Reviewer #1

✧ **General comments:**

In this study, the authors established long-term Hg emission maps for anthropogenic sources in China using an integrated P-CAME model. The temporal trends and spatial distributions of sectoral Hg emissions were analyzed. Key sectors and spatial hotspots of cumulative Hg emissions were identified. This dataset could provide crucial input for chemical transport models and Hg budget models. The yields of this study are of broad interest. The manuscript is well organized and written. Overall in my opinion, the manuscript is acceptable for publication on Earth System Science Data after minor revision.

Response: We appreciate the reviewer's comments and have addressed the detailed comments in our revised manuscript and the detailed responses below. Thank you for the comments!

✧ **Detailed comments and replies**

1. Introduction: Emission inventories are fundamental inputs for chemical transport models (CTMs). Applications of existing Hg emission inventories in CTMs and their performances in different regions can be introduced. The emission maps in this study could contribute to future atmospheric Hg simulations.

Response: Thank you for your comments. We have added a discussion on the applications and performance of current emission inventories in CTMs across regions and trends in the Introduction section.

"Amidst a wide array of studies, four main global emission inventories stand out for their comprehensiveness and broadly implication in CTMs (Chemical transport models): those established by Streets (Streets et al., 2011; Streets et al., 2019), EDGAR (Muntean et al., 2018; Muntean et al., 2014), AMAP/UNEP (AMAP/UNEP, 2013, 2019), and WHET (Zhang et al., 2016b). The annual emission magnitudes across inventories are ranked as WHET > Streets > AMAP/UNEP > EDGAR. Spatially, higher-emission grids are observed in WHET, Streets, and AMAP/UNEP for 2010, whereas EDGAR shows lower emissions, particularly in East and South Asia. Regarding long-term trends, EDGAR and Streets exhibit a gradual increase in emissions from 1980–2012 and 1980–2015, respectively. In contrast, WHET shows a decline followed by an increase during 1990–2010. These emission inventories have been extensively used in CTMs to simulate the atmospheric transport, transformation, and deposition of Hg. Comparing simulated Hg^0 concentrations with observations provides a critical metric for evaluating the performance of emission inventories in CTMs. Despite discrepancies among inventories in terms of emission magnitudes, species composition, and spatial distributions, a study employing the ECHMERIT model (Jung et al., 2009) reported no statistically significant differences in regression slopes when inventory-based simulations were compared with observational data (Simone et al., 2016). In terms of trends, both Streets and EDGAR indicate increasing emissions. However, when Streets inventory data were used as CTMs input, the simulated Hg^0 concentrations conflicted with the observed decline in atmospheric Hg^0 concentrations in the Northern Hemisphere during 2005–2020 (Feinberg et al., 2024). Anthropogenic emissions were identified as the primary driver of the divergence between simulated and observed Hg^0 concentrations and the associated declining trend (Feinberg et al., 2024). The WHET inventory, which incorporates updated country-specific emissions for China,

India, the U.S., and Western Europe, successfully reproduced observed atmospheric Hg concentration declines in GEOS-Chem simulations (Zhang et al., 2016b). Emission estimates from WHET for 1990, 2000, and 2010 were 1.3 to 2.4 times higher than those reported by Streets or EDGAR, highlighting the pivotal role of regional emissions in accurately capturing global emission trends and aligning them with observational data."

See revised Manuscript, Lines 48-68.

2. Section 2.1.2: The method of Monte Carlo simulation should be mentioned here instead of only in Section 2.2, with an introduction to the basic principle.

Response: Thank you for your comments. We have included an introduction to the application of Monte Carlo simulation in the calculation probabilistic technology-based emission factors in Section 2.1.2.

"To estimate mercury emissions with greater accuracy and reduced bias, Monte Carlo simulations were applied to produce probabilistic technology-based emission factors, addressing the variability and uncertainty in key parameters. Emission factors were calculated based on the provincial mercury concentration in fuel or raw materials (log-normal distribution), release rates associated with combustion or production technologies (as specified for coal-fired sectors), removal efficiencies of APCDs (normal or Weibull distributions), and the proportions of mercury species determined by APCDs combinations (Equation S2). Raw mercury concentration data and their standard deviations were sourced from previous studies (Zhang et al., 2012; Wu et al., 2012; Liu et al., 2018; Liu et al., 2019), while mercury removal efficiencies and release rates were obtained from prior research based on field experiments (Zhang et al., 2016a; Zhang, 2012; Chang and Ghorishi, 2003; Omine et al., 2012). Speciated mercury proportions for various APCD combinations were derived from our earlier work (Liu et al., 2019; Zhang et al., 2023; Wu et al., 2016). By incorporating these parameters into Monte Carlo simulations, probabilistic emission factors were generated, providing a robust and comprehensive estimation of mercury emissions across Tier 2 sectors**"**

See revised Manuscript, Lines 126-137.

3. Section 2.3: Did the authors adopted the improvement of the GEOS-Chem model in their recent study (Liu et al., 2022)?

Response: Thank you for your question. We did not use the improvements to the GEOS-Chem model as described in Liu et al., 2022. One significant limitation is the availability of long-term and consistent organic aerosol concentration. Additionally, our study primarily focuses on developing a gridded emission inventory by creating a long-term point source database, without incorporating these model adjustments. To clarify, we have provided further details on the GEOS-Chem mechanism used in Section 2.3.

"We applied a global 3-D atmospheric chemistry model (GEOS-Chem, v12.6.3, http://geos-chem.org) to simulate atmospheric mercury concentrations from 2006 to 2021. A three-year spin-up (2006-2008) was used to achieve balanced concentrations, which serve as the restart field for analysis year (2009-2021). The global simulation was conducted at a resolution of 2.0° × 2.5° to provide boundary conditions for a nested simulation over the China region, which had

a finer resolution of 0.5°×0.625° and 47 vertical levels. Meteorological input was driven by the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA2) (Gelaro et al., 2017). For the global simulation, the EDGAR emission inventory was used as it provides long-term emissions data for the entire simulation period. However, since EDGAR tends to underestimate emissions in China, we replaced China's emission with the P-CAME inventory. Biomass burning emissions were calculated based on GFED4 (van der Werf et al., 2017), while geogenic activities, soil emission and re-emission followed the calculation scheme outlined in Selin et al., (2008). The chemical scheme in v12.6.3 involves the oxidation of Hg^0 through a two-step mechanism initiated by Br. Photoreduction of Hg^{2+} occurs in the aqueous phase and is governed by the NO_2 photolysis rate and organic aerosol concentrations (Horowitz et al., 2017)."

See revised Manuscript, Lines 169-180.

4. Line 183: It should be "tended" instead of "tented".

Response: Revised.

"Overall, proxy method tended to overestimate emissions in densely populated areas"

See revised Manuscript, Line 212.

5. Line 196: It should be "reflects" instead of "reflecting".

Response: Revised.

"This trend reflected substantial shifts across key sectors"

See revised Manuscript, Line 225.

6. Lines 239–241: What is the confidence level of the uncertainty ranges?

Response: The confidence level of the uncertainty ranges is 95%CI. The uncertainty range, defined by the 2.5% and 97.5% quantiles, represents a 95% confidence interval, indicating a 95% probability that the true value lies within this range. We have added this explanation to the context.

"The uncertainty range, defined by the 2.5% and 97.5% quantiles, represents a 95% confidence interval, indicating a 95% probability that the true value lies within this range. For P-CAME emission inventory, the uncertainty range was subjected to (-16.1%, to 15.9%) in 2021, reflecting lower uncertainty in the parameters."

See revised Manuscript, Lines 269-272.

7. Line 251: It should be "NME" instead of "MNE".

Response: Thank you for your comment. Since we revised this section based on feedback from another reviewer, the original content no longer exists. We have carefully reviewed the article to ensure all instances of "NME" or "NMB" are correct, and the issue is resolved.

8. Sections 3 and 4: The sub-sections in these two sections are more like parallel ones instead

of results and discussion, respectively. Therefore, I recommend the authors to change the structure to a combined section “Results and Discussion”. More discussion is encouraged for the current Section 3.

Response: Thank you for your suggestion. We have combined Sections 3 and 4 into a single "Results and Discussion" section, as recommended, and expanded the discussion to provide more insights. This section now consists of two main parts: emissions (Section 3.1-3.4) and simulation results (Section 3.5-3.6). In the emissions part, we analyze the spatial distribution and improvements of P-CAME over the proxy method, temporal trends, comparisons with previous long-term emission inventories, and the identification of cumulative emission hotspots. In the simulation results part, we present the results of long-term simulations and evaluate the performance of P-CAME compared to the proxy method.

See revised Manuscript, Lines 196-384.