

General comment of Referee#4

I agree with the comments of Reviewers I and II that the model could/should have been used to model nitric acid entrainment and deposition for which there are lots of ambient data. There is not enough rainfall Hg data collected on an event basis to constrain (or verify) the model. However, I disagree that the paper should not be published without such data. I also do not agree that the authors MUST propose a set of testable hypotheses to go with their model. This can be the focus of future work. The model framework described in this paper may prove useful for modeling the behavior of other scavengable gases and particles in the atmosphere, and is therefore worthy of publication. The subject of this paper, an effort to explain why Hg rainfall deposition is disproportionately higher in the southeastern US while the US emission sources are located much farther north, is a very important topic. Atmospheric deposition of Hg is ultimately responsible for elevated fish Hg levels throughout the SE US and the Gulf of Mexico, yet most of the Hg emission sources are located much farther to the north. It has been proposed that tall convective thunderstorms are responsible for enhancing rainfall Hg deposition in the SE US, and this paper represents an attempt to model why that should be the case. If this model proves to be reliable, then it offers a tool that can be used in to predict how changes in Hg emissions in the US and on a global scale will affect fish Hg levels in the region. This tool could also be used along with estimates of the sources and in-situ formation rates of gaseous oxidized Hg in the atmosphere over the SE US for more accurate source apportionment analyses.

Our response:

The reviewer provides a clear summary of our study goals, which was previously lacking from the conclusions in the previous version of our manuscript. We have restated these study goals in the revised conclusions, paraphrasing from the reviewer, whom we credit in the acknowledgments:

“This study examines why the Southeast US receives more mercury deposition in rain than the Northeast, despite the lower mercury emissions in the Southeast. Since wet deposition contributes to high mercury concentrations in fish, other wildlife, and ultimately humans, understanding the cause of high regional deposition is of concern to environmental agencies. Past studies suggested that tall convective thunderstorms are responsible for the enhanced mercury deposition in the Southeast and our modeling work explores the dynamical and scavenging mechanisms at work in thunderstorms that might explain the regional differences.”

As mentioned in responses to other referee comments nitric acid is not an exact surrogate for mercury.

Specific comments and our responses:

1. p3579, 10: “Small-scale turbulence”

No modification was made for this. The sentence is about “Small-scale turbulent...entrainment”

2. p3579, 15: Subsequent evaporation of cloud droplets

Modified as corrected by the referee.

3. p3580, 6:”in more stable environments.”

Modified as “in a more stable environment.”.

4. p3583, 21: naming convention is used (c2500s10p60_s) before it has been defined on 3584, line 21.

Modified as “(with CAPE=2500 J/kg, SHEAR=10ms⁻¹, and PW=60 mm)”

5. p3585, 22: “whereas it is only 19%...”

We re-phrase the entire sentence as:” ...show that the atmosphere of the Southeast is climatologically more unstable than the Northeast, with ~65% of the soundings having CAPE of $\leq 2000 \text{ J kg}^{-1}$ at the Northeast sites versus only ~19% at the Southeast sites.”

6. p3588, 16: “Spatial (domain) and temporal averages”

Modified as corrected by the referee.

7. p3591, 17-18: Last sentence is not a complete sentence. Delete the word “since” and it would be OK.

Modified as suggested by the referee.