Referees' comments on "Toward resolving the budget discrepancy of ozone-depleting CCl_4: An analysis of top-down emissions from China" *by Sunyoung Park, Shanlan Li, Jens Mühle, Simon O'Doherty, Ray F. Weiss, Xuekun Fang, Stefan Reimann, Ronald G. Prinn*

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 made changes to the manuscript to answer the suggestions of the reviewers and clarified a few points raised in review. We respond to the referee's comments below and a revised version of the manuscript including most of the changes suggested by the reviewers will be submitted to the editor. 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:

Paper Summary: This paper uses observations from the Korean Gosan station to identify the location of CCl_4 sources and the specific industrial processes involved with the CCl_4 emissions. There are two basic techniques used to analyze these data. First, trajectories are used in a source/receptor analysis technique to identify the CCl_4 emission locations. The major sources originate in Eastern China. Second, a positive matrix factorization (PMF) analysis technique is used to finger-print the specific sources of CCl_4. This analysis reveals that the emissions are primarily from chloromethane production (CH_3Cl), perchloroethylene production, and fugitive emissions from feedstock usages. These estimates are larger than those from the SPARC CCl_4 report, with the fugitive emissions being 10x larger than SPARC!

Review Summary: This is an excellent paper that NEEDS to be published. My overall comments are with regard to improving the writing in the paper and some of the structure.

Paper Suggestions:
While the paper mentions the SPARC (2016) CCl_4 report, there ought to be more discussion of how this paper seems to resolve the discrepancy between their emissions based upon measurements. SPARC had a top-down emissions calculation of 40±15 Gg/y, a hemispheric gradient method of 30±5 Gg/y, and a regional emission

estimates of 21.4±7.5 Gg/y. The SPARC regional 21.4 Gg/y had a 15 Gg/y contribution from China. The higher estimate herein of 24 Gg/y from China would bring this 21.4 SPARC number up to 30 Gg/y - in precise agreement with the gradient method and within the uncertainties of the 40 Gg/y top down estimate.

>>> Yes, we agree with the reviewer. This is an important point to mention and we have included the following sentences in the second paragraph of the Conclusions section: "According to Liang et al. (2016), global top-down emissions were estimated to be 35 ±16 Gg yr^{-1} as an average estimate between 40±15 Gg yr^{-1} based on the new 33-year total lifetime of CCl_4, and an independent top-down method using the observed inter-hemispheric gradient in atmospheric concentrations which yielded 30±5 Gg yr^{-1}. The SPARC sum of the regional emissions estimates of 21±8 Gg yr^{-1}, where Chinese emission of 15 (10–22) Gg yr^{-1} contributed 71±33%, was still less than the aggregated top-down values. Instead, if we apply the higher emission estimate of 23.6±7.1 Gg yr^{-1} from China suggested here, the summed regional estimate would be calculated to be 30±10 Gg yr^{-1}, largely in agreement with the best estimate of global emissions of 35 ±16 Gg yr^{-1} from Liang et al. (2016)."

The discussion in the summary of the CCl4 sources should be broken out with more definitive statements. The SPARC report used industrial estimates to characterize potential sources [Sherry et al., 2016], and this paper provides the **first observational basis for these sources, but this paper also makes the case that Sherry et al. is perhaps too conservative in their estimates.**

>>> This comment is also very helpful. The factors were re-named as (A), (B), (C) and so on, and their descriptions in the section of "Industrial source apportionment of atmospheric CCl_4 in East Asia" were also updated to make it easier to compare them to the SPARC report, as suggested by reviewers 1 and 2. The figure legends in Fig. 4 were changed accordingly.
As suggested, we've revised the conclusions to better discuss a link of the industrial sources identified from a factor analysis based on atmospheric observations to the SPARC bottom-up inventory-based estimations.
The revised conclusions now read: "A factor analysis combining the observed concentration enhancements of 18 species was used to identify key industrial sources for CCl_4 emissions and to link our atmospheric observation based top-down identification of potential sources with bottom-up inventory-based estimates (e.g., Liang et al., 2016; Sherry et al., 2017). Three major source categories accounting for 89±6 % of CCl_4 enhancements observed at GSN were identified as advertent or inadvertent co-production and escape of CCl_4 from CH_3Cl production plants (factor (A)) and during industrial C_2Cl_4 production (factor (C)), and fugitive emissions (factor (B)) from feedstock use for the production of other chlorinated compounds (e.g., $CHCl_3$) and process agent

use. These sources are largely consistent with the bottom-up CCl_4 emissions pathways identified in SPARC (Liang et al., 2016). The SPARC estimate of global CCl_4 emissions from chloromethanes and PCE plants (pathway B from Liang et al. (2016) and Sherry et al. (2018)) was 13 Gg yr^{-1}, as the most significant source. Fugitive feedstock/process agent emissions, denoted as pathway A by Liang et al. (2016) and Sherry et al. (2018), were estimated to be ~2 Gg yr^{-1}. These emissions for pathways B and A had contributions from China of 6.6 Gg yr^{-1} and 0.7 Gg yr^{-1}, respectively.

If we assume that the emission rates from the sources correspond to the relative contributions of the corresponding source factors to the total Chinese emission rate (23.6±7.1 Gg yr^{-1} for the years 2011–2015), source factors (A), CCl_4 emissions from chloromethane plants, and (C), emissions from PCE plants, amount to 13±4 Gg yr^{-1} for China. This is as high as the global bottom-up number of 13 Gg yr^{-1} for pathway B emissions, and more than 50% higher than Chinese estimate of 6.6 Gg yr^{-1}. This could point to a higher than assumed ratio of CCl_4 being emitted from these processes into the atmosphere, although factor (C) could possibly include influence of fugitive emissions as a chlorination feedstock for PCE production. Furthermore, also source factor (B), fugitive feedstock emissions are estimated at ~7±2 Gg yr^{-1} from China alone, which again contrasts with Chinese estimate of ~0.7 Gg yr^{-1} and even a lower global estimate of only 2 Gg yr^{-1} for pathway A from Liang et al. (2016) and Sherry et al. (2018)."

The paper is fairly well written, but many of the current paragraphs need to be broken up into more distinct sections or primary thoughts. The extended paragraphs of the current version obscure the thoughts, logic of the paper, and the overall content of the text.

>>> Based on the reviewer's perspective, we realized that discussions should have been better structured in various places in the previous version breaking a long body of paragraph by a single topic. We do think the revised manuscript has been improved according to reviewer's suggestions. Thanks for the reviewer's editorial comments!

For example, the 2nd para of the Introduction (P2, 4-30) talks about top down emissions, bottom up emissions….I would break this up into paras on: 1) top down emissions (4-12); 2) a SPARC bottom up para (12-16); and 3) a discussion of regional emissions.

>>> As the reviewer suggested, we divided this long paragraph into three to make it easier to follow. First, we started with a discussion about the updated bottom-up emissions in the SPARC report, and introduced the global top-down and hemispheric gradient top-down emissions, pointing out that the revised bottom-up estimate of 25 Gg yr^{-1} is still lower than the average SPARC-merged top-down emission estimate of 35±16 Gg yr^{-1}. Then we added the summed

regional emissions estimate from Australia, East Asia, U.S. and Western Europe, and mentioned its lowering than the global total and the relative significance of East Asia contribution.

In the 1st para of section 3 (P. 4 line 18 to P.5 line 32 - 46 lines!), there are a broad range of paragraph thoughts. The paragraph starts with a discuss of the interspecies correlation and ends with a thought on an underestimate of Chinese emissions. Please break this up to improve the flow of the text.

>>> We have broken up the original, long paragraph, which is now in section 4 of the revised version, into four paragraphs corresponding to "introduction to an interspecies correlation method", "a reference compound and its emission estimate", "determination of the empirical correlations between the observed enhancements of CCl_4 and reference, HCFC-22" and "comparison of the annual CCl_4 emissions in China estimated in present study with previous results".

The "Data Overview" section both discusses the data and shows results. I would restructure sections 2 and 3 into: a data, methods, and results sections. The Supporting Information ought to flow better into these data and methods sections.

>>> We have completely restructured section 2 of the manuscript by breaking it up into two sub-sections (2.1. Measurements of CCl_4 at Gosan and 2.2. Results), and one independent section (3. Potential source regions of CCl_4 in East Asia). The new section 3 is comprised of three paragraphs: introduction to trajectory statistics as a tool to illustrate the regional distribution of potential CCl_4; input data and conditions for calculation; and description of the resulting map of potential source areas. We've also added specific information on corresponding SI text accordingly in the new section 3. Air mass source country classification that had been discussed in the last paragraph of Data overview section in the original manuscript, now moved to the beginning of section 4, as a transitional paragraph to the following country-specific emission analysis.

Again, break up the single paragraph of the conclusions into short paragraphs. The main messages are lost in this "run-on" paragraph.

>>> We have re-organized the conclusions with four short paragraphs. We hope this can convey ideas more clearly to readers. For the text revision, please see the earlier response.

Figures are good. For Fig. 4, put some vertical lines on the plot to see how the bars line up with chemical names at the

bottom.

>>> Done

Fig. S5. What are the colors for? Do they indicate statistical significance?

>>> We now say in the figure caption: "The colors by shade indicate statistical significance."