Referees' comments on "Continuous increase in East Asia HFC-23 emissions inferred from high-frequency atmospheric observations from 2008 to 2019"

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We thank the referees for their thoughtful and thorough reviews. We are pleased that all the reviewers see our manuscript as a valuable contribution to the field. We have taken almost all of the reviewers' suggestions and concerns into account in the revised version, as we note in detail below, and we feel we have been able to make an even stronger presentation of our findings in light of their insights and questions. We thank the reviewers and the editor for their time and effort and appreciate the recommendation for publication in Atmospheric Physics and Chemistry. [In the following, Reviewers' comments are in bold Courier New and our responses and are in Time New Roman font]

Reviewer comments:

Referee #1:
Primary comments:
This manuscript describes important emission information on key Montreal Protocol controlled and climate related gases. It should be fully published.
The methods used to determine the emissions are widely accepted and appropriate for this study.

My primary concern is one that can be remedied with some careful attention to the discussion to make the reading of the flow a bit better. The discussion is rather disjointed, with a bunch of factual statements of decisions made for the analysis.
>>> Based on the reviewer's perspective, we realized that discussions should have been better structured in the previous version without including HCFC-22 observations and emission estimates in the main text. They are now discussed in the Supplementary Information. The HCFC-22 production section of the original manuscript (3.4 Estimation of HCFC-22 production in eastern China) has now been moved to a newly added subsection (2.3 Bottom-up Emissions Estimates) of the Method section to clarify that the HCFC-22 production information for eastern China is linked to the inventory-based bottom-up HFC-23 estimation. We do think the revised manuscript has been improved according to reviewer's suggestions. Thanks for the reviewer's editorial comments!

There is little discussion of why the decisions were made for many of the assumptions. Such discussion would help to make the logic flow a bit better. One such example is at line 220. An

assumption is mentioned with no statement of why. The same is true for the discussion of the results.

>>> There were three assumptions in the original section 3.4 of the Result and Discussion section. The statement at line 220 in the previous version that "we assumed the eastern China production fractions were correlated exponentially with time", has now been clarified with an addition of the following clause, and is now shown in a new subsection 2.3 of the revised Method section: "because HCFC-22 production rates in developing countries exhibited an exponential growth over time until mid-2010 (https://ozone.unep.org/countries)". The second assumption was that China HCFC-22 plants information reported to UNEP was correct, because we estimated the annual fractions of eastern China HCFC-22 production to the Chinese total based on those reported data. As stated in the text, even if unreported and/or newly built plants exist at unknown locations, our estimation of the abated HFC-23 emissions for eastern China, for which the fractions were primarily used, would not be affected because the emissions abatement action under both the CDM and HPPMP programs must have been taken in the reported facilities. The third one was that a varying rate between the 2015 and 2018 production fractions in eastern China can be applied consistently over time. Given the limited information, the extrapolation approach would not be considered unreasonable.

All the discussion related to the figures (which I mostly like) does not naturally flow. For example, a better discussion of the comparison of HFC-23 vs. HCFC-22 emissions and what that REALLY tells us would be helpful.

>>> We have reorganized the figures in the Results and Discussion section by moving the original Figures 5, 6, and 7 (the timeseries plot of atmospheric HCFC-22 observations, observation-derived annual HCFC-22 emissions in eastern China, and annual HCFC-22 production fractions for eastern China, respectively) into the Supplementary Information (Figures S6, S7 and S3, respectively). We think this organization is more concise than our originally submitted version of the manuscript.

The agreement between our top-down HCFC-22 emissions in China and inventory-based bottom-up emissions estimates was taken to suggest that the HCFC-22 production information linked to the bottom-up estimates would also be reasonably accurate, which has been stated both in Section 3.6 and in the completely rewritten Conclusions.

A useful discussion point that this paper COULD address is the distribution of the inferred emissions relative to what is seen for CFC-11.

>>> The spatial distribution of the top-down CFC-11 emissions in eastern China (Rigby et al., 2019; Park et al., 2021) showed large emission flux in or around the provinces of Shandong and Hebei for the periods 2014–2017. Given that the emissions of CFC-11 primarily occur during foam blowing, rather than directly during production, the regions where emissions have been identified are not necessarily the same as the locations where the compound has been produced. Whereas the high emission flux densities of HFC-23 were found around the

provinces of Shandong and Jiangsu and were relatively well correlated with the location of known HCFC-22 factories. Considering their distinct emission sources (i.e., CFC-11 use-related emission vs. HCFC-22 production-derived emission), it is unclear whether we could expect a certain correlation between the emission distributions of CFC-11 vs. HFC-23. Also note that it is not possible to discern the location of emissions sources at finer scales than provinces, or groups of provinces in eastern China. Therefore, an explicit discussion of the spatial distribution of HFC-23 emissions in comparison with that of unexpected CFC-11 emissions is beyond the scope of the current manuscript, and thus we have used the annual top-down estimates of CFC-11 only to validate our modeling performance, as in our originally submitted version of the manuscript.

Other areas that could be described more clearly are how the priors are really constructed relative to the TEAP information regarding the location of the probable production facilities. It was not clear to me how the initial emissions were distributed among the various facilities for the priors, and how they changed.

>>> We use an ensemble of 27 different inversion runs with a range of prior emissions, which have three different prior flux distributions combined with nine different total prior emission magnitudes with corresponding uncertainties. Detailed information on prior construction is provided in the section of "Three different spatial distributions of a priori emissions" in the Supplementary Information.

As another check, we examined a specific point source distribution for prior emissions based on the likely locations of HCFC-22 production facilities. All plant locations were presumed to have an equal emission flux, which was assumed to be the average of unabated HFC-23 emissions derived from TEAP-based production of known HCFC-22 facilities. For all other areas, 20% of the population density distribution was applied to ensure some minimum prior emissions for all inversion grids. The resulting posterior emissions were consistent with those run by the population density distribution (see the plot appended below).

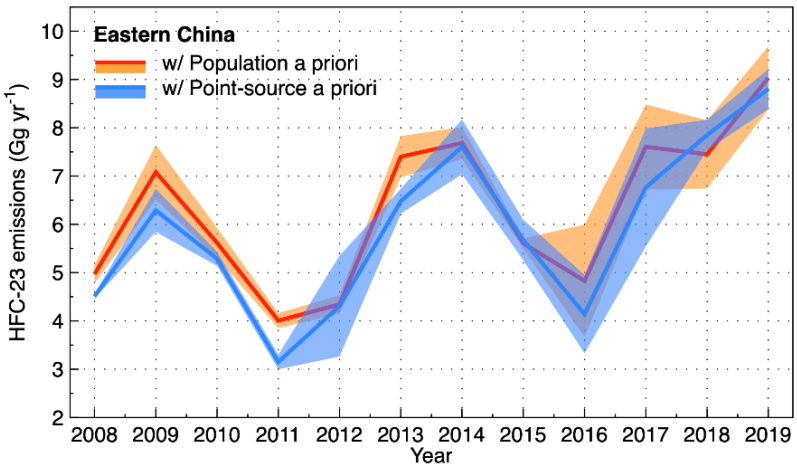

Figure: HFC-23 emissions from eastern China derived using two different *a priori* distributions: "Population" and "20% of population-based point source" *a prioris*. Each line represents the annual mean of nine different model set-ups for each *a priori* distribution. Shading denotes 2σ uncertainties.

However, we decided not to include the point source distribution in our inversion analysis based on the following reasoning: (1) potential bias due to inaccuracies in the point source information could be problematic because details on the retirement period of each plant, production magnitudes, and changes in production capacity were not fully available; and (2) applying a point source distribution for prior emissions, which is constructed based on locations of the known HCFC-22 plants, would have already presumed that inferred HFC-23 emissions should be strongly associated with the known HCFC-22 production. Therefore, this approach would not be relevant to investigate the potential existence of unreported, unknown HCFC-22 production.

A few minor points are below…

Abstract: How are the known production locations used in the analysis?

>>> The production plant locations were compared with the spatial distribution of the inferred emissions in Figure 3 (which was Figure 4 in the originally submitted version). All available information we could find about the facilities was summarized in Table S1 in the Supplementary Information. As noted above, it is unfortunate that we were unable to provide complete information about how long the plants existed, how large their production capacities were/are, and how the production magnitudes have been changed.

Line 39: the term "basket" is a bit informal. I'd change it to "group"….though did Kyoto really regulate?

>>> Changed.

The term "top-down" is loosely defined in the abstract, but not in the body of the paper. I suggest doing so, and with a bit more detail than what is done the abstract.

>>> We have updated the text in line 23 to read as follows: "A recent study on atmospheric observation-based global HFC-23 emissions (top-down estimates) showed significant discrepancies over 2014–2017 between the increase in the observation-derived emissions and…".

Figure 3…so that these maps can be shown without the caption, I'd put the years of the analysis in the upper left corner of each map.

>>> A good suggestion. Updated.