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Atmospheric Chemistry and Physics Discussions

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

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Response to Anonymous Referee #3

We thank the reviewer for her/his fair and constructive comments. We have revised the manuscript based on the reviewer's concerns and believe the paper is improved as a result. A general response to the major issues raised by both reviewers is being submitted separately as a General Comment. This response will address the concerns raised specifically by this reviewer, in the order presented in the review.

From General Comments

"I think that showing how well the model prediction compares with observations with Slade et al (2010) (and others if available) will help to clear these concerns."

The reviewer makes a good point that validity of the approximations can be evaluated by comparing the results of this modeling study to the observations of Slade et al. (2010). Unfortunately, a direct comparison is not possible because the selected modeling period (summer 2004) did not overlap with the observational period (summer 2009). This choice of modeling period was made for practical reasons- WRF results from the 2004 summer were readily accessible for the continental US and that WRF-Chem has been previously evaluated during that period (McKeen et al., 2007).

While a direct comparison is not possible, we can make a qualitative comparison of the two studies. Figure 2 of Slade et al. (2010) indicates that particle enhancements over the lake were ∼300 cm-3 at altitudes of ∼200m, when the surface wind was ∼5 m/s. Figure 6b of this study suggests enhancements of ∼100 cm-3 at an altitude of 200 m when 22-m winds were ∼10 m/s. These results are qualitatively comparable, and if anything they suggest that the observed enhancement was larger than what the model results indicate. We also note that the aircraft measurements of Slade et al. (2010) are only available for altitudes above ∼200 m, and thus do not include the surface layer where the model predicts the most significant impacts.

We have revised the manuscript to include a qualitative comparison with the Slade et al (2010) results.

Specific Comments

1. Page 16214; line 23 to 25 – This sentence is confusing or perhaps wrong. The two-moment sectional method in Tzivion et al. (1989), which is on the reference list, is not for aerosol coagulation but for condensation and evaporation. However, there is the coagulation paper by Tzivion et al (1987) – see below. Perhaps you need to add another Tzivion et al. (1987) for coagulation? Tzivion, S., Feingold, G. and Levin, Z. (1987). An Efficient Numerical-Solution to the Stochastic Collection Equation. J Atmos Sci 44:3139-3149.

- We thank the reviewer for catching this error in our manuscript. MOSAIC models the

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transfer of particle mass and number between bins due to condensation/ evaporation using either the moving center algorithm of Jacobson (1997) or the two-moment algorithm of Simmel and Wurzler (2006), which is built on the method of Tzivion et al (1989); the latter is used for this study. MOSAIC does not use Tzivion et al (1987) for coagulation. We have revised the manuscript to correct the misstatement in the original manuscript.

2. Page 16215; line 8 – Does MOSAIC model uses the two-moment sectional method by Tzivion et al. (1987) or Jacobson et al. (1994)?

- MOSAIC uses Jacobson et al (1994) for model coagulation, as stated in the manuscript.

3. Page 16215; line 12 (and Page 16226; line 10 and Page 16227;line 3 to 10) – Several questions here.

a. “newly-formed particles are assumed to have the smallest. . .”. What do you mean by newly-formed particles? I guess the particles formed from nucleated particles that survive to the smallest particles in the model?

- Yes, we refer to particles that nucleate in the atmosphere and grow, primarily by condensation, to 40nm. The model does not actively simulate this process- the nucleation parameterization simply adds a number of new particles to the smallest size bin. We have added text in sections 2.1, 3.1, and 4 to make clarify this point.

b. I don’t understand how the WRF-chem model accounts for the growth and loss of freshly-nucleated particles up to Dp âĹij 3-10 nm? Please add the description of the parameterization. However, new particle formation rates are very different between 3nm and 10 nm. How come it is from 3nm to 10nm?

- Our original text was not clear. The model directly applies the binary nucleation rate of Wexler et al (1994) as the new particle formation rate at Dp = 40nm. There was no adjustment to growth to 3, 10 or 40nm. We have revised the manuscript in sections

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2.1, 3.1, and 4 to clarify how new particle formation in WRF-Chem is modeled.

c. If new particle formation rates at 3nm or 10nm assumed to be at 40 nm, this might overestimate the contribution of the nucleation to number flux, although the real impact might be more complicated (for example, it can affects the frequency and magnitude of nucleation event due to the overprediction of the condensational sink by the "newly-formed particles"). Nevertheless, is the binary nucleation good enough for that region? If not, how about doing additional simulations using the activation nucleation (i.e. empirical parameterization)?

- We strongly agree with the reviewer that the sensitivity of WRF-Chem and other models to the choice of nucleation scheme is a ripe topic for further analysis. However, we feel that such analysis is outside the scope of the current study. The nucleation parameterization selected is used frequently for simulations within the continental US, and thus was a reasonable choice given that our goal was to evaluate the potential impacts of the proposed lake-surface particle source. We discussed the weakness of the model in simulating new particle formation (page 16227, lines 3-11) in the original manuscript. To further stress the point we have now added more text in that section to discuss the potential non-linear effects on particle number concentrations arising from how WRF-Chem models new particle formation.

4. Page 16219; line 9 to 14 (and Page 16226; line 6 to 20) – I agree that F10nm is a better upper bound estimate of total number flux (as mentioned in Page 16217). However, I do have some concerns on how the number flux predicted by F10 nm is emitted at 40nm. Although particles in between 10 nm < Dp < 40 nm are removed quickly by coagulation (and also by dry deposition), applying the same number concentration at 40 nm will overestimate the condensational sink and can suppress nucleation event. Do you have any size distribution data on F10 nm account for number flux larger than 40 nm? Knowing that Geever et al (2005) is for ocean water, please explain why the parameterization by Geever et al (2005) is chosen as the lake emission among available sea-salt emission parameterizations.

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- Our motivation for selecting the Geever et al. (2005) parameterization was discussed in section 2.3 of the original manuscript, but based on the reviewers' concerns we have revised the manuscript to make our reasoning on this point more clear. We also already discussed the potential non-linear impact of this choice on total particle number concentrations (page 16226, lines 11-16).

We specifically chose a parameterization that focused on number emissions only. While the size distribution of emitted particles would be drastically different for freshwater vs. marine conditions, the number of particles emitted should not change much. Our reasoning is that the number of particles is linked directly to the number of droplets mechanically generated at the water surface, by bubbles bursting or waves breaking. Each droplet results in one particle. The residual particles left behind by an evaporating salt water droplet would be considerably larger than the residual from a freshwater droplet, but in each case there would be a residual particle.

For this reason we feel justified in using a number-based marine emission parameterization as a first approximation of what the freshwater particle emissions would be. Within that constraint the decision to use the Geever et al. (2005) parameterization was somewhat arbitrary. Still, it should again be emphasized that our fundamental goal here was not to determine definitively the exact contributions of freshwater particles to the regional aerosol, but rather to evaluate whether the contribution was potentially significant enough to merit more intensive observational study. With respect to that goal, we feel our methodology for this study is justified.

This point was also raised by another reviewer. We encourage readers to also refer to the response to reviewer #1 and to our General Comment.

5. Page 16219; line 15 to 18– I think that the test simulations with/without nucleation are interesting and also meaningful to compare it with lake emission, but I do not understand why you want to do the simulations with/without coagulation and dry deposition. Can you please explain more why these tests are needed or interesting?

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- Given the underlying concerns regarding how nanoparticle processing is handled within WRF-Chem, we thought that we should consider the sensitivity of our results to different sources and sinks of particle number- nucleation, coagulation, and dry deposition. As expected, nucleation turned out to be the most significant of these processes, but we have also reported the other results for completeness.

6. Figure 4 (b) – The diurnal pattern before 20 GMT can be expected as the number fraction (del N%) is almost proportional to the change in the lake aerosol flux at the fixed other emissions or background aerosol concentrations. However, after 20 GMT, it shows the opposite trend between two. Can you explain what happens after 20 GMT?

- The percent change in the number concentration is of course sensitive to the rate of particle emission from the lake surface, but it is also quite sensitive to the background particle concentration. The percent change in aerosol number concentration is calculated as (delN/N) * 100%. Throughout most of the time period shown in Figure 4(b), the emissions and background concentrations trend in opposite directions, such that the percent increase due to surface emissions is enhanced (i.e., delN increases and N simultaneously decreases). This is the proportional trend the reviewer describes. After 20 GMT, both the emissions (and hence delN) and the background concentration (N) are decreasing. In this instance, the net effect of these conflicting trends is for the percent change to increase, as seen in the figure.

Technical Corrections

1. Page 16215; line 8 – MOSIAC should be MOSAIC.

- Good catch. This is fixed in the revised manuscript.

2. Figure 6(a) and Figure 8 (b) and Figure 10, the simulation name above each figure does not contain “0”, unlike Figure 3 (a) and (b) where the simulation name in the figure contains “0” such as BASE0 and LAKE0-BASE0. Please make consistent simulation name throughout the manuscript.

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- We appreciate the reviewer's attention to details in catching these errors. They have been fixed in the revised manuscript.

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