Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2022-367-RC1, 2022

Comment on acp-2022-367

Anonymous Referee #1

Referee comment on "Measurement report: Atmospheric mercury in a coastal city of Southeast China – inter-annual variations and influencing factors" by Jiayan Shi et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-367-RC1, 2022

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.

Specific comments:

Line 236: The authors call data from two months “trend over 2012”; however, it corresponds only to teen 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.

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

Line 243: “aggressive” what does it mean?

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

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.

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”.

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.

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

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

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.

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.

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.

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.

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 modelling. 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.