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Comment on acp-2021-857

Anonymous Referee #1

Referee comment on "Global emissions of perfluorocyclobutane (PFC-318, *c*-C_4F_8) resulting from the use of hydrochlorofluorocarbon-22 (HCFC-22) feedstock to produce polytetrafluoroethylene (PTFE) and related fluorochemicals" by Jens Mühle et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-857-RC2, 2021

The manuscript by MuÌ□hle et al. investigates the relationship between calculated global emissions of PFC-318 and estimates of HCFC-22 production used as feedstock for production of fluorinated ethylene products used to manufacture Teflon. This manuscript extends previous work (MuÌ□hle et al., ACP, 2019) by updating several years of PFC-318 emission and by looking in more detail at the use of HCFC-22 as a feedstock. The material is well-presented, the underlying data on PFC-318 temporal variation is excellent, and the model used to evaluate emissions has been applied successfully to other AGAGE data. I am not sufficiently expert to evaluate the accuracy of the HCFC-22 feedstock production data, which is the other significant compilation of this manuscript. Any errors in these reported production numbers will impact the conclusion, and, if relevant, the authors should comment on the reliability of these reports (especially since inconsistencies are noted for other gases regulated by the Montreal Protocol).

The major contribution of this paper is the observed linear relationship between HCFC-22 feedstock production and PFC-318 emission rates after about 2002. Attribution of the increasing PFC-318 emission due to industrial activity in China is also interesting. However, I thought the manuscript could have done a better job in describing why the relationship observed after about 2002 or so, is different from the preceding 10 years, and what was contributing to PFC-318 emissions prior to 1990. The unknown nature of the period prior to 2002 adds a level of uncertainty to the analysis that is not discussed. Between 1990 and 2002, the calculated PFC-318 emission rate remained nearly constant (even decreasing early in the period) while cumulative HCFC-22 FS production totaled about 1500 Gg. For approximately the same HCFC-22 FS production from 2005-2010 (1405 Gg), the calculated PFC emission rate increased from 0.96 to 1.43 Gg/yr. Similarly, the last part of the record shows constant HCFC-22 FS production but continued increase in PFC-318 emission. How is this rationalized? I would like to understand the different emission response of PFC-318 to HCFC-22 production to have better confidence in the quantitative results presented. The correlation presented looks compelling but deserves more detailed analysis of potential factors that might contribute to the correlation.

It might have also been interesting, if the data is available, to discuss the relative importance of HCFC-22 FS production to other sources of HCFC-22 and to the global

burden and trend of HCFC-22. AGAGE data shows a consistent linear increase in PFC-318, while the rate of HCFC-22 increase has slowed. Does this mean that HCFC-22 FS emissions are becoming a more significant component of ambient HCFC-22 relative to other changing sources? In a similar vein, the authors mention CHF3 associated with the production of HCFC-22, and thus it has a source related to PFC-318 emission. Can temporal trends in CHF3 vs PFC-318 observed at AGAGE sites provide additional insight into the relative importance of the different source emissions over time? Data from at least one of the AGAGE sites (THD) seems to show a change in slope of the relationship between CHF3 and PFC-318 occurring around 2015. It seems a shame not to get better use of the excellent measurements from the AGAGE sites. This may well be beyond the scope of the manuscript intended by the authors, but it could make the manuscript more interesting.

Other minor comments/questions:

Line 84: Given the locations of the sites used for the 12 box model, it brings up the question of how much impact was observed in the pollution events that are removed from the data set for further analysis? And, if significant, could further analysis of these pollution events provide further insight into regional or long-range sources? Though not critical, it would be interesting to know the fraction of data that were removed due to pollution filtering.

Line 110: The authors note that the results here agree with MuÌ□hle et al., 2019. Aren't the data, model, and methods identical between this study and earlier? I thought only the last few years of data were new.

Figure 2: The black diamond curve is not identified in the caption or figure as the PFC-318 emission rate.