Responses to comments of Reviewer 1

We wish to express our great appreciation to Reviewer 1 for the careful review, supportive comments, and author-like editing. Your very careful comments and editing have significantly improved the quality of our draft. In the revised manuscript, we have incorporated all your comments. In the response below, we address each of major comments and all specific comments. The Reviewer's comments are italicized and our responses immediately follow.

This is an interesting study showing meaningful results. I would recommend publication in the Atmospheric Chemistry and Physics with consideration of the comments below.

<Major comments>

1. This paper examines both the individual and the combined effects of climate and mercury emissions changes. However, the analyses in this study are not solid enough to show 'climate effects'. 'Climate' was used in a very unspecific and broad meaning here. This study considered only a) changes in natural mercury emissions due to increased temperature and b) changes in anthropogenic mercury emissions in the future. Please be more specific whenever mentioning climate effects in the manuscript. I would suggest defining 'climate effects' in the introduction.

Revised: We revised the text to clearly define the climate effects in this study. We count the total effects including: (1) caused changes in natural emissions without considering the change in land/ocean Hg storage. (2) caused change in gaseous chemical transformation (mainly temperature dependent reactions). (3) caused change in aqueous chemical transformation (through water vapor, clouds, radiation effects), together as the climate effect.

2. One of the reasons to use the dynamic scheme is to consider seasonal variations of natural mercury emissions (Page 20168, Line 17-23). However, all figures in this paper show annual averages only. There should be at least a paragraph about seasonal variability of mercury concentration.

Revised: This is a very good suggestion. We added one figure after global TGM concentrations to show the global Hg seasonal variability.

3. Accelerated oxidization of mercury related to increased future temperature is mentioned several times in the manuscript. Authors should provide a plot similar to Fig. 5 to demonstrate it.

Revised: We totally agree with it. A plot is necessary. We added a figure by using curves of Hg(II)/Hg(0) under each scenario to demonstrate it.

4. Section 5 and 6 focus on the air quality in the US. Is this the first study on this topic? Otherwise, results from previous studies on the same topic must be compared with CAM-Chem/Hg results. Also as a reader, I would like to see some background of this study in the introduction regarding mercury pollution in the US.

Revised: we added some background studies on US mercury pollution in both introduction and section 5 and 6.

5. When I read Section 2, it seemed like all simulations had been carried out using meteorological fields from CCSM3 for the present climate. Did all simulations use the same meteorological fields representing present atmosphere? The authors also explain their

simulations at the beginning of Section 6. This first paragraph in Section 6 needs to be moved to Section 2. Please rewrite Section 2. The authors need to add a table summarizing all simulations used in this study.
Revised: They are using different meteorology fields, which are all from CCSM3 based on different scenarios and projections for future. We have revised the section 2 and 6, and added a table to summarize all simulations. That will be very helpful.

<Specific comments>:
For the benefit of readers, a number of points need clarifying and certain statements require further justification. Some examples are given below. Please rewrite some sentences. My suggestions are double quoted.
Revised: We should say much more than "Thanks" for your editing and considerations on following details of this manuscript. All your editing are accurate and very helpful to us.

Page 20166 Line 17: This pattern => "This spatial difference" Line 25: The sensitivity analyses presented show => "The sensitivity analyses show"

Page 20167 Line 12: What about "Changes in climate and mercury emissions determine atmospheric concentrations of mercury compounds in the future"? Line 14: "both the concentration and composition of atmospheric mercury in the future." Line 14: This sentence is not explained well with too much assumed knowledge. Is there any reference? Line 20: Please remove 'By contrast'. Line 23: over past centuries => "over the past centuries" Line 24: Did Selin et al., 2008 and Holmes et al.2010 only mention the changes in composition? Not concentration? Please revise the first sentence of this paragraph. Line 27: Please explain 'the source-receptor relationships'

Page 20168 Line 1: The uncertainties will not impact pollution. They impact accuracy of mercury pollution in the future simulated by models. Also large uncertainties => "considerable uncertainties" because you cannot directly compare uncertainty of climate and uncertainty of mercury emissions. Line 10: To analyze uncertainties, you need model evaluation against reliable observations. Line 11: Please rewrite this sentence. I would suggest "future atmospheric levels of mercury compounds are influenced by potential changes in emissions as well as changes in climate". Line 13-15: "Global anthropogenic emissions of mercury which is associated with social and industrial developments were estimated to be 2190Mg in 2000." Line 20: Please explain 'the simple scaling method' with references. This is important to highlight improvement of this study from previous ones. Doesn't the simple scaling method consider any seasonal variability and spatial inhomogeneity of future Hg emissions? Line 22: Is the dynamic modeling method important only for natural emissions of mercury or any other chemical species? Line 1-23: Please rewrite these two paragraphs. Some sentences are redundant. I would suggest Mercury is emitted into the atmosphere from both anthropogenic and natural sources (1st paragraph) - natural emissions are affected by climate change (which variables are important for mercury emissions from natural sources? temperature? humidity?) (2nd paragraph) anthropogenic emissions Line 24: A reference to CAM-chem is required (Lamarque et al., 2005) Line 27: What this study addresses is future effect of climate and emissions on atmospheric mercury, not uncertainties. To address uncertainties, we need ensemble simulations from multiple models. Please clarify this. Line 29: climate warming => "climate change"

Page 20169 Line 1: Projections => The projections Line 9: "The model used in this study, CAM-Chem/Hg, is a three .." Line 10: This sentence ('Details of the mercury model ...') should be placed in Line 17. Line 11: "The CAM-Chem model considers fully coupled gas-aerosol phase chemistry that originates from the MOZART." Line 19: Is there a reference to air-sea mercury exchange scheme? Line 21: *"The model's mercury chemistry includes oxidation of elementary mercury and gaseous mercury. Elementary mercury is oxidized by ozone and the oxidation is temperature dependent (more oxidation with higher temperature?). OH, H2O2 and chlorine oxidize Mercury in gas phase." Line 25: "After balancing all chemical reactions, transportation and deposition of mercury are calculated in each time step."*

Page 20170 Line 1 & 11: CAM-Chem => "CAM-chem/Hg" Line 9: "Biomass burning of mercury"

Page 20171 Line 24: the projected => " the present and projected"
Page 20173 Line 4: Is this about 'natural' emissions from soil and ocean? I would suggest "3.3 Natural emissions from land and ocean". Line 5: 'modify' means that the authors changed the original dynamic emission scheme. Please explain in more detail. Line 6: What is F2? Line 10: What is sensitivity? Line 16: What is F? Is it different from F2?
Revised: We change the original emission scheme by bring in variables that represent the mercury storage change in land and ocean reservoirs. F2 is the projected future natural emission flux from land. F is the emission flux from ocean surface.

Page 20174 Line 21: mi is a typo.
Page 20175 Line 14: 'trend' is a term for temporal change averaged over a certain period. Line 16: Please remove 'the result shows that' Line 21: Please quantify statistical significance of the difference. Is it really statistically significant across the globe? It seems like most of TGM increases occur over land.
Revised: We added the percentage of increase and decrease. Also stating that TGM increase significant over land.

Page 20176: Line 1: What is 'mercury emission industrial regions'? Line 5: The authors should mention that interhemispheric difference in the future is much larger compared to present. Line 19: This sentence is redundant. There should be a sentence explaining about mercury and air quality in the US. Why does this study focus on US air quality? Is it for comparison with previous studies? Explanation added on the beginning of paragraph. *Line 22: Please revise this sentence. Line 26: Is there any supporting results? How can we separate Hg increase due to transportation from neighboring countries?*
Revised: Supporting reference added.

Page 20177: Line 5: Why do the authors show changes in wet deposition? There should be a sentence like this to summarize Section 5: "Wet deposition of mercury increases in the future but the increases are not enough to set off emission increases. Therefore.... " Figure 5: Please redraw this. The contour levels are not readable at all. Line 6: "the peak wet deposition is located in the southeast" Line 11: Please remove 'only' Line 18 – 25: This paragraph should

move to Section 2. Line 20: which incorporate => "considering", but keep => "while keeping" Line 22: for the three scenarios => " for the three future scenarios (B1, A1B and A1F1)."

Page 20178: Line 4: cylinder represents => "bars represent", the average concentration over the US => "the mercury concentration averaged over the US" Line 5: Do minimum and maximum mercury concentrations in the US? Please revise 'projected concentration range'. Line 7: the increases are continuous, not just in 2050. Line 8: climate warning => "warming" Line 12: Please show evidences to support the accelerated oxidation under A1F1. This may be more important than Figure 5. Line 14: Does this mean that natural emissions are still different according to the equations introduced in the previous section?
Revised: Yes. We follow the major comments and clarify the climate change impact.

Table 2: It is hard to separate scenarios. Please add horizontal lines between scenarios. Line 16: So what do the small differences mean? Line 19: The authors should show temperature changes over the US, not the global mean temperature change. Line 20: Please rewrite this sentence. Does this refer to the far right column of Table 2? Line 21: It should be mentioned that the contribution of climate change is made through natural mercury emissions. Line 27: Again, there is no supporting plots to show the accelerated mercury oxidation. Line 28: relatively higher => "relatively high"
Page 20180 Line 1: under the B1 scenario Line 3: What about the impact of rising temperatures? Line 16: imbalanced change => "difference" Line 16-18: Please do not guess. This study did not show anything related to mercury transport.
Page 20181 Line 2: see => "show" Line 4: This sentence is redundant.