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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.

Anonymous Referee #2

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General comment

This work investigates the regional scale impacts of the potential aerosol emissions from freshwater bodies using WRF-chem model that includes the MOSAIC aerosol module. Given that very little is known about ultrafine particle emissions from freshwater bodies, this study is a timely contribution to the literature. I initially had some major concerns with how their model dealt with nucleation and lake emission number flux parameterization, but these issues were addressed to some extent in the discussion section. However, I still have some concerns on the discussion about the sensitivity of the findings to current model setup (e.g., lowest size boundary, insufficient representation of nucleation, etc) (see specific comments), and the long list of the required model improvements gives the impression that this WRF-chem model is insufficient for this

study. 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. Once these concerns and the specific comments below have been addressed, I recommend that the manuscript be published.

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.

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

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? b. I don't understand how the WRF-chem model accounts for the growth and loss of freshly-nucleated particles up to Dp $\sim$ 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? 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)?

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.

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?

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?

Technical corrections

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

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16207, 2011.