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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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General Comment

We thank both reviewers for their helpful comments. We have revised the manuscript based on their suggestions and believe that the final paper will be significantly improved as a result of their contributions.

One of the major concerns raised in the reviews was whether a parameterization based on measurements of marine sea salt emissions could be reasonably applied to the potential freshwater sources that are the subject of this study. That question cannot be definitively answered without measurements targeting freshwater particles, and this work is not intended to imply otherwise. Our effort here builds on the earlier work

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Interactive Discussion

Discussion Paper

of Slade et al. (2010), who observed an interesting phenomenon and proposed a plausible cause- that the enhanced nanoparticle concentrations over the Great Lakes were a result of wind-driven surface emissions. The most obvious next steps would be to make further measurements to verify and quantitatively characterize this potential new source, and ultimately to build a new freshwater source parameterization based on direct observation. However, acquiring enough data to develop a freshwater particle emission parameterization would demand substantial additional resources, and it was not clear from the earlier work whether that cost was justifiable. Our goal in this work was to determine whether the proposed lake-surface source had the potential to be a significant source of particle number on a regional scale. We believe that we have now demonstrated that this potential does exist, and thus that additional work is merited to quantify freshwater emissions of particles with ambient measurements.

Our purpose in this work was not to suggest that the specific parameterization that we chose is the optimal one, and we would certainly not advocate that it be used to model lake emissions operationally. Rather, our goal for this study was to provide a reasonable estimate of the potential for lake-derived particles to impact the regional aerosol population, while working under the constraint that direct measurements of the emission rates of lake-derived particles have never been done. Given our goal, we believe that our methodology for the study was entirely appropriate. Methodological choices were consistently made to highlight the potential contributions from the lake source without straying from conditions that were physically reasonable. Our justifications for these choices are discussed in detail in the manuscript, but some will be highlighted again here since these choices were the focus of many of the reviewers' comments.

- As suggested by Slade et al. (2010), it was clear from the initial observations that the biggest potential impacts would be on particle number concentration, rather than aerosol mass. The observed particles are too small to make significant mass contributions. Thus our efforts in this work focused on particle number concentrations, which are a key aerosol property for estimating aerosol cloud-forming potential, among other

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things. In keeping with this focus, we chose a sea-salt emissions parameterization developed by Geever et al. (2005). This parameterization focuses on particle number emissions. We further chose the version of that parameterization that included particles as small at 10 nm (rather than 100 nm), in order to get an upper bound estimate on the number of particles emitted. This choice might be disputed, but it was consistent with our goal of determining the potential impacts of this source in the absence of direct measurements.

- Our decision to use the Geever et al. (2005) parameterization as opposed to some others was made specifically because we were attempting to use a salt-water parameterization for freshwater conditions. Many parameterizations specify emissions with an explicit size-dependence (cf. Lewis and Schwartz, 2004), but the Geever et al. (2005) parameterization does not. As we stated in the manuscript (section 2.3), our assumption was that the same wave-breaking mechanism is the cause of particle generation in marine and freshwater environments. Given that, it was reasonable to assume as an initial estimate that while the size distribution of emitted particles would shift dramatically between marine and freshwater environments, the number of particles would not. To a good approximation, the number of particles emitted is the same as the number of droplets produced mechanically at the water surface. Droplets are produced by bubble-bursting or wave-breaking, after which the water evaporates to leave residual particles as they move away from the surface. We found it reasonable to assume as a first approximation that the number of droplets released mechanically at the surface will not change from marine to freshwater conditions. And for each droplet that is released, one residual particle is produced upon evaporation, though of course the residual particles will be much smaller under freshwater conditions. If both of these assumptions are accepted, then it holds that a number-based parameterization for marine particle emissions would also hold for freshwater particle emissions.

- While we felt that the arguments above justified our selection of parameterization for the wind-driven particle emissions from freshwater surfaces, we also realized that

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the small size of the emitted particles might affect the aerosol population dynamics in unanticipated ways. Mechanisms that we identified as potentially impacted included nucleation (i.e., atmospheric new particle formation), coagulation, and dry deposition. We examined the sensitivity of the results to these processes by including simulations with the individual processes turned off. Ultimately though, we cannot fully explore these problems without a model aerosol scheme that includes better size resolution for nucleation mode particles. This same limitation exists for any source of atmospheric nanoparticles, including primary combustion emissions and atmospheric new particle formation.

These points had been raised already in the original manuscript, but given the reviewers' concerns we have clarified them further in the revised version of the manuscript. We have also addressed the additional concerns raised by each reviewer individually in separately submitted responses. With these changes we hope that the editors of ACP will find that this paper is a worthy contribution to the journal. We have not determined conclusively that the wind-driven freshwater particle source is real or that it has a major impact on the regional aerosol, but this was never our intent. We have demonstrated that the potential for such impacts is sufficiently strong to merit additional work, and we look forward to more observations and modeling work to better characterize this phenomenon.

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