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Comment on acp-2021-753

Anonymous Referee #1

Referee comment on "Odds and ends of atmospheric mercury in Europe and over the North Atlantic Ocean: temporal trends of 25 years of measurements" by Danilo Custódio et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-753-RC1, 2021

This paper presents trends in total gaseous mercury concentrations in Europe and the North Atlantic Ocean and the regional sources affecting TGM inferred from concentration-weighted trajectory (CWT) analysis and the Positive Matrix Factorization (PMF) model. Ten to twenty-five years of TGM data at six locations were analyzed in the study. Given the long term data available, there needs to be a more detailed and deeper analysis of the data than the one currently presented. The paper summarized the distribution and general statistics for the 10-25 year period. There should be more detailed analysis comparing TGM data over various time periods and among the sites and examining changes in the frequency of Hg depletion events, etc. as well as the explanations for the TGM variability. Long term declines in TGM appear to be related to decreasing anthropogenic Hg emissions; however, Hg emissions data from the region were not presented in the paper.

The detailed methodology for the CWT and PMF analysis are missing. It is unclear why back trajectory data are combined for the five sites rather than analyzing the data for the sites individually. CWT results likely differ from one site to another. The PMF analysis in this paper was very similar to the one conducted by Custodio et al. (2020) published in this special issue for the 5-year period at Mace Head, and does not seem to provide substantially new results and insights. The PMF results are also not as detailed as those in Custodio et al. (2020) despite a longer time-series in this study. The baseline factor extracted from the PMF model needs to be described in detail given that this factor explains the largest proportion of the TGM variability. It was not clear from the paper which pollutant markers were used to assign a PMF factor to the baseline factor. The lack of a clear definition of the baseline factor makes it challenging to evaluate the effectiveness of mercury control measures that have been implemented. In my view, the source apportionment results should provide a better understanding of the different anthropogenic source sectors contributing to TGM and whether their contributions have changed over time, and if there are emerging Hg sources that we need to be concerned about. The role of re-emissions of previously deposited Hg is also not well understood.

Specific comments

Abstract. It is not necessary to describe the PMF model in the abstract. It is more meaningful to focus on the PMF factors extracted and what sources they represent. Please also clarify the last sentence of the abstract.

Lines 57-65. The updated review paper on atmospheric mercury (Lyman et al., 2020) should be discussed and cited. This review paper summarized trends in TGM and provided potential explanations for the trends.

Lines 67-70. The paper by Custodio et al. (2020) is a five year study of the TGM during the 2013-2018 period, while historical mercury releases are discussed in Horowitz et al. (2014). The timescales are very different; thus it does not seem reasonable to attribute the TGM decrease over that period to historical changes in mercury releases.

Lines 71-73. There have been several studies examining long-term trends in TGM and relating that to mercury emissions trends. Please discuss and reference these studies and provide some explanations on how this study is different.

Lines 77-79. Were 5-day trajectories analyzed over the 25-year period? Please clarify.

Line 80. Delete "On this raw,"

Line 81. Please clarify the meaning of "baseline factor"

Line 128. It is useful to label the TGM sites on a map and show the spatial distribution of the mercury point sources in Europe.

Line 130-146. Please clarify which analyzer model was used to measure TGM and the sampling interval of the TGM data. There should also be some discussion on quality control, calibration and maintenance of the analyzers.

Line 150. Was the CWT analysis conducted at five or six stations? Long term TGM data are available at six stations as mentioned in the previous section.

Lines 151-152. Some justification for the back trajectory model parameters are needed. It seems that two trajectories per day is not sufficient given that TGM data are available every 5 minutes. Why did you choose a start time of 0:00 and 12:00 local time and two arrival heights?

Line 161. "For a 120-hour trajectory duration, 84 trajectory segment end points were calculated." It's not clear how the 84 end points were obtained. There should be 120 trajectory segment endpoints for each 120 h trajectory.

Line 169. PMF stands for Positive Matrix Factorization.

Lines 171-175. It is unclear as to which year(s) of data the PMF model was applied to. Why was PMF applied to only the Mace Head site and not the other five sites? If the purpose is to examine changes in the sources affecting the TGM sites, then PMF should be applied to the yearly data or different time periods rather than the 25-year period.

Lines 177-184. The introduction to the CWT method should have been discussed in the back trajectory section where the CWT calculation was described.

Figure 1. The time-series shown is not very meaningful as it is difficult to see the variations due to the large number of data points.

Lines 188-215. A more detailed analysis of the TGM data is needed given that there are almost 25 years of data measured at six sites. For example, a comparison of the TGM data over the three time periods, comparisons between sites, changes in the frequency of Hg depletion events, etc.

Line 218. Please label the site names on one of the maps.

Line 252. LSQF has not been defined.

Lines 264-273. Some studies in North America (Weiss-Penzias et al., 2016) and Mace Head (Weigelt et al., 2015) observed a smaller decreasing trend in TGM during the recent decade. Was this observed at the six sites analyzed in this study? Explanations on long term TGM trends were provided by Lyman et al. (2020) and should be discussed in this paper.

Lines 281-294. The discussion of the trend at Villum is quite interesting. From Figure 3, there was a substantial increase in TGM from 2010 to 2014, which you attribute to melting of sea ice and subsequent GEM emissions. However, what caused the abrupt decline in TGM from 1.7 to 1.0 ng/m3 after 2014? Was that related to changes in sea ice?

Line 295. Did you analyze the CWT results for each site separately? The probable source regions are likely different depending on the site. Why were the CWT results combined for all sites?

Lines 316-342. It would be useful to include a plot of the annual Hg emissions from central Europe and discuss the changes in anthropogenic emissions alongside with changes in TGM.

Line 348-349. "We compared the regional patterns of TGM with other pollutants (CO, CO2, CH4, O3, CHCl3, CCl4, and CFCs) also measured at Mace Head and find that TGM shows a similar pattern concerning source location as the other species closely related to anthropogenic sources." Are these results shown in the paper? Have you analyzed the trends for these co-pollutants and are they decreasing?

Lines 352-367. This discussion of the CWT results and Figure 4 were discussed in the previous section. This discussion should be combined with the previous section.

Line 376-377. "The source apportionment indicates a baseline factor characterized by high load of anthropogenic species accounting for 65% of TGM mass." Has the percentage change over time? Was the PMF analysis conducted on the yearly data or the entire 25 year period?

Line 383. It is not clear from the discussion how the baseline factor was extracted from the dataset. What pollutant markers were used to assign a PMF factor to the baseline factor? Please show a plot of the factor profiles from the PMF analysis.

Line 390. What was the magnitude of the trend for the combustion factor and was it greater or smaller compared with the baseline factor? The lack of a clear definition of the baseline factor is an issue especially if the objective was to evaluate the effectiveness of mercury control measures. The focus should be on understanding the different anthropogenic source sectors contributing to TGM and whether their contributions have changed over time, and if there are emerging sources that we need to be aware of. Also, what is the role of re-emissions of previously deposited Hg? Are the source apportionment tools able to improve the understanding of Hg re-emissions?

References:

Custodio, D., Ebinghaus, R., Spain, T. G., and Bieser, J., 2020. Source apportionment of atmospheric mercury in the remote marine atmosphere: Mace Head GAW station, Irish western coast, Atmos. Chem. Phys., 20, 7929–7939, https://doi.org/10.5194/acp-20-7929-2020

Lyman, S. N., Cheng, I., Gratz, L. E., Weiss-Penzias, P., and Zhang, L., 2020. An updated review of atmospheric mercury. Science of the Total Environment, 707, 135575, https://doi.org/10.1016/j.scitotenv.2019.135575