Response to the Reviewer comments on the manuscript “Odds and ends of atmospheric mercury in Europe and over northern Atlantic Ocean: Temporal trends of 25 years of measurements”.

The authors mentioned Hg emissions inventory are inaccurate and methods of estimating emissions have changed over time, and thus argued that the Hg emissions data are not suitable for trends analysis. While emissions data have uncertainties, the data are subjected to quality assurance and quality control. The emissions data are being used to inform domestic and international policies including the Minamata Convention on Hg. It is not meaningful to quantify long term trends without explaining the underlying causes. The PMF analysis does not provide enough details on sources of Hg (only three factors were identified); thus, the analysis of TGM with Hg emissions inventory is necessary.

Response: The authors are aware of the importance of emission inventories, their assurance and quality control and their relevance to international policies. Governments use emission inventories to help determine significant sources and target regulatory actions. Emissions inventories are essential to mathematical models that estimate mercury released to the environment. Inventories also can be used to raise public awareness regarding sources of pollution. Indeed, the emission of anthropogenic sectors and their change over time can be assessed with periodic updates of the emissions inventory. Methods to determine emissions are many as continuous monitoring of emissions from a source; besides the inconsistence those methods as mentioned in response to reviewer.

However, there is a conundrum in the global inventories emission trend. It does not show the downward trend observed at many long-term monitoring stations. An upward trend is observed at the emission inventory (as shown in the figure below), even while considering only the northern hemisphere.

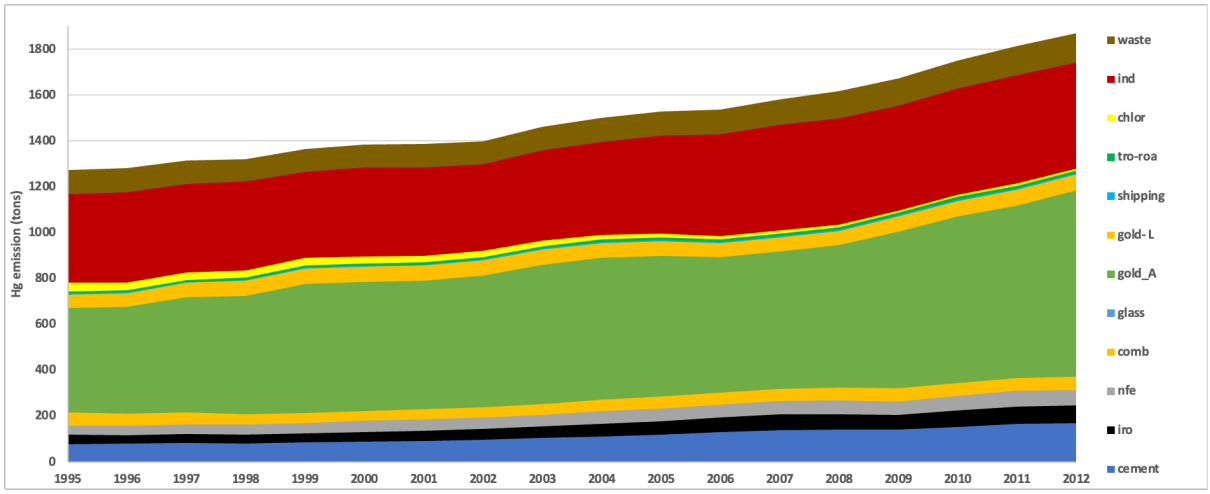

Figure 5S (reviewed manuscript): Time-series of global mercury emission. Emission inventory provided by Emissions Database for Global Atmospheric Research (EDGARv4.tox2, 2018). The inventory data is available at https://edgar.jrc.ec.europa.eu/dataset_tox4#sources. *The time-series displays the time variability of 12

sectors reported as cement production (cement), combustion in residential and other combustion (comb), glass production (glass), artisanal and small scale gold production (gold_A), large scale gold production (gold_L), shipping emission (shipping), road transportation (tro-roa), chlor-alkali industry, mercury cell technology (chlor), combustion in power generation and industry (ind), and solid waste incineration and agricultural waste burning (waste).

In addition, the 2018 Global Mercury Assessment (UN, 2018) indicate that the increase of mercury emission is linked to an increase in the primary anthropogenic sector, which estimation raised up to 20%. The 2018 UNEP Report (AMAP/UNEP, 2018) presents an inventory indicating increased emissions since the '90s.

The conundrum in the global mercury emission is already reported in the literature (e.g. Zhang et al., 2016). The missed compliance among observation and emission inventory trends has been linked to the miss estimation of Hg released from commercial products and emissions from coalfired utilities after the implementation of gas emission controls. Lyman et al. (2020, and references in) reported that the observed TGM trend is not consistent with the global anthropogenic emissions inventory, in which uncertainties ranged from -33% to 60%.

As the authors stated before, compiling a global assessment based on inventories requires several assumptions and generalizations (AMAP/UNEP, 2018). Several discrepancies are observed in the mass balance-based estimation; there are large differences between estimates. The estimation itself can explain the inconsistency among the decreasing trend observed at the monitoring stations and the increased emissions from 1990 to 2015, as indicated by anthropogenic Hg emission inventories (e.g. UN, 2018, and AMAP/UNEP, 2018).

The authors understand the importance of discussing the inventories in the manuscript, even its constrained time availability. For this reason, we presented a time series of mercury emission for the northern Atlantic and Europe in the new version (Figure 5 in the new version). The emission inventory considered by the authors is reported by EDGAR, which is based primarily upon information provided by states, local, and tribal air agencies for sources in their jurisdictions and supplemented by data developed by EPA. Even constrained in time availability, the trend observed in emission inventory for the northern Atlantic and Europe is complacent with the downward trend displayed by the observation and reported in the manuscript.

A new statement is presented in the new version of the manuscript concerning this point highlighted in the comment from the reviewer.

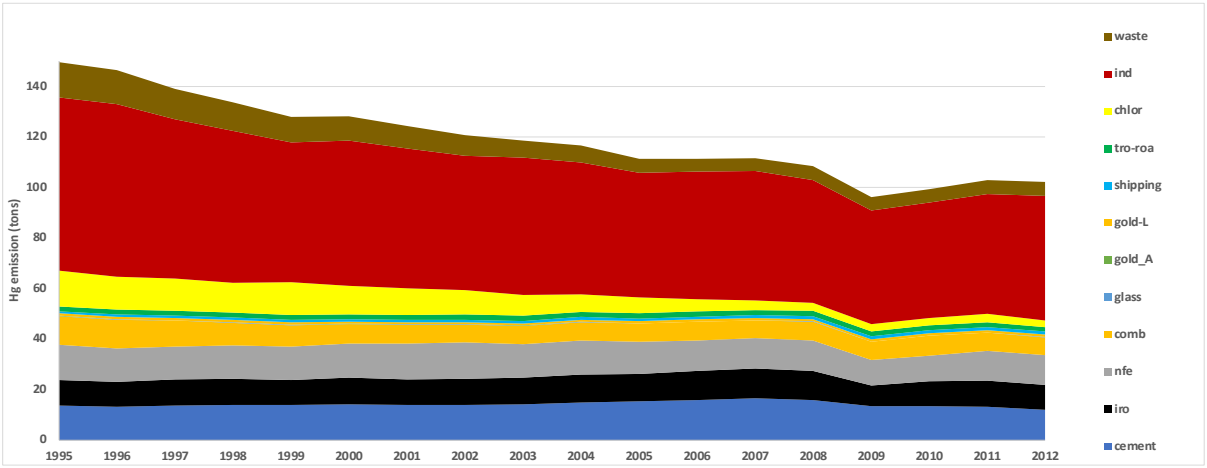

Figure 5 (in the new version of the manuscript): Time-series of Europe and North Atlantic mercury emission. Emission inventory provided by Emissions Database for Global Atmospheric Research (EDGARv4.tox2, 2018). The inventory data is available at https://edgar.jrc.ec.europa.eu/dataset_tox4#sources. *The time-series displays the time variability of 12 sectors reported as cement production (cement), combustion in residential and other combustion (comb), glass production (glass), artisanal and small scale gold production (gold_A), large scale gold production (gold_L), shipping emission (shipping), road transportation (tro-roa), chloralkali industry mercury cell technology (chlor), combustion in power generation and industry (ind), and solid waste incineration and agricultural waste burning (waste).

Based on the available dataset, the PMF factors comprised a baseline, combustion and ocean/marine factor. The baseline factor refers to background, but do we know what sources are contributing to the background factor? Does this include all Hg sources, anthropogenic, natural or re-emissions? If it includes anthropogenic sources, does it include combustion sources which was resolved in a separate PMF factor? It is important to identify specific types of sources in order to inform mercury pollution control policies. The study reported a 2.7% decrease per year in the baseline factor; however, there was no explanation on what is driving the decline in the baseline factor. Similarly what is causing the increasing trend in the marine factor?

Response: The authors are afraid that most of the questions addressed in this comment, which are indeed important, cannot be answered by the PMF solution here presented. Even neither by the current possibility of receptor model technology here deployed to assess gas mercury fluxes. The authors hope that the emission information from EDGARv4 presented in the new version gives new insights on the anthropogenic sectors driving the atmospheric mercury trend down.

As presented by the authors, the significance, implication and causes of variability in the marine factor remain to be determined. It can be associated with the changing the ocean's biogeochemistry as acidification of oceans, climate change, excess nutrient inputs, or other phenomena affecting mercury ocean-air fluxes.

The authors discussed some disadvantages and limitations of receptor models like PMF in their response. Such discussion should be included in this paper considering that a major source of

atmospheric Hg is from re-emissions of previously deposited Hg, which cannot be resolved using current receptor modeling tools. Are there other parameters that can be included in the PMF model to identify Hg re-emissions (e.g. temperature)?

Response: A new statement on the limitation of the PMF solution obtained on this study is presented in the revised version “Due to a lack of source markers that could allow the propagation of the eigenvector from axis rotation to reconstruct more realistically the complexity of mercury sources, only four factors solved our factorization. However, such an approach provided be a valuable method to evaluate mercury fluxes”. The re-emissions factor could be addressed only by a marker of such “source”.

Given the 10-25 years of TGM data available at six monitoring sites, there needs to be a more detailed and deeper analysis of the data than the one currently presented in the paper. There were no additional analyses conducted to address this comment in the revised paper.

Response: The experimental section was reworded to attend to the information requested by reviewer 2 (it appear from lines 168 to 198). In addition, an extensive description of the experimental features is avoided since it is already reported in the literature. The authors are restricted to providing the main features of the observational sites, besides information about sampling and analytical methods. Detailed information about the six sited (as well information about the sampling in each one of them) is presented at references provided by the authors.