

Supplemental information

Aerosol acidity, the main driver of aerosol hygroscopicity and $\text{H}_2\text{O}_{\text{ptcl}}$ concentration is driven by sulfate (Figure S1.). Other contributors to aerosol acidity (NO_3 , Cl) do not contribute substantially and are not visible when plotted at the same scale in Figure S1 for this simulation. The total aerosol free acidity in Figure 5 of the main text ($\sim 50 \text{ nmol m}^{-3}$) is less than the sulfate acidity ($\sim 200 \text{ nmol m}^{-3}$) due to the neutralization of ammonium.

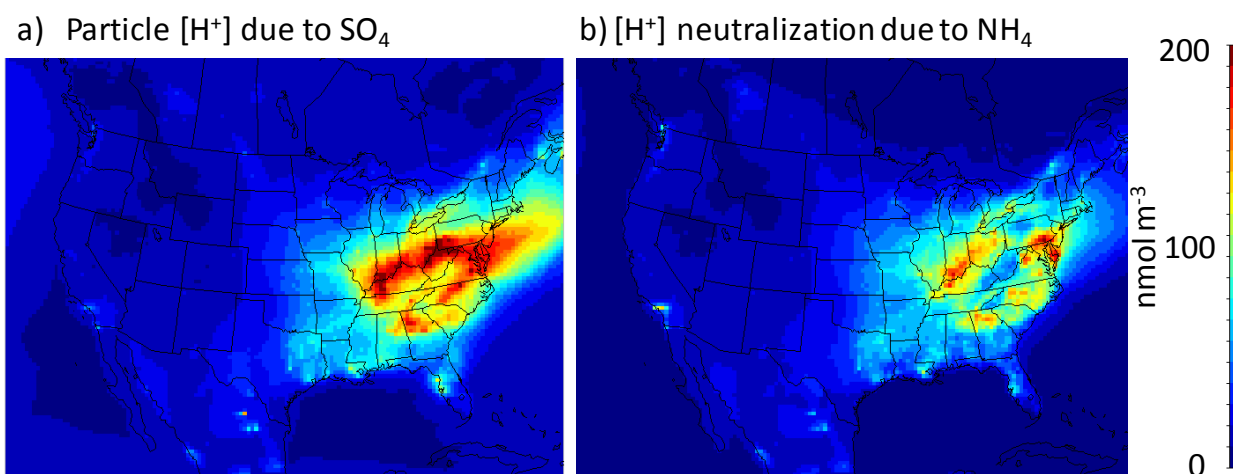


Figure S1. Particle phase H^+ concentration due to sulfate (a) and the neutralizing power of ammonium (b). Values averaged for July 2003.

When anthropogenic emissions are removed from emissions as described in detail by Carlton et al. (2010), aerosol acidity for that 3 week period is lower than in the base case (i.e., when all controllable and non-controllable emissions are present). This suggests that $\text{H}_2\text{O}_{\text{ptcl}}$ in the U.S. is largely anthropogenic and controllable.

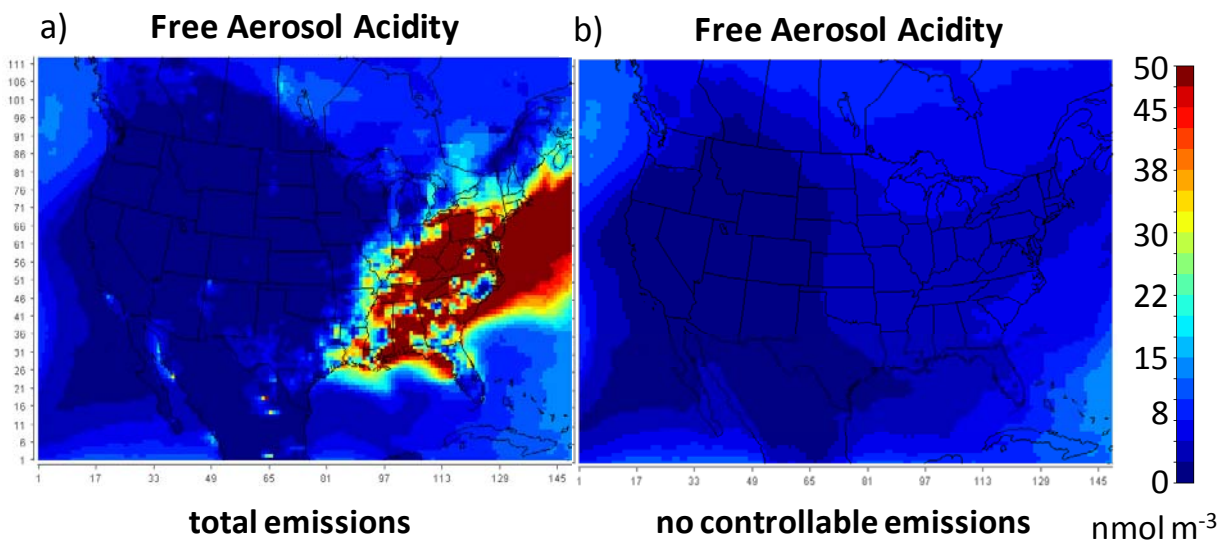


Figure S2. Free aerosol acidity in the base case CMAQ simulation, when all emissions are present (a) and when controllable, anthropogenic emissions are excluded (b). Values averaged for August 18 – September 4, 2003.

Note that modeling details for the work described here and in Carlton et al., (2010) are the same, with the exception of the model time period July 2003 (here) versus August 15 – September 4, 2003 (Carlton et al., 2010).

References

Carlton, A. G., Pinder, R. W., Bhave, P. V., and Pouliot, G. A.: To What Extent Can Biogenic SOA be Controlled?, *Environ. Sci. Technol.*, 44, 3376-3380, 2010.