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Comment on acp-2021-699

Anonymous Referee #2

Referee comment on "Top-down and bottom-up estimates of anthropogenic methyl bromide emissions from eastern China" by Haklim Choi et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-699-RC2, 2022

Review of "Top-down and bottom-up estimates of anthropogenic methyl bromide emissions from eastern China" (Haklim Choi et al. 2021).

This manuscript presents a dataset of both methyl bromide and CFC-11 from Gosan Station, South Korea. The authors present an analysis of CH_3Br emissions based on correlating CH_3Br to CFC-11, and conclude that the bulk of these emissions are from eastern China.

Major Comments

This manuscript presents a high quality dataset, but falls short in its presentation and interpretation. The organization of the paper needs significant work, and several figures need to be changed to better illustrate the author's main points. There are several minor mistakes, which can be corrected.

MC1: Inter Species Correlations: This is the part that I am struggling with the most. While this method can work well, and can provide robust results, the authors do not present a convincing analysis here. The large outliers in either CH_3Br or F11 are dominating your regressions. To estimate emissions, I would expect that you would want representative ratios for these two compounds, and not slopes dominated by a few high pollution events.

There are ways to assess the consistency of the slopes with one of the simplest methods being to remove the largest 5, 10, and 15% of data (as an example) and run the regressions to see what the differences are. From your supplemental plots, my eye tells me you are going to get drastically different numbers for several years if you remove the outliers. However, I would suggest you consider not using regression slopes for this analysis.

Regional scale tracer-tracer ratios are typically represented by broad distributions and are decidedly non-gaussian. A alternative method is to simply ratio each enhancement and take the median value, and the uncertainty of the median, which is more robust to outliers than either the arithmetic mean or regression slopes (see Miller et al. 2012). You can further assess the individual ratios by either performing bootstrap (removal with replacement) Monte-Carlo simulations or manually removing the largest 5% or 10% of the ratios and taking the median of the reduced dataset.

MC2: Organization: I suggest the authors significantly re-organize the paper. Sections 2 and 3 are so short as to not warrant stand-alone section headings. Furthermore, sections 4 and 5 both contain methodological descriptions that could be combined with sections 2 and 3 into a comprehensive "methods" section, to be relabeled as section 2. In particular, sections 5.1 and 5.3 contain a significant amount of method description that disrupts the flow of the results and makes re-finding results after an initial read difficult. I suggest that all methodological description be placed into a new section 2: methods.

The introduction is one of the longest sections of the paper and contains a lot of information that is relevant to CH_3Br, but not specifically to the conclusions of this paper. I suggest carefully going through the intro and trimming it down.

MC3: Lack of support for possible causes, I.E. SO_2F_2: Medusa systems measure SO_2F_2, and the authors suggest that a slow transition to SO_2F_2 as a replacement for CH_3Br is a possible reason for the continued high emissions they detect at Gosan. While the authors present the Gosan SO_2F_2 record, they do not do any further analysis than the potential source regions are similar (which is trivial given the nearly identical uses of both chemicals) and showing pollution events are concurrent in time. Given that SO_2F_2 is commonly used to replace CH_3Br, it would strengthen the paper to add an analysis of this compound to support the notion that China is not using SO_2F_2 and rather is choosing to violate the Montreal Protocol.

MC4: Several points in the paper are underdeveloped/lack explanation: Firstly, the authors focus in on eastern China, and then do not present analyses of any of the other source regions they show in figure 5. Why are Korean emissions not also assessed? Or Japan (even though they are likely to be low based on figure 5)? Based on Figure S2b, air masses from Korea seem equally as elevated in CH_3Br as the air masses originating in China. Without these analyses, the support for the conclusion that emissions, and overall atmospheric burden, are almost entirely from eastern China is incomplete.

Minor Comments

For entire paper: the term "concentration" is used throughout. Please change to "mole fraction" as the AGAGE data is published as mole fraction. See IUPAC Green Book for further reference (https://iupac.org/what-we-do/books/greenbook/)

L26: -0.13 ppt yr^{-1} is not the number I get when I divide 1.1 by 12.

L118-119: Add countries for both sites in addition to Lat/lon. Additionally, I suggest adding a panel to Figure 1 with the globe showing the location of Gosan, Mace Head, and Cape Grim. This allows the reader to quickly see the spatial relationship between the three sites at a glance, rather than having to look it up.

Section 3: Add in justification for why Mace Head and Cape Grim are appropriate comparison sites to Gosan. Given CH_3Br's lifetime of about 9-10 months, I'd like to see more explanation of why CH_3Br at Cape Grim is included here.

L124: State which site the baseline mole fractions declined for.

L138/139: State the dates explicitly for the periods of missing data.

Figs 3 and 4: Given the missing data, the reader is left wondering how the authors have dealt with the lower number of samples during 2016, 2017 and 2018. For example, Figure 3 shows monthly means for 3 year periods, yet based on figure 2, for 2017-2019, the months of March, April, and possibly May are really only 2019 data, as no data exist for those months for 2017 and 2018, at least from eye balling figure 2. See comment above about stating the periods of missing data explicitly.

L143: HYSPLIT citation is out of date per HYSPLIT website: see Stein et al., 2015 AMS: https://doi.org/10.1175/BAMS-D-14-00110.1.

L155 and section 4 overall: It is worth noting that while this section details and discusses the HYSPLIT back trajectories, at Line 155, the authors reference Figure S1, which denotes that FLEXPART was used. Please clarify what specific model you are using for this part of the analysis.

L170 and figure S2a/b: It would be helpful if these two figures shared the same color scheme. I realize the colors are denoted in the legend, but in S2b, east China is bright red, wheras in S2a, bright red corresponds to the sea of South East. Korea is green in S2b and lumped into one category, yet neither North Korea nor South Korea are green in S2a. It is easier for the reader to quickly interpret the pollution events per zone in S2b using S2a if the colors match between figures.

L219 and 221: Equation 2: please subscript α, E_{MB}, and E_{CFC-11}. i.e $\sigma_{E-MB} = \sigma_{E-CFC-11} + \sigma_{\alpha}$. As it is written it is easy to mistake this as $\sigma * E_{MB}$, etc…

Section 5.3: The term "significant" is well defined in statistics, and without supporting statistical analysis to show the significance of these regression slopes, I do not find the R values presented here convincingly significant. Additionally, the Pearson correlation coefficient R is known to be non-robust in the presence of outliers, and given the large outliers in your data, the R values are almost certainly biased. See for example Devlin et al. 1975 (doi: 10.2307/2335508), or Zhou 1987 (doi: 10.1007/BF00897747).

Furthermore, figure S4 is somewhat integral to your paper and should be included in the

main text, and not in the supplement.

L355: This sentence has a typo or is missing a word after soil, I believe.

L359: same comment as above for L26.

L369-374: This is confusing. You state there is a 3 Gg yr^{-1} discrepancy and then state there is an additional 3.5 Gg yr^{-1} discrepancy. What is this additional discrepancy?

L395: I went to download the data and while this link does provide access to the AGAGE Gosan station data, it a) does not include CH_3Br, and b) does not contain the data as presented in the paper (i.e) with the background filter applied. Please add the CH_3Br data at a minimum, and it would be nice to have this papers data set with all years included, and an additional column marking the samples as background and pollution as per the filter used in this paper.