Supporting Information for "Lake spray aerosol emissions alter nitrogen partitioning in the Great Lakes region"

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Contents of this file

Text S1. In Table S1 the monthly mean bias of the NEI+Lake and NEI simulations compared to five IMPROVE sites close to the Great Lakes is shown for November 2015. For SO_4, NO_3 and TOC the bias is reduced in NEI+Lake compared to NEI, except for TOC at EGBE. For Ca, Na and Cl the change in bias is more diverse. At BOWA and MKGO the bias is reduced in NEI+Lake for all three species and for Cl at EGBE, while at SENE, ISLE and EGBE the bias increases. Because the simulations have a relatively

coarse horizontal resolution (12x12 km) a comparison to individual stations is difficult. Therefore we also compute the average bias for the stations in the vicinity of the Great Lakes. The ISLE station is excluded from this average as the location of the ISLE station in the model simulations is over lake (not over land). This average bias is reduced in the NEI+Lake simulation compared to NEI for all investigated aerosol species by 0.02 to 0.08 μg (kg dry air)$^{-1}$ except Cl, which turned from a small underestimation in NEI to a small overestimation in the NEI+Lake simulation and the bias increases by 0.003 μg (kg dry air)$^{-1}$.

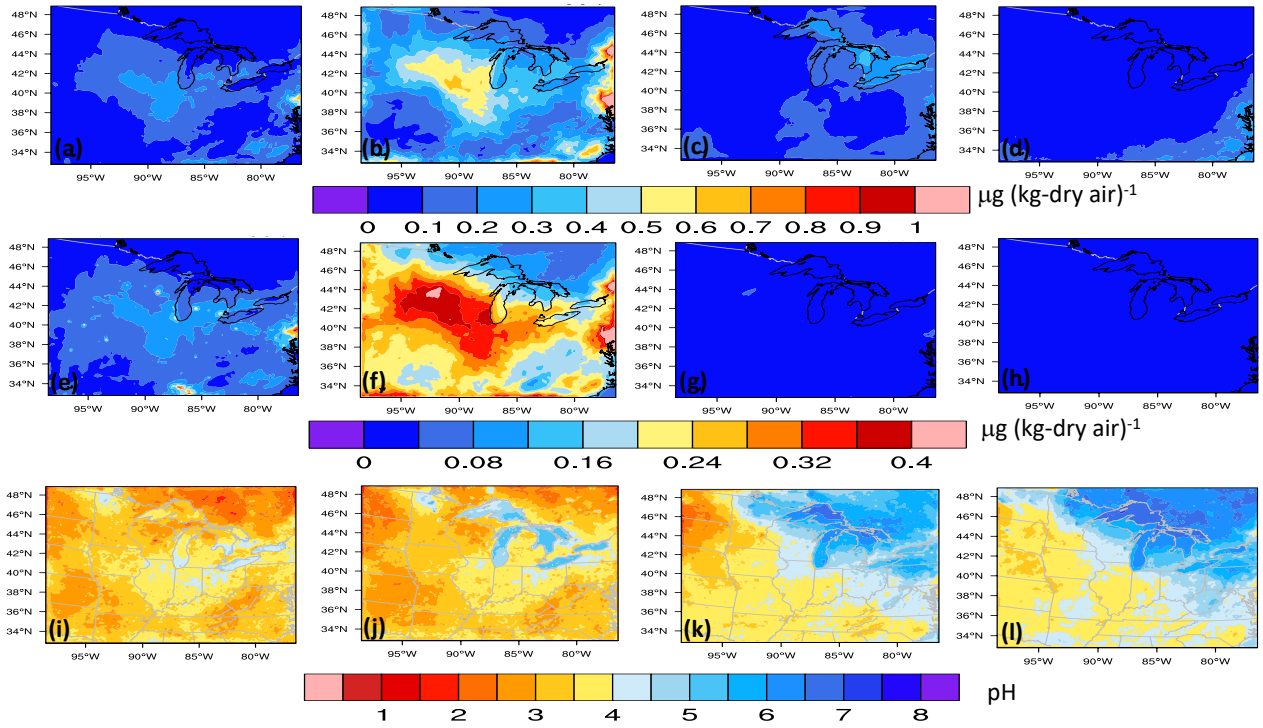

Figure S1. NEI+Lake simulation of particulate NO_3^- (a−d), NH_4^+ (e−h) concentrations (μg (kg dry air)$^{-1}$), and pH (i−l) for the four MOSAIC size bins (0.039−0.156 (Bin 1), 0.156−0.625 (Bin 2), 0.625−2.5 (Bin 3) and 2.5−10.0 (Bin 4) μm). Monthly average values for the surface model layer in November 2015 are shown for the whole simulation domain.

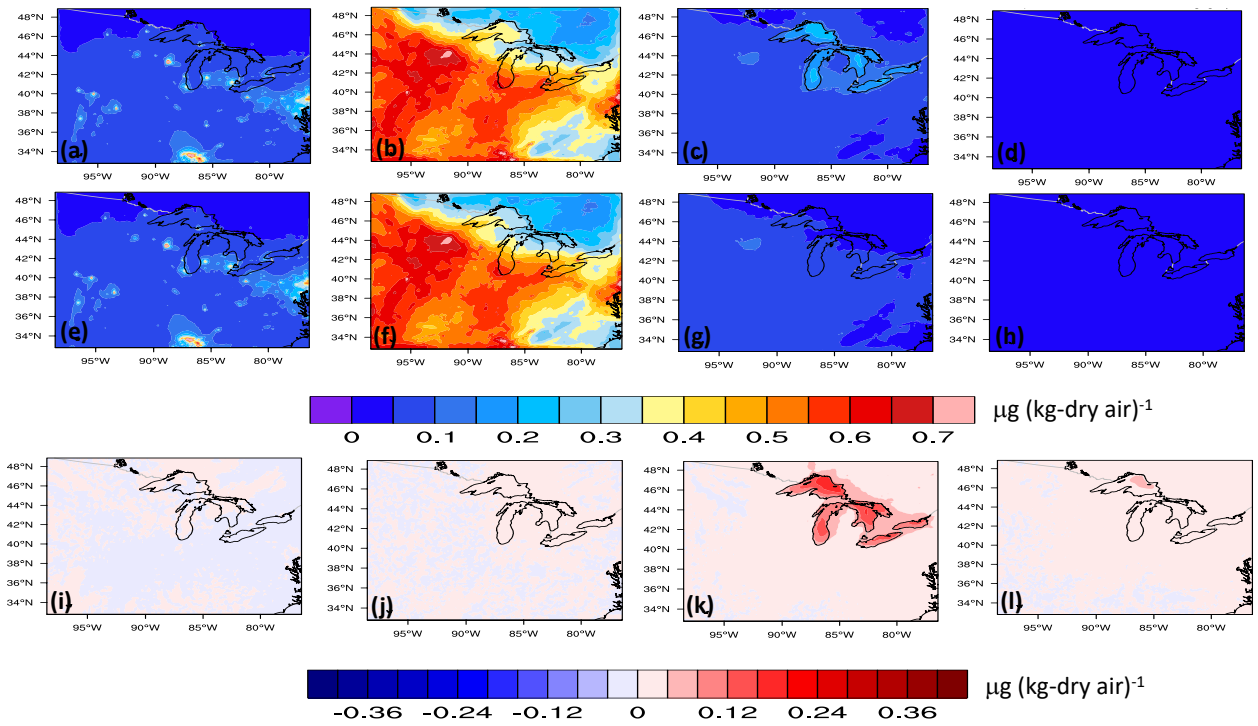

Figure S2. Particulate SO_4^{2-} in the NEI+Lake (a−d) and NEI (e−h) simulations. The difference between NEI+Lake and NEI simulations of particulate SO_4^{2-} concentrations (μg (kg dry air)$^{-1}$) (i−l). Values represent monthly average mixing ratios for the surface model layer in November 2015 for the four MOSAIC size bins for the whole simulation domain.

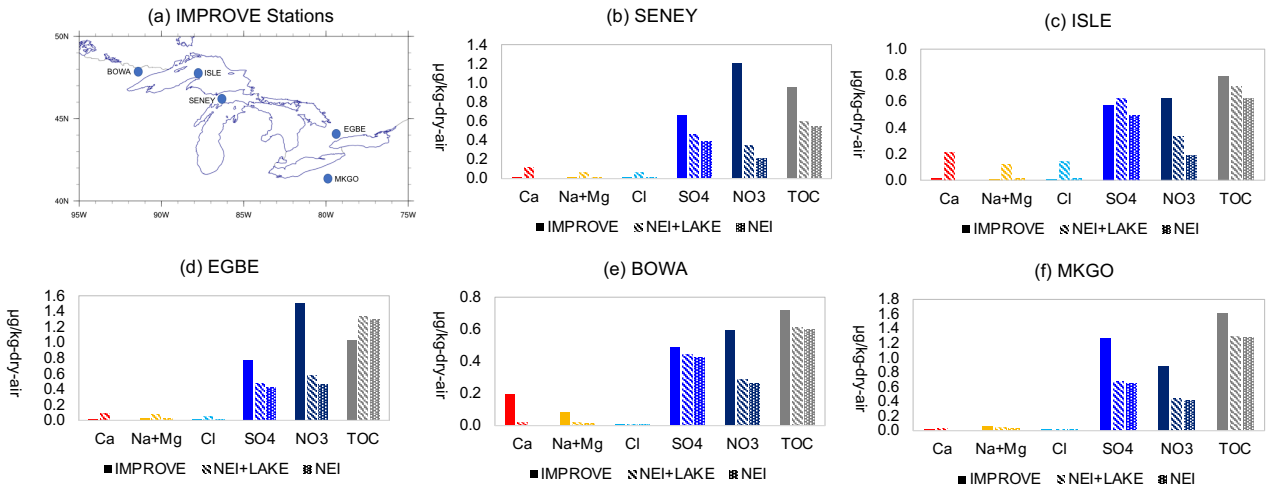

Figure S3. (a) Locations of selected IMPROVE sites for comparison with model output in November 2015. (b−f) monthly mean mass mixing ratios at the selected IMPROVE sites, NEI+LAKE, and NEI simulations. For the simulation data the nearest model grid points to the IMPROVE sites are chosen for the comparison. Based on the availability of data for November 2015 five stations close to the Great Lakes are chosen. Note that the ISLE station is on the island for IMPROVE data, but it is over the lake in simulations.

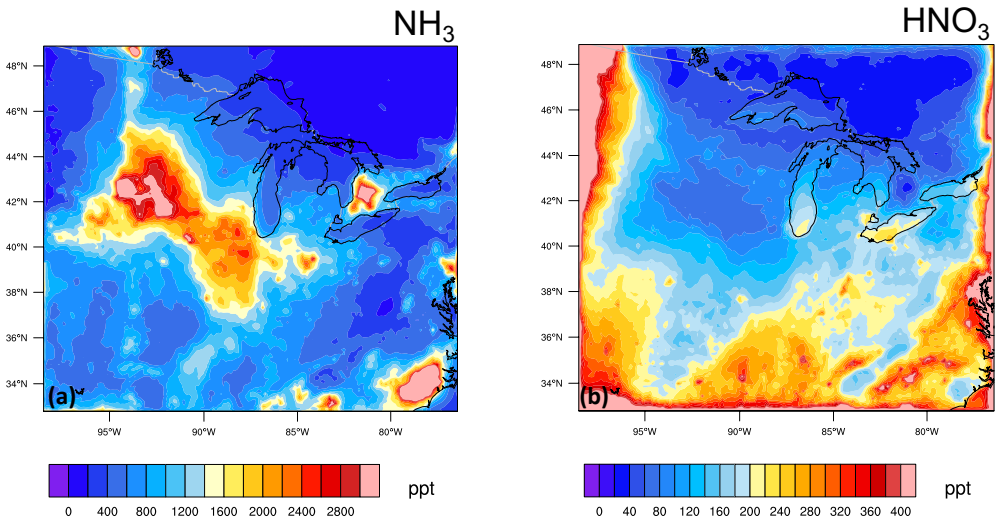

Figure S4. (a) NH_3 and (b) HNO_3 for the NEI+LAKE simulation. Values represent monthly average mixing ratios for the surface model layer in November 2015 for the whole simulation domain.

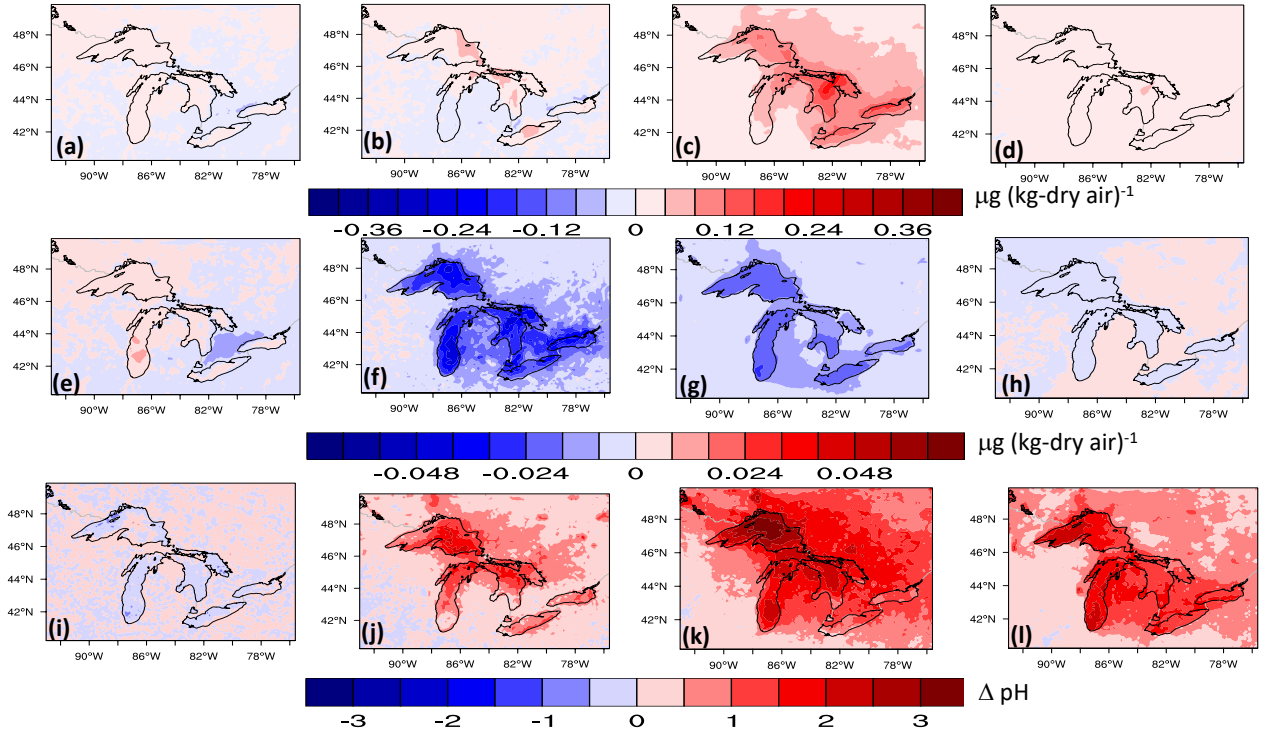

Figure S5. The difference between NEI+Lake$_{\text{no het}}$ and NEI$_{\text{no het}}$ simulations (NEI+Lake$_{\text{no het}}$ - NEI$_{\text{no het}}$) with the heterogeneous calcium reactions (Equations 1 and 2) turned off of particulate NO_3^- (a–d), NH_4^+ (e–h) concentrations (μg (kg dry air)$^{-1}$), and pH (i–l) for the four MOSAIC bins (0.039–0.156 (Bin 1; a,e,i), 0.156–0.625 (Bin 2; b,f,j), 0.625–2.5 (Bin 3; c,g,k) and 2.5–10.0 (Bin 4; d,h,l) μm). Changes represent the difference in monthly average mixing ratios for the surface model layer in November 2015.

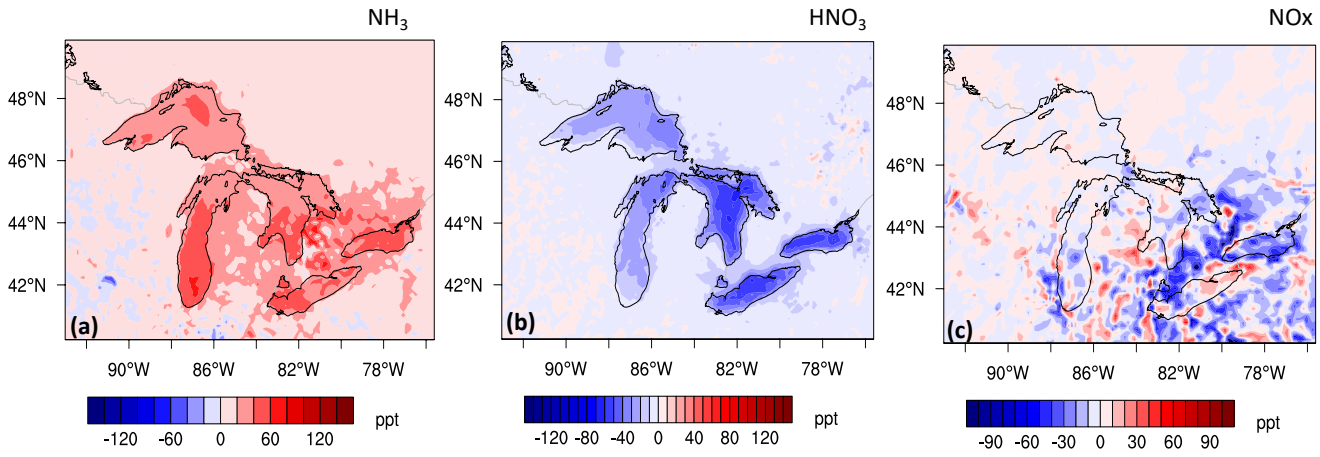

Figure S6. The difference between Lake+$NEI_{no\ het}$ and $NEI_{no\ het}$ simulations ($NEI+Lake_{no\ het}$ - $NEI_{no\ het}$) with the heterogeneous calcium reactions (Equations 1 and 2) turned off of surface model layer mixing ratios (ppt) for (a) NH_3, (b) HNO_3, and (c) NOx in November 2015.

Table S1. Monthly mean difference between the surface model layer concentrations of several aerosol species of the NEI+Lake and NEI simulations and co-located IMPROVE site data concentrations for November 2015. Since IMPROVE data is for fine mode aerosols, the sum over the first three aerosol size bins is used for NEI+Lake and NEI simulations. The simulation with the lowest bias is shown in bold.

NEI+Lake - IMPROVE	SENE	ISLE	EGBE	BOWA	MKGO	Average[a]
ΔCa [μg (kg dry air)$^{-1}$]	0.10	0.20	0.07	**-0.18**	**0.01**	**0.00**
ΔNa [μg (kg dry air)$^{-1}$]	0.06	0.11	0.05	**-0.07**	**-0.01**	**0.01**
ΔCl [μg (kg dry air)$^{-1}$]	0.05	0.14	**0.02**	**0.00**	**0.00**	0.02
ΔSO_4 [μg (kg dry air)$^{-1}$]	**-0.20**	**0.05**	**-0.29**	**-0.05**	**-0.59**	**-0.28**
ΔNO_3 [μg (kg dry air)$^{-1}$]	**-0.86**	**-0.29**	**-0.93**	**-0.30**	**-0.44**	**-0.63**
ΔTOC [μg (kg dry air)$^{-1}$]	**-0.36**	**-0.08**	0.30	**-0.11**	**-0.32**	**-0.12**
NEI - IMPROVE	SENE	ISLE	EGBE	BOWA	MKGO	Average[a]
ΔCa [μg (kg dry air)$^{-1}$]	**-0.01**	**-0.01**	**-0.02**	-0.20	-0.02	-0.06
ΔNa [μg (kg dry air)$^{-1}$]	**0.00**	**0.00**	**0.00**	-0.07	-0.03	-0.03
ΔCl [μg (kg dry air)$^{-1}$]	**-0.01**	**0.00**	-0.02	0.00	-0.02	**-0.01**
ΔSO_4 [μg (kg dry air)$^{-1}$]	-0.27	-0.08	-0.34	-0.06	-0.61	-0.32
ΔNO_3 [μg (kg dry air)$^{-1}$]	-1.00	-0.44	-1.04	-0.33	-0.48	-0.71
ΔTOC [μg (kg dry air)$^{-1}$]	-0.42	-0.18	**0.26**	-0.12	-0.34	-0.16

[a] The average is taken over the stations SENE, EGBE, BOWA and MKGO. The ISLE station was excluded because the values of the simulations are over the lake and not over the island and therefore less representative.