRF-Chem. However, as discussed in the manuscript, in WRF-Chem this sea-salt emission parameterization was also applied to all water bodies, including the Great Lakes. It was not the intention of this study to compare sea-salt parameterizations. We chose Geever et al. (2005), also derived for the marine environment, because Geever et al. (2005) is based on measurements that included submicron particles (whereas the Gong et al. 1997 scheme strictly applies only to particles with diameters in the 1.6-10 micron range) and because the Geever et al. parameterization focuses on total number fluxes.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Our main assumption was that lake aerosol particles are produced mechanically the same way that sea-salt particles are generated and thus the number flux, not mass or size distribution, should be the same in both environments, to first approximation.

5. Page 16219, lines 12-13: “emitted particles are somewhat larger than what was observed by Slade et al (2010)”. Once again I’m wondering about the role of evaporation and whether the Geever parameterization is for particles (i.e. evaporated droplets) or droplets of sea-water, or droplets of fresh water. This needs to be clarified.

- We have revised the text to improve clarity, but we feel the important points were described in section 2.3. Within this model structure, the smallest particle size bin available was 40-80 nm; that is just how the model was developed. Because that is the smallest size bin available, we put all of the particles emitted by the lake there. The clause quoted above was intended to remind the reader that this was a source of uncertainty- the observations of Slade et al. (2010) suggested that the particles from the lake surface were smaller, 20-40 nm. If a smaller size bin had been available, we would have put the lake-emitted particles there instead.

6. Page 16222, and figures 2 to 4: what evidence is there to suggest that the mass of solute in all of the Great Lakes might be the same? This is implied in the use of a single emission function for all of the lakes.

- Again, we made no assumptions about mass whatsoever- only about number. We did assume a single composition for all lake-emitted particles, based the composition provided in Table S1 of Slade et al (2010). Sensitivity simulations with different chemical compositions of emitted particles indicated that the composition had only minimal impact on our results (results not shown). We have added text in section 2.3 in the revised manuscript to include this information.

7. Page 16222, discussion on Figure 5: it would be worthwhile to comment at this point on the likely range of accuracy of the nucleation process itself. Figure 3 suggests that, with nucleation, the surface flux from the water can account for another 20% on the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

particle number. Without nucleation, the surface flux accounts for another 160% on the particle number. But more importantly, what is the range of masses typically predicted for different nucleation parameterizations? Quoting a few references that compare different nucleation processes would be useful to underscore the authors' point here.

- On page 16226, lines 26-27 of the original manuscript we discussed how new particle formation models used in the literature vary by several orders of magnitude. We also discussed on page 16227, lines 4-8 that ambient measurements have suggested that binary nucleation theory underestimate observed nucleation rates, but that these results are not conclusive for all regions. We have added text in the revised manuscript to further emphasize that in some instances the binary nucleation theory can overpredict nucleation rates under ambient conditions.

8. Page 16227, lines 15 and 16: At this late point in the paper, the reader learns that the Geever et al parameterization is for ocean water; i.e. for sea-salt solution water. I think that this is not just another significant source of uncertainty in the work, it's the most significant one. The paper's conclusions are predicated on the idea that amount of aerosols formed from ocean waves will be the same as that from fresh water. In the case of ocean waves, the source of the mass is the dissolved NaCl. What is the source of the mass in the fresh-water waves? The Geever parameterization's origin as an ocean aerosol flux parameterization needs to be mentioned earlier in the paper. More importantly, the authors need to make a much stronger case that an ocean aerosol flux algorithm would give accurate lake aerosol fluxes. What evidence is available in the literature that suggests that the concentration of dissolved solute in lake water is as high as the amount of dissolved NaCl in ocean water? What is the chemical nature of that dissolved solute? This discussion is missing from the paper, but must be included before I could recommend publication.

- The whole premise of the paper was that particle emissions from freshwater lakes are a newly proposed phenomenon- how could a freshwater parameterization already exist? We feel that section 2.3 made it quite clear that the Geever et al. (2005) parame-

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

terization was developed for a marine environment. We discussed early in that section that a marine approximation is a reasonable proxy for number emissions, and later stated directly that the parameterizations were based on measurements collected at Mace Head, Ireland (a marine coastal site) and verified using ship-borne observations from the Atlantic Ocean. Still, we have revised the manuscript to make this connection more explicit.

For the rest of this comment, we refer again to the earlier discussions in this response and in the General Comment.

Minor comments

1. Page 16209, line 25: Suggest using “spatially inhomogeneous and locally intense” rather than just “intense” in this sentence.

- Thank you for the suggestion. We have the word ‘locally’ here, but do not want to suggest that natural emissions are spatially homogeneous and have therefore left out the other part of the clause.

2. Page 16210, line 3, “because the emissions intensity of natural sources can be so small” would make more sense as “despite the emissions intensity of natural sources being so small locally”.

- We have rewritten this sentence to clarify its meaning.

3. Page 16214, lines 1 through 12: one question I have with regards to a laundry list of model options such as presented here, is the extent this combination of options have been tested. That is, is this a configuration that is a standard operational version of the model (and if so, what are the references for the statistical tests of same)? Another way of looking at it: are these parameterizations part of a well-established configuration of the model, or are some new/different/particularly suited to/sensitive to the parameterization being examined in the current work?

- The WRF-Chem model is a community model with several options for physics. There

is not a set of options that has been universally recommended. How various physics options impact the model results is an area of active research, and many physics options are also being improved continuously as more scientific knowledge are gained. The physics option used here are commonly used and are the same as those used by Kim et al (2006) for studying the same eastern US domain used for this study. Among the physics option available in the model, we believe the planetary boundary layer (PBL) and the associated surface-layer schemes are most likely to have an impact on our results because they affect the boundary-layer height, which affects vertical mixing of emitted particles, and surface wind speed, which affects how much lake aerosols are emitted. For this reason, we chose to do sensitivity simulations using the MYJ PBL scheme in place of the more commonly used YSU PBL scheme.

4. Page 16214, lines 20 to 22: what was the composition of emitted aerosols from Slade et al, and how well does this match the composition assumed for the aerosols emitted as part of the current work? Ditto, lines 3 to 5 of page 16215.

- Slade et al. (2010) measured the composition of Lake Michigan water and the results are listed in their Table S1; we are not aware of any measurement of the composition of nanoparticles in ambient air over the Great Lakes. For our study, we used the results of Slade et al (2010) for the composition of emitted lake particles, except that we reclassified Mg²⁺ and Ca²⁺ as Na⁺ because the WRF-Chem speciation does not include the former two ions. We ran sensitivity simulations in which we changed the composition of the emitted particles and found that the results were very similar. We have added text in section 2.3 of the revised manuscript to include the composition information.

5. Page 16215, line 9: period missing after Jacobson reference.

- Good catch. This has been fixed in the revised manuscript.

6. Page 16215, lines 15 through 28: I was a bit surprised that 1999 emissions are still being used – there are more recent emissions databases being used in both the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

USA and Canada (2005, 2006) by the US EPA, NOAA, and Environment Canada, respectively. I see that some of the major point sources were updated using CEM data – but why not use the more recent inventories?

- This was a purely practical choice. We had access to 2004 simulations that used the 1999 emissions, and we did not have easy access to later results. Since our goal was to assess the potential impacts of the proposed lake-surface source, we did not feel that the choice of emissions inventory was critical to our methodology, within a reasonable range.

7. Page 16217, line 21: a few more details on the wind speed calculation are needed: presumably the same similarity theory stability functions as in the model were used, for example?

- This is correct. We used the same logarithmic wind profile with the same stability function as those used in WRF's 10-m windspeed calculations. The stability functions used are dependent on the choice of surface-layer scheme chosen in WRF. We have added text in section 2.3 to clarify this point, but we refer the readers to the WRF physics documentation (Skamorack et al. 2005) for more details.

8. Page 16218, line 28 to next page: I was actually a bit shocked by this statement (that the WRF-CHEM model in its default configuration does not distinguish between sea water and fresh water, and treats fresh water as ocean) – I have to take it for granted since I'm not familiar with the WRF-CHEM model.

- The reviewer is not the first to express shock at this. We were quite surprised as well. There are few water bodies where this shortcut would have a significant impact, and we happened to focus on one of them. We have added new text in the manuscript to note that versions 3.0 and later of WRF do allow for the option of having inland water bodies as one of the land-use categories; however, this is not the default option for running WRF and the WRF-Chem source code does not make use of the new inland water body category even if this land-use category is utilized in the meteorology simulation.

9. Page 16220, line 14: please include the mass emission rate, for comparison purposes, as well as the number.

- We have intentionally framed our entire study to focus on number and not mass. As noted both by Slade et al. (2010) and in section 1 of this manuscript (page 16211, lines 18-23), the mass of the lake-emitted particles would only be a very small fraction of the mass of primary particles emitted in the upwind region. Further, the emissions parameterization is a number-based parameterization, without any size or mass information. This is all by choice, with the reasoning described at length in the manuscript. To include a mass emission rate would confuse our argument in a way that we would like to avoid.

10. Page 16220, line 17: in addition to percent difference, the authors should show the non-normalized differences, as a third plot.

- We have followed the reviewer's advice and have added non-normalized difference plots to Figures 3, 5, 8, and 10. We have also modified the text to include discussion of these addition figures.

11. Page 16222, discussion on Figure 8: once again, a difference in addition to a percent difference on the figure would give the reader a better idea of the importance of the process. Ditto for Figure 10.

- Please see our response to comment #10 above.

12. Page 16226, lines 22, 23: I assume that by "new particle formation rate" you mean "nucleation rate" here – please clarify. The former could be taken to include, eg, sea-salt particle formation.

- Our word choice was intended to avoid confusion for a different part of the aerosol research community. For many researchers, nucleation rate refers specifically to the formation of molecular clusters below detectable size that may or may not grow to become detectable particle. The new particle formation rate in this manuscript refers

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

to those particles that nucleate and grow to detectable size. We have revised the manuscript in sections 2.1, 3.1, and 4 to avoid the confusion the reviewer describes.

Reference

Kim, S.-W., Heckel, A., McKeen, S. A., Frost, G. J., Hsie, E.-Y., Trainer, M. K., Richter, A., Burrows, J. P., Peckham, S. E. and Grell, G. A.: Satellite-observed US power plant NO_x emission reductions and their impact on air quality, *Geophys. Res. Lett.*, 33(22), L22812, doi:10.1029/2006GL027749, 2006.

[Interactive comment on Atmos. Chem. Phys. Discuss.](#), 11, 16207, 2011.

ACPD

11, C11573–C11583,
2011

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

C11583

