Huang et al., "Evidence for mass independent fractionation of even mercury isotopes in the troposphere"

General Comments:

This study is aimed at understanding the origin of even MIF in Hg and suggests possible mechanisms. I find that the study doesn't harness the full potential of the dataset gathered. Furthermore, very poor writing combined with flawed approaches lead to weakened findings. Several sections are speculative, grammatically incorrect and lack appropriate referencing. The key aspects such as back trajectory analysis and statistical analysis lack methodological description and are not appropriately employed.

I do not recommend the publication of this manuscript in ACP.

Specific Comments:

Line 102 -109 : This section is poorly written and needs significant improvement. The HYSPLIT modelbased air mass back trajectories shown in the supplement has not been referenced in the text. I suggest overlaying the back trajectories (Figure S6) with the cruise tracks in Figure 1.

While trajectory frequencies are useful, they are not direct enough to understand the air mass origins. I suggest the authors to conduct air mass cluster analysis using the trajectories. This would enable them to better segregate air mass origin source regions.

While computing the air mass trajectories ancillary information needs to be computed as well e.g., mixing layer height and trajectory height, rainfall etc. This will enable the authors to compute what fraction of the air mass transport occurred within the MBL. A table could be shown in the supplement.

Please note that the starting height of the trajectories (500 m) in Table S3 is too high. It should ideally be 50 m. Please ensure through cross examination if this height difference is affecting the back trajectories or not.

Line 111 -113 : Please provide more description of the sampling protocol. Why were two samplers used? Which samples were collected from which sampler at which heights ? Any blanks collected ? How many samples were collected in total ? How were the samples stored ? Which were the periods when sampling was stopped

I note that some of the information provided in the supplement has not been referenced in the text making it difficult for the reader. Every Note, Table and Figure in the Supplement should be referenced in the text in the main manuscript for enabling ease of reading.

Line 167 -168 : Why was the recovery > 100 % ?

Line 192-202 : Very poorly written section. This needs significant improvement. I understand what the authors intend to do but it doesn't translate with enough fluidity on paper. Perhaps this point is referring to the point I made above regarding computing the fraction of air mass trajectory that is within the MBL. However, it is first important to know the height of the mixing layer. The cut-offs at 500m , 1500m, 3000m may be valid but not useful until the height of the mixing layer is known.

As this is a key component of the paper, I request the authors to please recompute the back trajectories (BT) with a starting height of 50 m with ancillary information for each BT. Then conduct cluster analysis to identify source regions. Following which, they should compute the fraction of BTs

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within the mixing layer. These BTs should be shown in the main manuscript as an overlayed figure in Figure 1.

Line 204 -215: Again, a very poorly written section. This in my opinion is a key section of the methods. It is unclear what was the metric used to choose these three heights. The 'real' metric that should be used is the height of the mixing layer which the authors have not reported.

Furthermore, the Monte Carlo approach needs more description.

This choice will greatly affect the results and as such weakens the findings in the present form.

Line 269 -270 : This statement is not true. I would not refer to these poor correlations as strong in any manner. Why did the authors combine data for Cruise B&C in Figure 4b ? No justification has been provided for the same and given that they are entirely different cruise tracks and from different years which are clearly also affected by air masses from different origins, combining this dataset for a correlation plot is not correct.

Line 276 – 284: Too much speculation with no real proof. The authors do not provide a strong justification for their deduction.

Line 304 -320 : The justification for GEM oxidation pathway is not substantiated with any evidence. It is merely a speculation. Also, this section is poorly written.

Line 335: What is the abbreviation TGM ? It is not defined.

Line 313-314 and Figure 6: I find a fundamental flaw in the logic. The authors have self-imposed a cut off for the even-MIF and thereby left out a major chick of their data without any correlation in Figure 6. Also, why have the authors combined Cruise B &C data ? This approach to define even-MIF and further determine the mechanism is not convincing.

Along with this, the air mass frequencies need a real metric to determine the MBL transport and without the mixing layer height these self-imposed cut-offs (Line 382) yield speculative results.

Line 413- 417 : These numbers are speculative and self-imposed. A solid evidence is lacking and thereby weakens the discussion and main findings.

Line 490 -510 : The authors do not discuss at all how the end members were compiled and used. The authors should present the results from the Monte Carlo scheme used such as probability distribution functions and related plots. Overall in the present form the numbers are not convincing enough.