

Reply on RC1:

In this article, Harb and Foroutan describe a measurement and complementary modeling study of lake spray emissions focused on the Great Lakes region. The measurement section of the article describes the development of a lake spray source function using a marine aerosol reference tank, while the modeling section uses the Community Multiscale Air Quality model to simulate the lake spray impacts on aerosol number and mass concentrations during a period of strong winds. Generally, I find the measurement section of the article a useful development in the field of lake spray emissions but find the modeling section incomplete. Please see the specific comments below.

We thank the reviewer for taking the time to review our manuscript. Our reply to each comment is provided below.

Major Comments:

1) CMAQ model: Despite being a widely-used model for research and policy, I don't believe that CMAQ is the appropriate tool to simulate the potential cloud impacts from lake spray aerosols. The lack of aerosol-cloud interactions in the version of CMAQ used in this study means that the simulations are limited to the prediction of aerosol number concentrations without any information on the associated changes of cloud condensation nuclei or ice nuclei on cloud properties. Quantifying the impacts of this aerosol source on cloud properties requires an online-coupled model.

We agree with the reviewer that a more comprehensive evaluation of the impact of this aerosol source on cloud processes requires the use of an online-coupled model. However, the main goal of this manuscript was to report an experimentally-developed LSA emission parameterization and provide a preliminary estimate of freshwater emissions impacts on regional aerosol loading in the Great Lakes region. Therefore, we believe that implementing the LSA parameterization in an online-coupled model is beyond the scope of this work but is definitely worth considering in future studies that are revolved around the impacts of LSAs on cloud properties. In the original submission, we mentioned that future studies should consider LSA-cloud interactions (please see lines 545-548 in the revised manuscript). We have also mentioned this limitation when discussing the impacts of LSAs on the aerosol number concentrations in the cloud layer (i.e., Figs. 11 and 12) (lines 473-475 in the revised manuscript).

[Revised text as it appears in the manuscript \(lines 473-475\):](#)

[However, the reader should keep in mind that a more comprehensive assessment of the potential impact of LSAs on cloud processes, specifically the associated changes in cloud condensation nuclei \(CCN\) and ice nucleating particles \(INPs\), requires the use of an online-coupled model \(Zhang, 2008\).](#)

2) CMAQ model configuration: I found little benefit from the SEA simulation to the study, and was surprised that it was included after the description of the lake spray source function having substantially lower emission fluxes than the sea spray source function. If CMAQ continues to be used in this study (see comment above), I'd strongly suggest that the SEA simulation be replaced

with a simulation incorporating chemical speciation of the lake spray aerosol so that the impacts to regional atmospheric chemistry be quantified.

One of the main takeaways from this work was that using a sea spray source function to represent freshwater emissions will lead to a considerable overestimation of LSA emissions (please see lines 538-540 in the revised manuscript). Therefore, we have considered the SEA simulation to highlight this overestimation. Moreover, we have already provided the rationale behind our choice of each emission scenario in the original submission (please see lines 243-246 in the revised manuscript).

We agree with the reviewer that including LSA chemical speciation is essential to better quantify the impact of LSA emissions on regional atmospheric chemistry. However, the main goal of the modeling section was to quantify the contribution of freshwater emissions to regional aerosol loading and compare it to anthropogenic emissions in the region. It is obviously true that the inclusion of LSA chemical speciation will result in a more realistic simulation, however, we believe that using inert LSA particles provides a good preliminary idea about these impacts. In the original submission, we have already discussed the limitations brought about by not including LSA chemistry in light of the study goals (please see lines 406-413 in the revised manuscript) and suggested including LSA chemical speciation in future studies (please see lines 544-548 in the revised manuscript).

3) CMAQ model evaluation: When discussing the model results, it's unclear whether including this emission source improves model performance. I'd suggest that PM2.5 and PM10 observations from regulatory and IMPROVE sites in the region be compared to the model simulations. If chemical speciation of the lake spray aerosol is included (see comment above), I'd also suggest that PM2.5 speciation from the IMPROVE and CSN sites in the region be used in the evaluation.

We thank the reviewer for this suggestion. We think that without the inclusion of the LSA chemical speciation (please see our reply to comment 2 above), such a comparison is not very useful. Therefore, we believe that such an analysis would be suitable for future modeling studies that include LSA chemical speciation (please see lines 548-549 in the conclusion section of the revised manuscript).

Revised text as it appears in the manuscript (lines 548-549):

The inclusion of LSA chemical speciation in the simulation will also allow for evaluating model results against PM observations from regulatory air-quality networks in the region.

4) Typos:

Page 2, line 49: Should be "These distinct..."

Page 16, line 371: Should be "as shown in Fig. 4b..."

Figure 8 caption: Should be "concentrations"

Figure 9 caption: Should be "concentrations"

Figure 10 caption: Should be "concentrations"

Page 22, line 470: Should be "Great Lakes..."

Page 22, lines 478-479: Should be "further north or south..."

Figure 11 caption: Should be "from north to south..."

Page 25, line 514: Should be "impacts of these emissions..."

We thank the reviewer for catching these typos. They were corrected in the revised manuscript.