The manuscript by Shi et al. measured GEM concentrations in January and July in five individual years from 2012 to 2020, and the data were used to explore the potential factors controlling the inter-annual variations. Long-term measurements of GEM concentration are a useful tool for assessing of controls of regional anthropogenic emissions and global changes, and thus the data presented here are valuable. In the present study, the authors combined the multiple approaches including the analysis of GEM concentrations, criteria pollutants, backward trajectory and generalized additive model. I agree that it is practicable and relevant for using these kinds of methods to explore the controls in the change of atmospheric GEM. The manuscript is overall well organized, and can be read easily. I broadly agree with the discussions and findings of this manuscript. I therefore suggest a minor to moderate revision of this manuscript before final publication in ACP.

Response: Thanks very much for your overall positive review and valuable comments, which are very helpful for improving the quality of our manuscript.

As mentioned at the beginning of the manuscript, the major objectives of long-term observations are to evaluate the changes in anthropogenic emissions, that is an important part for the implementation of the Minamata Convention on Hg. However, after a comprehensive analysis, the authors mostly highlight that the changes in meteorological conditions were the most important variable in controlling the long-term trend in GEM. This is valuable, but not very striking findings to me because it is well accepted that variations in GEM among different short periods (e.g., monthly) could be impacted by changing atmospheric transport (air transport would change with different periods and subsequently affect the source-receptor relationships). Thus, I would suggest the authors to focus on the impact of changing local and regional anthropogenic emissions and climate on the trends in GEM concentrations, which would better serve for their research objectives.

Response: According to your suggestions, we summarized the published data of China's Hg emissions inventory so far **(Figure S3, S4)**, and placed greater emphasis on the impact of changing local and regional anthropogenic emissions and climate on the trends in GEM concentrations in the whole text as well as in the summary and conclusions **(e.g. Line 221-231, Line 357-363)**.

I am not clear why the meteorology is the major divers of changing GEM concentrations, and it also difficult to differentiate the impacts of meteorology, transmission, and emissions. I suspect that the transmission should be related to meteorology because the changes in local and regional meteorological conditions would further affect the transmission. Would the meteorology change land surface emissions and or atmospheric reactions that further affect the GEM? In addition, several previous studies reported declines in GEM concentrations in eastern China. Would this be an important cause for the changing contributions from transmission and meteorology? Thus, the authors may better define the three factors, which would help to better understand the real causes for the changes in GEM concentrations.

Response: Thanks very much for raising this critical issue. Yes, regional meteorological conditions and anthropogenic emission amount would affect transportation, and meteorological conditions also impacted land surface emissions and atmospheric reactions. Actually, it is impossible to differentiate the impacts of meteorology, transmission, and emissions in a realistic

scenario. Statistical methods simplify this realistic scenario and use representative parameters to indicate the factors.

The selection of representative variables for the factors was very crucial and decided what the factors mean. 16 variables we obtained from site observation and web downloads were screened by collinearity test, considering the meaning of variables, and statistical judgement. As a result, CO was used to represent the anthropogenic emissions factor; RH, T and BLH interaction was used to represent the meteorological factor; and 24h-Longtitude, 24h-Latitude interaction was used to represent the transportation factor. Given that the 6 selected variables passed the collinearity test, the three factors were considered to be independent of each other. We have added a detailed explanation of the factor's selection and the meanings of the factors in the revised manuscript. The detailed responses to this issue could be seen below.

Line 144-145: why did the authors only conduct two-month observations at the sampling site? Why not conduct a year of continuous observations for the selected years? A two-month observations in one year are sometime not adequate for assessing the inter-annual variations because of many factors mentioned in the manuscript.

Response: The main reason was that the period of instrument malfunction was different among years. For example, data for October, November and December were missing in 2013 and data for August, September and December were missing in 2015. We used representative months of data so that the period of GEM data was consistent and the GEM data could be comparable among years. We used January and July data mainly based on two considerations: (1) The measurement site, Xiamen, is located in the coastal region of Southeast China under the control of East Asian monsoon, which has a significant distinction in meteorology between summer and winter; (2) Based on our previous study on GEM observations in Xiamen throughout a year (Xu et al., 2015), January is very representative of winter and July is representative of summer. We added a description of seasonal representation to **Section 2.2 Observation period selection (Line 106-114)**. Furthermore, considering the influence of gap years, we have used "variation in winter/summer among years" instead of "inter-annual trend" in the revised manuscript.

Line 166-167: the definition of local impact relating to air mass within a province might over-estimate the local effect. Why not define the local impact within the city?

Response: In this study, polar plot was used to analyze the influence of local sources within Xiamen City. As for the regional source identification, we found that the endpoint of 72h backward air mass trajectories was relatively rare within Xiamen city. "Local" in air mass trajectory analysis means the emission source surrounding the site (mostly within Fujian province) compared to regional transportation. We have modified the backward trajectory analysis to a CWT analysis and changed "Local" in regional sources regions to "Fujian province" in the revised manuscript (**Figure 5 and Section 3.2.2**), which can display the source region more clearly.

Line 198-204: CO is mainly sourced from anthropogenic emissions but has a long atmospheric residence time, it may therefore a best proxy of local anthropogenic emissions. I would suggest the authors to consider using SO_2, NO_2, or PM_{10} to define the anthropogenic factor, although these parameters would have relative weaker correlations with GEM. Why use RH and SP to

define meteorology? How could these two factors affect GEM concentrations? What are the 24h-latitude and -longitude? Are they referred as the air massed originated outside the city to define long-range transport?

Response: Thanks very much for the valuable comment. We added a detailed description of the variables screening into **Section 2.5 Parameter Selection** and some factor explanations into the corresponding discussion in **Section 3.3.2**.

(1) CO is mainly sourced from anthropogenic emissions and has a long atmospheric residence time (compared to SO_2 and NO_2). In addition, Hg emissions in Fujian provinces were dominated by combustion sources (Liu et al., 2019). Hence, we used CO to represent anthropogenic Hg emissions.

(2) According to another reviewer's comment, we used the min-maximum method to standardize the parameters before the model running. SP was rejected by collinearity test. We filtered remaining meteorological parameters based on the performance of the model fit as well as the variables importance. In this step, WS, WD and LCC were rejected and T, RH, and BLH were retained. The meteorological factor was represented by T, RH, and BLH interaction. The meteorological factor influenced GEM variations by several pathways. To better illustrate how the parameters affect GEM concentrations, we plotted the corresponding curves of GEM with variables (**Fig. 8**). GEM concentrations increased with the raise of temperature, possibly because the increase of temperature would promote the Hg emissions from natural surface. GEM concentrations increased with the raise of RH, possibly because the high RH environment was favorable for the liquid phase reduction of reactive Hg to GEM (Horowitz et al., 2017; Saiz-Lopez et al., 2018; Huang et al., 2019). BLH primarily indicated the physical status of the atmosphere especially diffusion conditions. We also gave a case to the combined effect of T, RH, and BLH on GEM variations (**Fig. S8**).

(3) The interaction of 24h-Latitude and 24h-Longitude was defined as the regional air mass transport, which represented the backward trajectory endpoint location during last 24 h.

Line223: the range of background GEM concentrations of 1.5-1.7 ng m^{-3} is somewhat higher to me. Better to use recent global observations.

Response: The most recent data we found was the GEM concentrations at ground-based Global Mercury Observation System (GMOS) sites in 2013 and 2014. In the northern hemisphere, GEM measurements for background sites were ~1.5 ng m^{-3} (Sprovieri et al., 2016; Diéguez et al., 2019). We have updated the data in the manuscript **(Line 207)**.

Figure 2: a statistic of the annual GEM concentrations should be added

Response: It might be not very suitable to do a statistic of the annual GEM concentrations in this study, because we used only two representative months of GEM data. To be more accuracy, we have compared the Jan. and Jul. data with the corresponding winter and summer data from the references rather than annual average data (**Line 210-216**).

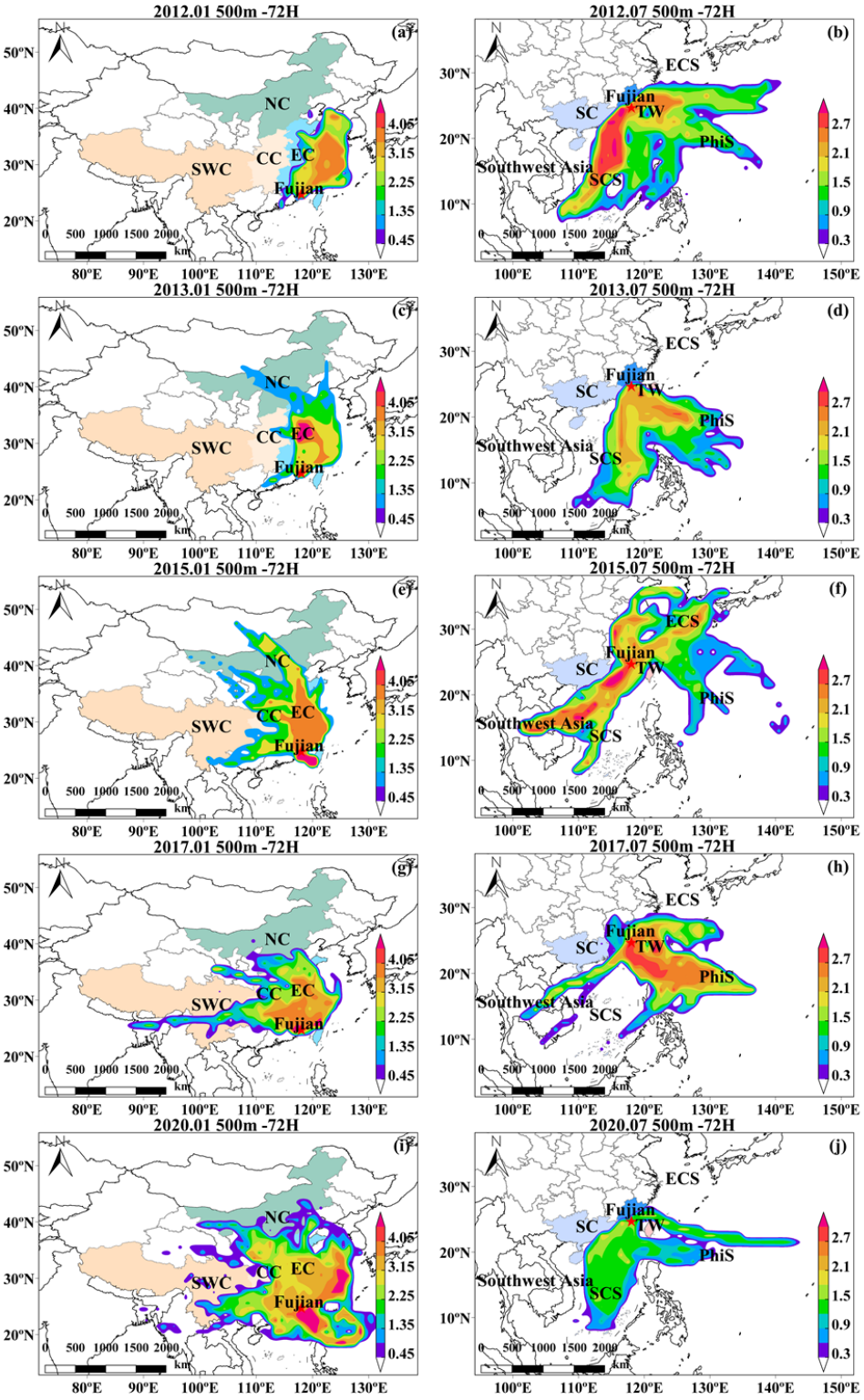
Line 274: the GEM lifetime here is not consistent with that in line 76

Response: We finally unified the GEM lifetime to 0.5~2 years (Schroeder and Munthe, 1998).

Line 297-298: elevated O_3 and decreasing GEM concentrations should be mainly related to subsidence of free troposphere and daytime production of O_3. If the daytime declines in GEM is caused by oxidation, we would expect a much higher oxidation rate than experimental studies.
Response: We agree with you. We have clarified that the oxidation of GEM might contribute a small part to the daytime reduction of GEM, but was not the driving factor due to its low oxidation rate. We have highlighted the impact of subsidence of free troposphere in the revised manuscript **(Line 289-291)**.

Line 317: a citation of references should be added here
Response: We added the reference (Air pollution emissions inventory of Xiamen city in 2016, unpublished report) here (**Line 309-311**).

As shown in Figure 5: a large fraction of air masses originated or passed over oceans, please add their weighted GEM concentrations in Table 2
Response: Thanks for your suggestion. In the revised manuscript, we have further performed CWT analysis based on backward air mass results, which brings a better insight into the mercury source locations. The results of CWT analysis are showed in below figures. The corresponding discussion was also revised.



Line 424-426: it is difficult to expect low regional anthropogenic emissions because the GEM measured are much higher the background levels in East Asia. I suspect that the other two factors of transmission and meteorology were also impacted by changing local and regional anthropogenic emissions. Actually, I do not know what are these three factors representing. Are the anthropogenic emission and transmissions representing local anthropogenic contributions and regional background? What is the meteorology? Is it representing natural emissions'?

Response: We agree with that the regional high GEM concentrations affected the GEM in the study region, and the other two factors of transmission and meteorology were also impacted by changing local and regional anthropogenic emissions. Actually, it is impossible to differentiate the impacts of meteorology, transmission, and emissions in a realistic scenario. As above responses mentioned, statistical methods simplified this realistic scenario and used representative parameters to indicate the factors. Thus, the representative variables for the factors mostly decided what the three factors represent. 6 variables including CO, RH, T, BLH, 24h-Latitude and 24h-Longitude were screened out to represent the factors. We have added a detailed explanation of the factor's selection (**Section 2.5**) and the meanings of the factors in the corresponding discussion (**Section 3.3.2**)

(1) The anthropogenic emissions factor was represented by CO. CO and GEM have a long lifetime in the atmosphere, and anthropogenic Hg emissions in Fujian were dominated by combustion sources (Liu et al., 2019), so we used CO to represent anthropogenic Hg emissions.

(2) The transportation factor was represented by 24h-Longtitude, 24h-Latitude interaction. 24-latitude and 24-longtitude referred to the direction and distance of the 24h backward air mass from the study site. We did not put the regional background concentration as a parameter into the model. But this factor (the interaction of 24-latitude and 24-longtitude) would cover the effect of regional background of GEM concentrations.

(3) The meteorological factor was represented by RH, T and BLH interaction. The meteorological factors affected GEM concentrations by several pathways: (a) Hg chemical transformation. For example, RH was highly related to the liquid phase reduction of reactive Hg in the atmosphere (Horowitz et al., 2017; Saiz-Lopez et al., 2018; Huang et al., 2019). (b) Hg emissions from the natural surfaces. Substantial flux measurements have clearly showed that the elevated temperature and solar radiation can significantly promote the Hg re-emissions from these nature surfaces. (c) physical state of the atmosphere. A higher boundary layer likely indicated good diffusion conditions and was more conducive to the reduction of pollutant concentrations.

In order to better illustrate the meteorological influence, we added a typical case for additional clarification (**Figure S8, Section 3.3.2**).

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