

This manuscript presents experimental measurements of GEM, RGM, Hg_p and key meteorological parameters, analyzes correlations among these variables, and presents modeling of RGM diurnal profiles. The topic is clearly appropriate for this journal.

The authors clearly and carefully describe experimental measurements of mercury in the tropical MBL and correlations (or lack thereof) between mercury and Chl a and temperature, and correlations among concentrations of various forms of mercury. By contrast, the modeling effort is not described or analyzed as well. Critically, the apparent change in RGM lifetime from ~ 3 hours to > 8 hours between panels (a) and (b) of Figure 6 is most likely explained by an error in the modeling. This error undermines much of the results and analysis of the modeling. Therefore, it is my judgment that this manuscript be revised and resubmitted for review. Once this error is fixed and the modeling results more carefully analyzed, I expect the manuscript to be acceptable for publication.

1) Lifetime of RGM loss in Figure 6a and 6b.

The measured lifetime of RGM and the modeled lifetime of RGM in panel (a) is roughly ~ 3 hours. This is consistent with other results, such as Holmes et al (2009) cited in the manuscript. Loss of RGM in the model is due to deposition, uptake on sea salt aerosol, and boundary layer ventilation. The fact that the RGM lifetime in panel (b) is more like 12 hours for **all** chemical mechanisms suggest that the loss mechanisms were not properly included in the modeling. Alternative explanations are possible, but in any case, the origin of this difference should be clearly explained.

In any case, the revised manuscript should report the relative importance of different RGM loss mechanisms in the model.

2) The manuscript states that “Entrainment of $Hg(II)$ from the free troposphere is calculated according to Holmes et al. (2009).” However, the method of Holmes et al (as described in the second paragraph of their model description) parameterizes free-tropospheric [RGM] to allow models to match the daily average [RGM] in the MBL. In the present study, daily average [RGM] clearly varies with different chemical models, so the method of the present manuscript appears to be different than described. Clarification of the method of computing entrainment is clearly needed.

In addition, the used of a parameterized entrainment (which accounted for 25-40% of the RGM source in Holmes et al) can hide significant errors in the modeling. I suggest the authors specifically report the level of entrainment in their model, so that readers can evaluate the robustness of the model results.

3) In reporting the model parameters, the authors do not provide sufficient information for others to reproduce their modeling results. They authors need to report diurnal concentration profiles of key species (input to or output of the model), along with time-dependent photolysis rate constants, and parameters relevant to loss of RGM. This can be placed in Supplementary Material in order to avoid page charges.

4) In section 3.6 the authors report, without providing supporting data, that there is no correlation between $[NO_2]$ and 24 hr average [RGM]. This is stated more forcefully in the

abstract, and the conclusion about the lack of correlation is extended to [HO₂] and [RGM]. Note that, with the rate constants for HgBr + NO₂ and HgBr + HO₂ being similar, a correlation analysis should consider a weighted average of [NO₂] and [HO₂].

More importantly, a correlation analysis is not necessarily sufficient. If the fate of HgBr were to be 99% reaction with NO₂ and 1% dissociation, then a 90% reduction in [NO₂] would lead to the fate of HgBr being 90% reaction with [NO₂]. The corresponding 10% decrease in the fraction of HgBr oxidation to Hg(II) would be lost in the noise of the data. In the present study, I suspect that [NO₂] and [HO₂] are so low that most HgBr dissociates, in which case the correlation analysis would provide insight. The validity of a correlation analysis should be specifically addressed. To this end, it would be helpful for the authors to report on the fraction of HgBr that dissociates instead of forming Hg(II) in their model of this site.

4) Page 21549. It is great that the authors estimated rate constants for HgBr with several radicals (copying the method of Goodsite, et al, 2004), and this probably deserves to be highlighted more than has been done here. The manuscript should, however, should point out that these rate constants are rather approximate. For example, mercury models almost all use the rate constant of Goodsite et al. (2004) for HgBr + Br → HgBr₂, but a more rigorous analysis by Balabanov, Shepler, and Peterson (*J. Phys. Chem. A* **2005**, *109*, 8765-8773) gives a rate constant about 6-8 times lower.

Note that the same paper by Balabanov, Shepler, and Peterson reports a gas phase rate constant for Hg + Br₂ → HgBr₂ of about 10⁻³¹ cm³ molecule⁻¹ sec⁻¹. This low rate constant is entirely reasonable for the reaction of a closed shell molecule with an atom possessing a closed subshell (electron configuration [Xe]5d¹⁰6s²). Therefore, this gas phase reaction should not be included in modeling mercury.

5) Table 1 has a reaction forming HgCl (a Hg(I) compound), but no reactions of the HgCl radical! Also, the use of (+Y) in the reactions of Hg + Cl is not explained.

6) The heading to section 3.3 indicates Hg(0) oxidation by iodine, but the section actually describes oxidation of Hg(I) (in the form of HgBr) to Hg(II).

7) Page 21566 line 25. The “correlation” between RH and RGM (or Hg_p) is actually an “anti-correlation”