The manuscript addresses the variability of atmospheric mercury concentration in a coastal city in Southeast China. The manuscript aims to report the main factors driving GEM variability by deploying the regression analysis method. The scientific question is relevant to the scientific community. However, many issues can be highlighted in the manuscript.

The main concern in the manuscript is its design, how the Generalized Additive Model was used, and the premises assumed for the pattern recognition of the factors driving GEM variability. The authors lack knowledge of the used method. The signal extracted from the matrix of trace gases, PM, and meteorological data used to reconstruct GEM, is not explicitly linked to GEM sources, transport, or processes. The factorization was constrained by a minimum concentration covariance that led to the meteorologic factor as the main cluster. I am afraid that the authors were misled by a spurious correlation in the propagation of the eigenvector, where the main factor explaining the GEM was seasonality. The main disadvantage of the unsupervised learning technique as the one used by the authors is the fact that the possible solution is no-unique.

Response: Thanks very much for your careful review and valuable comments which are very helpful for improving the quality of the manuscript. We have learned more GAM method through materials and literature. GAMs seem not like traditional unsupervised learning techniques such as PCA. We did not use GAMs to cluster, but to build a well-fitted nonlinear regression model and calculate the variation in factors interpretations rate. We carefully read the comments and revised the whole manuscript accordingly.

1. In this study, we assumed that the GEM concentrations were mainly affected by three factors: anthropogenic emissions, meteorology and transportation. 16 variables we obtained from site observation and web downloads were screened to represent the three factors using two methods: Statistical judgment and the meaning of the variables. The detailed screening processes have been implemented in the revised manuscript.

2. To eliminate the effect of seasonality on variance clustering, we have used “seasons” as an input variable when building the model with the whole dataset, and then run the model separately in summer and winter. We clarified this point in the **Section 2.5 Model establishment (Line 187-189)**. At the same time, we focused on inter-annual differences in individual seasons instead of seasonal comparisons when discussing the results.

3. We agree that it’s better to use the variables which are explicitly linked to GEM sources, transport, or processes. However, we encountered some difficulties to obtain the high spatial resolution of Hg emissions inventory in China. In addition, there is no known explicit parameters to represent GEM processes in the atmosphere. The advantage of GAMs is that it can use routine monitoring or easily obtained parameters to represent the influencing factors. In revised manuscript, we explained the meaning of the retained variables in detail.

4. Yes, the possible solution of GAMs is no-unique. In this study, the accuracy of GAMs simulation was assessed using a 10-fold cross-validation test. The principle of the test is dividing the whole dataset into ten subsets randomly, and in each round of cross-validation, nine subsets are used to fit the model and the remaining one is predicted. This process is repeated 10 times to ensure that every subset is tested. The 10-fold cross-validation results showed a good coincidence between the GAMs and cross-validated result. In addition, we also use the gam.check function (e.g. Quantile-quantile (QQ) plots) to ensure the validity and

accuracy of the model. The detailed introduction could be found in **Section 2.5 Model Quality Control (Line 190-200).**

Specific comments:

Line 236: The authors call data from two months "trend over 2012"; however, it corresponds only to ten months of data for a period of nine years. The terminology "trend" is incorrect throughout the manuscript and should be revised. After all, it is not clear why the authors used only January and July data.

Response: Thanks for your suggestion. We used "variation" instead of "trend" in the revised manuscript. In addition, we added the explanation of the period of GEM observation data to the text **(Section 2.2, Observation period selection)**. The main reason was that the period of instrument malfunction was different among years. 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 chose 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.

Line 239-249: The emission data should be presented, and regression with observation should be discussed.

Response: It's a pity that we do not have the Hg emission inventories data. We summarized the published data of anthropogenic Hg emission in China so far, and added it to the supporting information **(Figure S3,4)**. The published data did not cover the study period 2012-2020, and the small amount of annual emission data might be not suitable for regression analysis. According to the published data, Wu et al. (2016) estimated atmospheric Hg emissions in China decreasing from 547 tons in 2010 to 530 tons in 2014. The report from AMAP/UNEP showed that the anthropogenic Hg emissions in China were 565.2 t in 2015 relative to 575.2 t in 2010. An inventory over the period 1978-2017 revealed that China's anthropogenic Hg emission was highest in 2013 and then decreased until 2017 (Liu et al., 2019). It could be expected that the anthropogenic Hg emissions in China had a downward trend over the period 2012-2020 and the peak emission was most likely to occur in 2012 to 2014. A more detailed description of Hg emissions was added in the **Section 3.1.1 (Line 225-231)**.

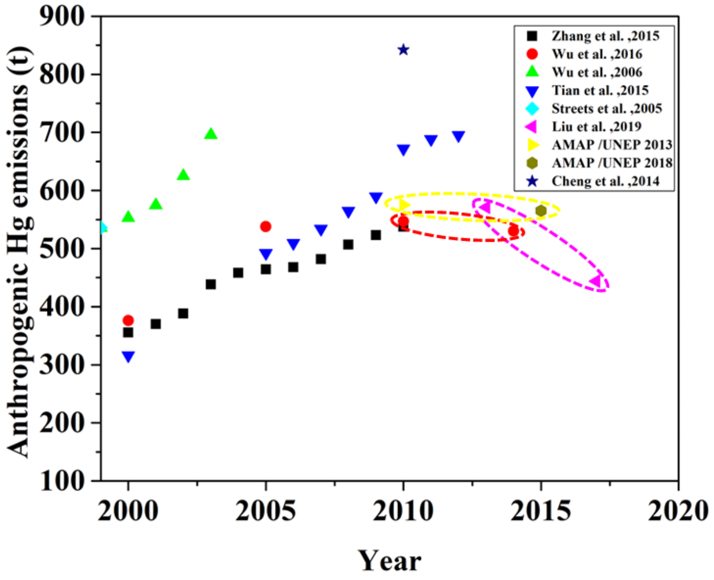


Figure S3. Anthropogenic mercury emissions from China reported in the literature (Streets et al., 2005; Wu et al., 2006; Cheng et al., 2015; Tian et al., 2015; Zhang et al., 2015; Wu et al., 2016; Liu et al., 2019).

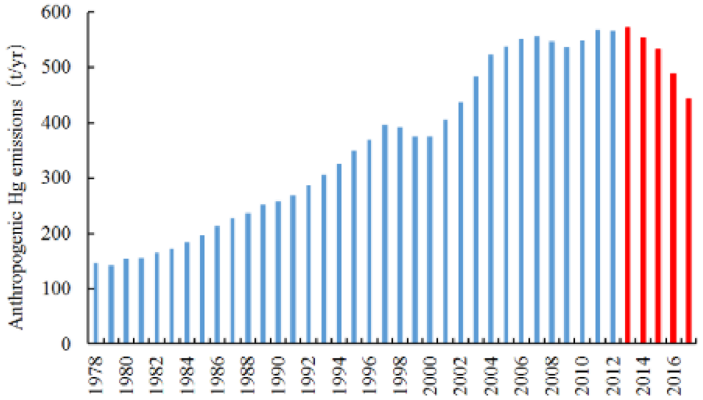


Figure S4. Anthropogenic mercury emissions during 1978-2017 in China (Liu et al., 2019).

Line 243: "aggressive" what does it mean?

Response: We mean "vigorous measures". In 2013, the Chinese State council issued an air pollution prevention and control action plan. Since then, plenty of emissions control measures, like accelerate the elimination of backward production capacity, accelerate the promotion of central heating, upgrades and building air pollution control devices, have been widely implemented in China **(Line 244-247)**. We changed "aggressive" to "vigorous measures" in the revise version.

Line 252: Would it be possible to show the coal consumption in Fujian and China?

Response: As you suggested, we provided coal consumption in Fujian and China during 2012-2020 both in **Fig. S5**. The data came from the Statistical Yearbook of China and Fujian (http://www.stats.gov.cn/tjsj/ndsj/: last access: 15 June 2022). The coal consumption in Fujian and China exhibited a similar variation, firstly decreasing to a valley in 2016 and then showing an upward trend from 2016 to 2020 **(Fig. S5)**.

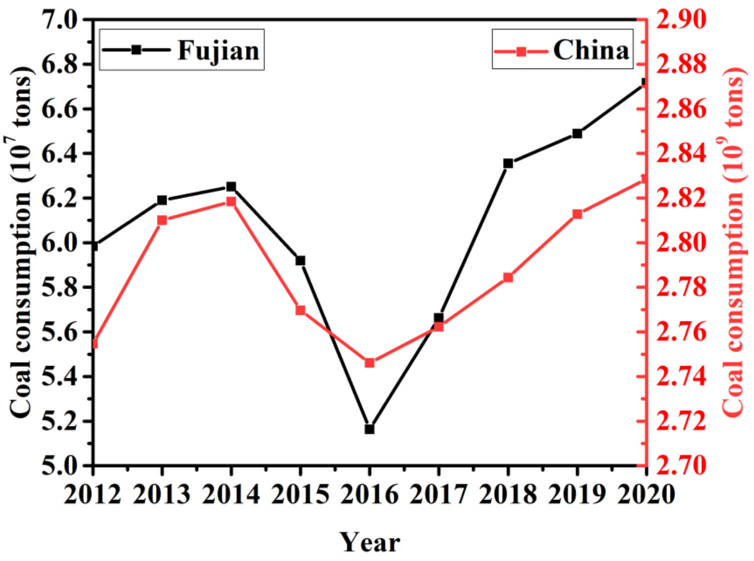


Figure S5 Statistics of annual coal consumption in China and Fujian Province during 2012 – 2020.

Line 259: Probably, the authors mean inter-annual variation rather than an inter-annual trend. I am afraid that the data exploitation presented by the authors does not allow a proper evaluation of the trend.

Response: We agree with you. We used "inter-annual variation" instead of "inter-annual trend" in the text and focused on inter-annual comparison of GEM concentration when revising the manuscript.

Section 3.1.2

I am afraid that using only two months is inappropriate for seasonality evaluation. In addition, one month represents only 1/3 of the season.

Perhaps it would be more appropriate to call the section January/July comparison rather than "seasonal".

Response: According to the climate features in the study region, January and July could well reflect the characteristic of the winter and summer seasons, respectively. The observation site Xiamen is located in the coastal region of Southeast China under the control of subtropical oceanic monsoon, which has a significant distinction in meteorology between summer and winter. In addition, a whole year GEM concentration observation in Xiamen supported that the GEM data in January can represent the GEM characteristics of winter and July can represent summer (Xu et al., 2015). We added the instruction of the season's representation of January and July in **Section 2.2 Observation period selection**.

Line 271-282: The polar plot does not support the statement of dominant wind from the North or a higher concentration of GEM on this wind. If the plots are correct, the predominant source of GEM in January is in the west, and long transport does not play a major role in the level of GEM at Xiamen. Actually, the plot shows only a low level of GEM at wind from the sea.

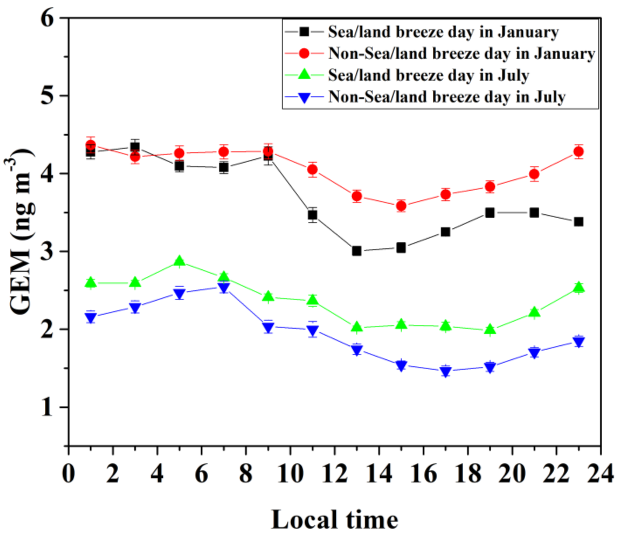
Response: We are sorry we did not explain the air mass and near-ground wind directions clearly. The wind speed/direction used in the polar plot analysis was observed near the ground and represents a very local scale (within Xiamen city) airflow condition. The wind speed/direction was strongly affected by the terrain. The "wind" here refers to the air masses which mainly affected by large-scale atmospheric circulation. The air masses in the study region was mostly originated from the land area in winter and from the ocean in summer. To be clearer, we've changed "wind" here to "air mass".

Line 283-288: It seems confusing; the authors should consider rewording it.

Response: We rewrote this passage as follows: "The diurnal variations of bihourly GEM concentrations were consistent among years (Fig. 3). In general, the GEM concentration peaked in the early morning, decreased to a valley in the afternoon, and then rose during the night. The diurnal pattern of GEM concentrations in January 2015 were gentle than other years of the same period, which might be related to the enhanced effect of air mass transport (Fu et al., 2012; Nguyen et al., 2022)". **(Line 277-281)**

Line 289: The diurnal pattern observed for July can be potentially constrained by sea/land breeze since it is a coastal place.

Response: We agree that the sea land breeze (SLB) is a potential factor of atmospheric mercury concentrations in coastal cities. Our statistics for 2017 – 2020 show that the average number of SLB days was only 6 days in January and 3 days in July. In addition, we compared the diurnal trend of atmospheric mercury in January and July of 2017 and 2020 on the SLB days and non-SLB days. As shown in the chart below, there was no significant difference in the diurnal pattern of GEM between SLB and non-SLB days. Thus, we infer that sea land breeze was not the dominant factor influencing the diurnal trend of mercury concentrations.



Line 297 – 298: For kinetic reasons, photo-oxidation cannot be the explanation for the observed reduction of GEM in the daytime. It is most like related to GEM fluxes. The authors speculate about the diurnal variation of GEM without a solid clue about the processes driving it.

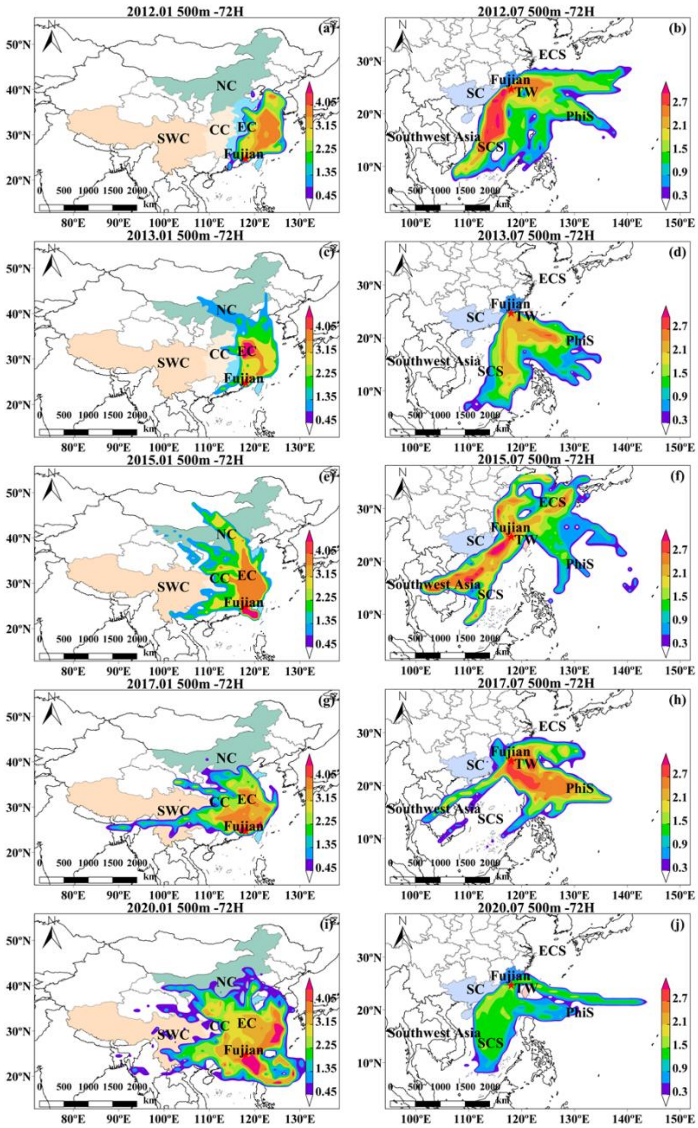
Response: We agree with your comment. The photo-oxidation of GEM possible contributed a small part to the diurnal variation of GEM, but it was not the dominant factor for the daytime

GEM reduction in the study region. We clarified this point in the revised manuscript **(Line 289-291).**

Polar plots are quite limited in providing emission locations. Concentration-Weighted Trajectory could improve this section; it would map GEM, allowing hotspot concentration identification.

Figure 5 does not bring insight into the mercury source location. A different kind of plot should be presented. In addition, a clearer CWT method should be presented.

Response: The polar plots analysis here was used for the identification of local point sources mainly within Xiamen city. According to your suggestion, we further performed CWT analysis and revised the relevant discussion in the main text **(Section 3.2.2).**



Line 365-370: It seems a last-minute explanation; since only fluxes can explain variation in the atmospheric mercury concentration, the authors should look into Hg emission to address a more convincing explanation.

Response: We rewrote this section and considered primarily the impact of Hg emissions, and secondarily the impact of meteorology **(Line 357-363)**.

“The high GEM concentrations in January 2015 was likely due to a combination of high-level Hg emissions and adverse meteorological conditions. The annual atmospheric mercury emissions in China were about 565 tons in 2015, which was roughly 20% higher than they were in 2010 (AMAP/UNEP, 2018). According to anthropogenic mercury emissions inventory in China during 1978 – 2017, mercury emissions might peak around 2013, and remained high in 2015 (Figure S2). In addition, an adverse effect of meteorological conditions due to extreme 2015 – 2016 El Niño event might result in an increase of GEM concentrations in 2015 (Nguyen et al., 2022).”

Section 3.3.2

This section has major concerns

Unsupervised learning techniques are power statistic methods applied successfully to extract signal and meaning information from high-dimensional data. Deploying nonnegative matrix factorization, we can more than explain covariance; we can extract the pattern of source and transport of atmospheric trace gases. However, I am afraid that the authors did not design the factorization properly. The species considered in the matrix were chosen without criterium. It was only convenient for the authors to have those species there. What is the sense of having PM in the matrix? Considering species that do not bring retrieval signals will not provide insight into mercury processes/source/fade. It only increases uncertainty and chances of spurious correlation, misleading the eigenvector's propagation.

Response: Thanks very much for your valuable comment. We added a detailed introduction of GEM factor selection into **Section 2.5 Parameter Selection** in the revised manuscript.

1, 16 variables we obtained from site observation and web downloads generally fell in four categories: anthropogenic emissions (SO_2, NO_2, O_3, CO, $PM_{2.5}$, PM_{10}), surface meteorology (T, RH, WS, WD, SP), high-altitude meteorology (BLH, UVB and LCC) and air transmission transportation (24h-Latitude and 24h-Longitude). All the variables were standardized by min-maximum method. The normalized data eliminates the effects of differences in dimension and ranges of values between indicators. The standardized variables were then screened using two methods: Statistical judgment and the meaning of the variables.

2, The detailed processes of factor screening are as follows. 1) We performed collinearity diagnostics with all the parameters. PM_{10}, SO_2, NO_2, SP and UVB were rejected into the model due to their high collinearity (VIF > 5). 2) we considered the meaning of the remaining parameters based on the literature and our research experience. 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. Parameters O_3 and $PM_{2.5}$ were easily rejected into the model. 3) After determining the first parameter CO, we put the remaining parameters (WS, WD, T, RH, BLH, LCC, 24h-Latitude and 24h-Longitude) into the model one by one. As the parameters were successively added into the model, the AIC decreased and R^2 increased. In this step, WS, WD and LCC were rejected. Eventually, 6 variables including CO, RH, T, BLH, 24h-Latitude and 24h-Longitude were selected into the model.

3, Considering that parameters of the same category may interact, we used interaction functions of tensors. RH, T and BLH interaction was used to represent the meteorological factor. 24h-Latitude and 24h-Longitude interaction was used to represent the transportation factor. Given that the 6 selected variables passed the collinearity test, the three factors they represented, i.e., anthropogenic Hg emissions, meteorological and transportation were considered to be independent of each other.

The major problem in this study was the correlations extracted from the species inserted in the factorization. The differences in the GEM concentration through the season, which are dependent on the seasonality of the emissions, were correlated with the seasonality of the meteorological parameters, which were extracted as the causes of GEM reduction in July. The direct incorporation of meteorological parameters into the factorization misled the eigenvector propagation. The seasonal differences created cluster minimizing the variance but do not sign origin/source/or fluxes of GEM.

The factor obtained by the authors does not provide any insight into the GEM reducibility (computationally speaking) since it does not bring information on the source/or fluxes of GEM. Seems that the authors did not plan the species to be considered in the calculation.

Moreover, the meteorological variables cannot be included directly in the factorization matrix. In order to evaluate transport, the authors should use an inversion accoupled with a transport model.

I hope the authors do not feel disappointed or frustrated with my comments. I`m very enthusiastic about unsupervised learning methods for pattern recognition and estimation of fluxes and the implementation of nonnegative matrix factorization into inversion modeling. Indeed, it has great potential to bring new insight into atmospheric mercury reducibility. I hope the authors only feel motivated to learn and improve their research.

Response: We really appreciate your comments and suggestions. We learned a lot from your comments. The inversion model is indeed a very meaningful work for regional and global flux estimation and has great potential to bring new insight into atmospheric mercury reducibility. Whereas, the main purpose of this study was to recognize the factors driving the inter-annual variations of GEM concentrations, which is a little different from building an inversion model of atmospheric GEM. Some explanations are listed as follows:

1, We agree with that the seasonal differences may create cluster minimizing the variance. We did take seasons into account. When we built the model with whole dataset, we have used "seasons" as an input variable. and then run the model separately in summer and winter. The accuracy of GAMs simulation was assessed using a 10-fold cross-validation test. We clarified this point in the revised manuscript **(Line 187-192).** As for the results discussion, we focused on inter-annual differences in individual seasons instead of seasonal comparisons.

2, According to your suggestion, we used the min-maximum method to standardize the parameters, including the meteorological variables, before the model running. The normalized data eliminates the effects of differences in dimension and ranges of values between indicators.

3, We have thought over the Hg species and from the characteristics of Hg species and the available representative variables **(Line 97-104).** GEM has a low chemical reactivity comparing to GOM and PBM in the atmosphere. GEM concentrations are largely affected by factors like anthropogenic emissions and atmospheric physical processes, which could be well represented by routine monitoring or easily obtained parameters, like CO, RH, BLH, etc. Whereas, GOM and PBM concentrations were strongly affected by chemical transformation processes, which have no known suitable indicators. In addition, involving chemical transformation effect in the GAM models would make it more complicate.

4, We have not done studies on the inversion model. We agree that it has great potential to bring new insight into atmospheric mercury reducibility. We will learn more about the inversion model and we are very willing to provide the atmospheric mercury observation data from the two coastal cities of China for the verification of the inversion model.

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