

## *Interactive comment on* "New evidence for atmospheric mercury transformations in the marine boundary layer" by Ben Yu et al.

## Anonymous Referee #1

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In this study, the authors present a comprehensive study on the isotope compositions of atmospheric total gaseous mercury (TGM) and particulate bound mercury (PBM) in the marine boundary layer over Chinese seas. As I know, this is a first study for investigating the isotope compositions of atmospheric Hg species and their underling mechanisms in MBL. It is therefore novel and would contribute significantly the Hg isotope research field. The manuscript is overall well written and organized. I agree with the interpretations on the mechanisms associated with the variations of Hg isotope compositions. Actually, I have revised this manuscript when it was submitted to another journal. I found the authors have address most of my and other reviewer's comments properly, and the paper is currently in a good quality. I would therefore suggest a publication of the manuscript in ACP with minor revisions. Some of the minor comments

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are listed below: 1. line 15: should be 'isotopic compositions of total gaseous mercury (TGM) and particulate bound mercury (PBM) collected....' 2. line 18: should be 'isotopic compositions of TGM in MBL were affected. ..' Line 19-20: the statement of 'lower air temperature could promote the positive..." is not clear. I would suggest to specify to 'TGM D199Hg values were significantly positively correlated with air temperature...', and then the author may interpret the potential mechanisms. Section 2.1: I would suggest to add a figure to show the cruise and sampling locations of the isotope samples in the main manuscript. Line 150-154: the diagnostics of using D199Hg/d202Hg ratios is great. But I think it should simply introduce the ratios obtained from previous laboratory studies or source materials, and this would help you to figure out the major factors. Line 168: 'TGM collected in two summer cruises ... ranges' should be " TGM collected in two summer cruises were characterized by significantly negative d202Hg and near-zero D199Hg values. Line 177-179: 'the back-trajectory....in TGM' should reword as 'Backward trajectory analysis showed that higher D199Hg values were associated with air masses originated from both mainland China and open oceans' Line 202-205: need to be rewritten. Note that the MIF signatures of gaseous Hg(II) emitted from anthropogenic sources and produced via atmospheric oxidation should be different. The former case would have near-zero value, while the latter case would have significantly positive value according to precipitation observations. These two should be differentiated here. Line 210-215: the D199Hg/D201Hg ratio for PBM should be presented Line 285-293: I agree with your interpretation that oceanic GEM emission should play a minor role. However, as the isotopic composition of oceanic GEM emissions have not been well constrained, I would suggest to soft these statement. Maybe this process could be completely excluded. Zheng et al., investigated the MDF of Hg isotope during aqueous Hg(0) evaporation. Note that GEM emission from water is driven by evaporation of Hg(0) from water (the isotopic compositions are unknown) and water-atmosphere interface photoreduction (the isotopic compositions of initial water are unknown). So it is difficult to known whether this source generate positive or negative MIF.

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