Reviewer's Comments

General comment

This study has analyzed data from multiyear measurements of the gaseous elemental mercury (GEM) concentration at a regional background site in eastern China and quantified the contribution of natural surface emission to GEM using the positive matrix factorization (PMF) model. The long-term observation data are valuable and the topic is of broad interests.

My major concern is the robustness of the PMF results. To what extent should we believe these results? The results need further verification. Figure 5 is one of the most important yields from this study. Suppose Figure 5 is basically correct, we can draw some important conclusions from this figure:

(1) Although cement production is believed to be one of the most important emission sources in China, it seems to contribute very little to GEM at this site. Could this be true?

(2) The current Hg emission inventories haven't considered ship emissions, but this emission source should be considered in the Hg emission inventory development, especially for coastal areas. This could be a very important finding if it is true.

(3) Iron and steel production has a large contribution to GEM concentration as well. Is this site under the influence of many large iron and steel plants (e.g., Baogang)?

If the contributions from different anthropogenic sources could be verified to some extent, it would be much easier for the readers to believe the contribution from natural sources. One possible approach for the verification that I can think of is to use the PSCF model to identify the potential GEM source regions from 2015 to 2018. If the key source regions for the monitoring site are consistent with the above conclusions (e.g., do not have many cement plants; have potential ship emissions from the seas or the rivers; have many iron and steel production activities; etc.), the robustness of the PMF model could be verified.

Overall, I think this manuscript is worth publishing on Atmospheric Chemistry and Physics after major revision.

Specific comments

1. Lines 47–48: It should be “non-ferrous metal smelters” instead of “non-ferrous smelters”.

2. Section 2.2: How many valid GEM data were included in the analysis?

3. Lines 195–196: This statement is not accurate and lacks evidence. Some of the anthropogenic emission sources vary significantly from season to season. For example, coal combustion for residential use has a much higher level in winter.

4. Lines 208–209: The p values for all the correlations should be given here. Have the authors investigated the correlations between GEM and solar radiation? Solar radiation and temperature could have collinearity to a certain extent. It is possible that the diurnal GEM trend has a more significant correlation with solar radiation. Solar radiation is related to the photoreduction process of Hg in soil, which could be the major natural GEM source in the study area.

5. Lines 241–245: The choices of NH_3 and O_3 as tracers should be more carefully examined. These two tracers are not directly linked to natural emission sources, but indirectly through temperature. If temperature is already chosen as a tracer for PMF and NH_3 and O_3 are only linked to natural sources through temperature, what is the point of choosing NH_3 and O_3? The authors should pay attention to the other links between NH_3/O_3 and natural sources. Say the links through solar radiation, land surface type, and so on. Moreover, the PMF method usually chooses primary air pollutants as tracers, e.g., VOC species profiles, ions on particles, heavy metal profiles, etc. Secondary air pollutants, such as O_3, are usually avoided to be used as a tracer for PMF, because all the coefficients resulting from the PMF model need to be positive while it is not always the case for secondary air pollutants like O_3, not to mention that O_3 and GEM are potentially not independent variables. O_3 might act as an oxidizer for GEM under certain conditions (e.g., high humidity),

although this mechanism is not clear so far. Therefore, the authors should either remove O_3 as a tracer or explain why in this case O_3 is applicable from PMF.