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

Anonymous Referee #1

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My main concern with this paper is that the authors have used a wave-induced aerosol flux parameterization that was designed for ocean aerosol flux calculations for use in fresh-water fluxes. The ocean flux calculations result from the water having a high mass proportion of salt, which, upon droplet formation in breaking waves, will at least partially evaporate, leaving particles of sea-salt. The authors made use of this parameterization in a series of sensitivity studies which suggest that the resulting fluxes might be significant on a local scale – but they do this without justifying the flux parameterization they use as the source of that information. It may be that all that is required here is a more in-depth discussion of the findings of Slade et al (2010) – wherein a surface source of fine particulate matter from the Great Lakes was postulated to explain the observations in that paper. I'm very concerned that this paper is suggesting possible importance of a process using a flux algorithm that may not be appropriate for

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the concentration and types of species that are observed.

At the same time, if this justification can be provided, and especially if the composition of the resulting aerosols can be discussed, I think that the paper would be publishable in ACP. The contribution may be an important one, given that changes in fine particle number up to 10% have been identified.

I'm calling it major revisions for now, pending a better justification for the flux parameterization used.

Major comments:

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

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)?

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.?

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?

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.

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.

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 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.

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.

Minor comments:

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

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".

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

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the parameterization being examined in the current work?

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.

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

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 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?

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?

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.

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

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

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.

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.

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