

Comments from Referee#1

This paper presents a modeling study of mercury scavenging in convective clouds. The detailed processes studied are outside of my area of expertise, but that said, I do have comments regarding the overall study design. Mainly, I am very puzzled why the authors chose mercury to study in this regard. The model results presented are completely unverified and unverifiable with current technology. For example, many of the assumptions made, such as the vertical distribution of reactive mercury, are completely unknown. I understand the value of a model where some new and verifiable prediction is made, but it is really hard to see exactly what observations could be made to verify these results. Differences in mercury deposition alone between north and south sites, would not provide the necessary information. If there is a straight forward way to verify this model, then it is essential that the authors identify this. If the model cannot be readily verified, then this is not really a scientific result and the paper should not be published. My suggestion for the authors would be to apply the same model to a compound or compounds that are also readily scavenged and can be observed. For example there is a wealth of data on HNO₃ from aircraft studies. Why not apply this model to HNO₃ and use the existing observations to see if it makes sense.

Our response:

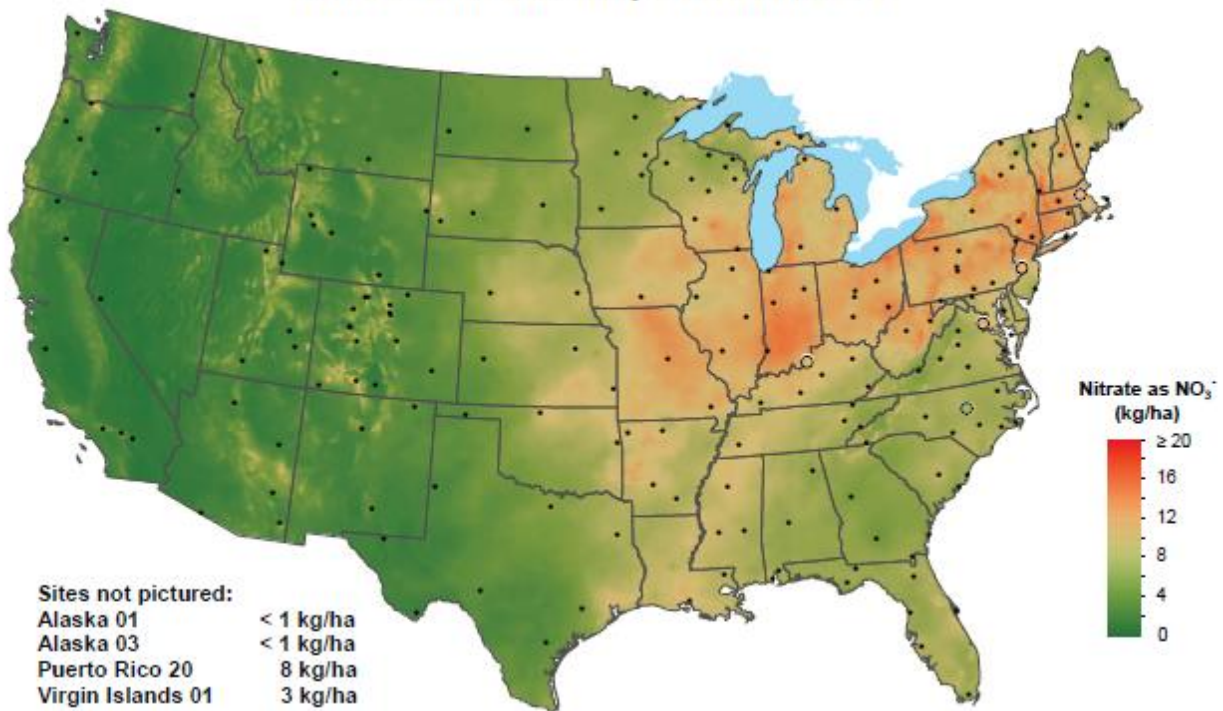
Our primary goal is to understand processes that could explain high mercury wet deposition that is observed in the Southeast US, especially along the Gulf Coast and Puerto Rico, and is disproportionately higher than emissions or concentrations in the boundary layer in this region. US mercury emissions peak in the Ohio River Valley and northeast, but wet deposition in this region is generally lower than along the Southeast. By contrast, nitrate and sulfate deposition occurs over emission regions or slightly downwind. Thus, while we agree that additional observational and modeling analysis of HNO₃ and nitrate concentrations and deposition would be valuable, these species are not direct analogues for mercury. Because of the human and environmental health consequences of mercury exposure, we believe our study of mercury deposition does not need additional analysis of nitrate. We apologize that these differences between mercury and nitrate deposition and reasons for excluding nitrate (and sulfate) from our analysis were not clear in our initial manuscript. We have clarified these points in the introduction and conclusion.

The scope of the present study is framed to examine the viability of the following hypotheses: 1) Deep convection is more efficient at removing atmospheric mercury since they access and process mercury from a deeper layer of the atmospheric column; 2) Efficiency of mercury removal by convective storms is dependent on thunderstorm morphology and thus environmental characteristics.

High-altitude observations of atmospheric mercury are indeed limited, as the reviewer says. However, aircraft observations in many locations [Sillman et al., 2007; Talbot et al., 2008; Slemr et al. 2009; Lyman and Jaffe, 2012], including Florida [Sillman et al. 2007], have established that Hg(II) mixing ratios generally increase in the middle and upper troposphere. The GEOS-Chem model reproduces these observed vertical gradients in the troposphere, including their changes with latitude and season [Holmes et al. 2010], which justifies using the seasonal mean concentrations from GEOS-Chem as an initial condition for the RAMS model. While the limited observations prevent us from comparing the RAMS model results against specific individual rain events, we instead we seek to explain broad regional differences and statistical differences between storm types. Our modeling results will further be tested by the upcoming Nitrogen, Oxidants, Mercury and Aerosol Distributions, Sources and Sinks (NOMADSS) field aircraft experiment, which will provide extensive vertical profiles and horizontal surveys of Hg(II) and associated trace gases over the over the East and Southeast US. This field experiment was designed to evaluate the sources and processes impacting mercury wet deposition, among other goals. Thus, our results and predictions are testable with current technology and we expect that our mechanistic understanding of scavenging in rain storms, developed through idealized simulations, will be helpful in

interpreting the upcoming field measurements. In the longer term, we hope that cloud microphysical

Nitrate ion wet deposition, 2008



measurements and hydrometeor sampling, using the NSF thunderstorm penetrating aircraft (facility under development) could be used to characterize vertical profiles of mercury in hydrometeors which can be related to thunderstorm morphology. We have modified the manuscript to include the above discussion.

Possibilities for testing the hypotheses are discussed in the discussion section of the manuscript. Specifically:

“It is difficult to confirm this hypothesis using existing databases, however, it will be tested during the upcoming Nitrogen, Oxidants, Mercury and Aerosol Distributions, Sources and Sinks (NOMADSS) field aircraft experiment, which plans to evaluate mercury wet deposition, among other goals, through extensive vertical profiles and horizontal surveys of Hg(II) and associated trace gases over the over the East and Southeast US.” (Section 4)