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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global emissions of HFC-143a (CH_3CF_3) and HFC-32 (CH_2F_2) from in situ and air archive atmospheric observations" *by* S. O'Doherty et al.

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On conclusions to be reconsidered: p. 6481-6482. The authors interpret changes in the ratio of mole fractions at Mace Head (56N) vs Cape Grim (40S) over time as suggesting that "the pattern of emissions is changing over time" and furthermore, that this "implies decreasing emissions in the NH and/or increasing emissions in the SH". While this ratio is effected by the inter-hemispheric distribution of emissions, it is also effected by the magnitude of emissions relative to the trace gas global atmospheric burden. Consider, for example, constant emissions (NH only) of a long-lived trace gas into an atmosphere with an initial concentration of zero. Over time, the NH/SH mixing

ratio will change dramatically (large initially and then decreasing) until steady-state is reached (and a constant N/S ratio), despite emissions and their inter-hemispheric distribution (NH only) being constant over the entire period. I imagine that this effect is the overriding reason for the observed change for this ratio over time that the authors discuss. Hence, this section needs substantive rethinking and revision.

We agree with the reviewer, the interpretation was too simplistic and will be removed from the text.

Furthermore, the discussion on lines 5-18 (p. 6482) is confusing and should be reconsidered (and whether or not Table 2 is a useful addition). It isn't clearly described how the emission ratio of HFC-143 to HFC-32 in an Australian inventory, or derived from C. Grim (is this also an estimate of "Australian" emissions?) help the reader understand NH vs SH emission magnitudes and their potential variation over time.

We agree with the reviewer, the discussion of the Australian inventory is misplaced and will be removed from the introduction (as will the associated Table 2).

On prior emissions and their uncertainty: p. 6480, lines 3-18. What emissions from UNEP reports? I presume you mean emissions reported to the UNFCCC here? If so, why are UNFCCC reported values, which are known to be underestimates of global emissions, used with EDGAR estimates to constrain the uncertainty on the a priori emission magnitudes (30%)? To what extent are the posterior emissions effected by the prior emission magnitudes and their uncertainty?

We thank the reviewer for pointing this out. A priori emissions were obtained from EDGAR, not, as stated, from the UNFCCC reports. However, we did use the comparison of the EDGAR and UNFCCC emissions growth rates to derive an order of magnitude uncertainty estimate. We acknowledge that this is likely to be a relatively poor approximation of the "true" uncertainty in the EDGAR dataset (partly because of the under-reporting noted by the reviewer). However, we do not find that the choice of uncertainty had a significant impact on the conclusions of this paper. The close

agreement between the “optimized” model and the observations indicates that the prior emissions estimate had a relatively minor influence on the derived emissions. We have modified the text to clarify which a priori emissions dataset was used.

p. 6474 HFC-23 has a lifetime of Âż50yr.

Changed the text, “HFCs are partially fluorinated hydrocarbons that have atmospheric lifetimes in the range of 1-50 years (with the exception of HFC-23 and HFC-236fa which have lifetimes of 222 and 242 years, respectively)...”

L 21, Reconsider the intent of this sentence, as it comes across as misleading. The US reports emissions data to the UNFCCC even though it didn't ratify Kyoto.

Changed the text, “However, countries that did not ratify the Kyoto Protocol (Non-Annex I or developing countries and the USA) were not required to submit national HFC data, although some countries such as the USA do”.

L 23, projected for what year? Is this sentence and the following one a discussion of the past or future?

The text is unclear, and has been changed to answer the reviewer's questions:

“It has been suggested that the total GWP weighted HFC emissions for baseline scenarios in 2050 will be $\sim$4 times larger than those previously reported in the Intergovernmental ...”

Citations are limited, and often include only those associated with the author team. Important points related to HFCs that should be referenced are not.

We agree with the Reviewer and have now included additional non-author team citations:

Fang, Xuekun, BR Miller, and SS Su. 2014. “Historical Emissions of HFC-23 (CHF3) in China and Projections upon Policy Options by 2050.” Environmental Science & Technology 23. doi:doi.org/10.1021/es404995f.

http://pubs.acs.org/doi/abs/10.1021/es404995f.

Laube, J. C., P. Martinerie, E. Witrant, T. Blunier, J. Schwander, C. a. M. Brenninkmeijer, T. J. Schuck, et al. 2010. “Accelerating Growth of HFC-227ea (1,1,1,2,3,3,3-Heptafluoropropane) in the Atmosphere.” Atmospheric Chemistry and Physics 10 (13) (July 2): 5903–5910. doi:10.5194/acp-10-5903-2010. http://www.atmos-chem-phys.net/10/5903/2010/.

Miller, B. R., and L. J. M. Kuijpers. 2011. “Projecting Future HFC-23 Emissions.” Atmospheric Chemistry and Physics 11 (24) (August 16): 13259–13267. doi:10.5194/acp-11-13259-2011. http://www.atmos-chem-phys-discuss.net/11/23081/2011/.

Miller, John B., Scott J. Lehman, Stephen a. Montzka, Colm Sweeney, Benjamin R. Miller, Anna Karion, Chad Wolak, et al. 2012. “Linking Emissions of Fossil Fuel CO 2 and Other Anthropogenic Trace Gases Using Atmospheric 14 CO 2.” Journal of Geophysical Research 117 (D8) (April 19): D08302. doi:10.1029/2011JD017048. http://doi.wiley.com/10.1029/2011JD017048.

Montzka, S. a., L. Kuijpers, M. O. Battle, M. Aydin, K. R. Verhulst, E. S. Saltzman, and D. W. Fahey. 2010. “Recent Increases in Global HFC-23 Emissions.” Geophysical Research Letters 37 (2) (January 29): n/a–n/a. doi:10.1029/2009GL041195. http://doi.wiley.com/10.1029/2009GL041195.

Consider updating GWP with the latest IPCC report values, and lifetimes with those from the recently published SPARC report.

We agree, and this has been done using the references detailed below:

Myhre, G., Schindell, F-M. Breon, W. Collins, J. Fuglestvedt, J. Huang, D. Koch, J.-F. Lamargue, D. Lee, B. Mendoza, T. Nakajima, A. Robock, G. Stephens, T. Takemura and H. Zhang: Anthropogenic and Natural Radiative Forcing. In: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K.

Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Ko, M., P. Newman, S. Reimann, S. Strahan (Eds.), SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species, SPARC Report No. 6, WCRP-15/2013.

The introduction contains much information not all that relevant to the main point of the paper. Also, why are emissions derived from Culbertson tabulated in the introduction instead of being discussed (or possibly displayed) along with the new results in Figures and or Tables? (name misspelled on p. 6476, line 5)

We feel the historical mole fraction data for HFC-143a reported in the Culbertson paper was an important addition to the introduction as a marker for the type of work that had preceded this study. It was initially decided not to present the Culbertson HFC-143a emissions data alongside the new results since they are reported as average calculated emissions over a five-year period and not as annual averages. In hindsight we have now added these data to the Figure 5 for comparative purposes (see revised Figure 5).

Corrected misspelling on p6475, line 25 and 6476, line 5.

p. 6481, It is stated that the radiative forcings associated with these two HFCs are estimated with the AGAGE 12-box model. I don't believe this is entirely accurate; some clarifying text is needed here.

The text should have read: "The radiative forcing due to HFC-143a estimated using the global average mole fraction obtained from the AGAGE 12-box model, was 1.7 ± 0.04 mWm-2 in 2012." and "The radiative forcing due to HFC-32, estimated using the AGAGE 12-box model global mole fractions, was 0.7 ± 0.02 mWm-2 in 2012". We have corrected this in the text.

The consideration of East Asian sources of these gases is interesting in light of the

discrepancies in global emissions derived in this work and those reported to the UNFCCC. In this discussion, the years associated with emission magnitudes derived in the regional studies need explicit speciïňĄcation.

The estimation period is from November 2007 to December 2008, I have now detailed this in the manuscript:

"Li et al. [2011] reported emission estimates for East Asia (China, S. Korea, Japan and Taiwan) for the period between November 2007 and December 2008, where the emission rates for each country were estimated using an interspecies correlation method. . ."

Are there any independent indications that the methods used in these regional studies provide reliable estimates of emissions from this entire region? Some discussion of limitations of regional studies seems necessary.

The regional studies quoted in the text have been published in peer-reviewed journals and detailed discussions of the regional studies and associated limitations are reported within these references.

Furthermore, the emission Tables mention that "Estimates of global emissions based on ...measurements...collected primarily at Trinidad Head and Cape Grim using the AGAGE... model." What does "primarily" mean here? Are the emissions derived from results only at these two sites despite the availability of data at many more sites? This should be explicitly discussed in the methods section.

In the original text the "primarily "referred to the fact that most NH flask samples were collected at Trinidad Head, but other flasks collected at different NH sites were also analysed. This section of text has been reworded for clarity:

"Table 3. Estimates of global emissions of HFC-143a (Gg yr-1) based on AGAGE in situ measurements and archived air samples (the NH flasks were collected primarily at Trinidad Head and the SH flasks at Cape Grim) using the AGAGE 2-D 12-box model. Estimates of total emissions prior to November 2003 are based only on archived air

samples. Also shown are global emission estimates derived from UNFCCC National Inventory Reports (2012 submission), and from the EDGAR (v4.2) database".

Figures 3 and 4, an indication of which sites are being represented is missing and should be added. Table 1 appears a bit misleading as results for HFC-32 at one site (as in Figure) are not available for all time after the measurements began.

These figures indicate semi-hemispheric average mole fractions. We have added the following lines to the figure captions: "Semi-hemispheric monthly average HFC-X mole fractions (30N-90N: blue, 0N-30N: green, 30S-0S: purple, 90S-30S: red). Averaged observations are shown as data points with error bars."

Velders et al., 2012 is not listed in reference section.

This was a mistake, we have now added:

Velders, Guus J M, A R Ravishankara, Melanie K Miller, Mario J Molina, Joseph Alcamo, John S Daniel, David W Fahey, Stephen A Montzka, and Stefan Reimann. 2012. "Climate Change. Preserving Montreal Protocol Climate Benefits by Limiting HFCs." Science (New York, N.Y.) 335 (6071) (February 24): 922–3. doi:10.1126/science.1216414. http://www.ncbi.nlm.nih.gov/pubmed/22362993.

Consider changing color scheme in ïňĄgures 1 and 2, the points dominate the colors of data presented in the ïňĄgure, but not the colors displayed in the caption.

We have given this comment close consideration and have decided to make no changes to the Figures 1 and 2. We feel that it is difficult to represent so many different stations data on one plot, but the colour scheme used is quite clear for each site with or without the points.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 6471, 2014.

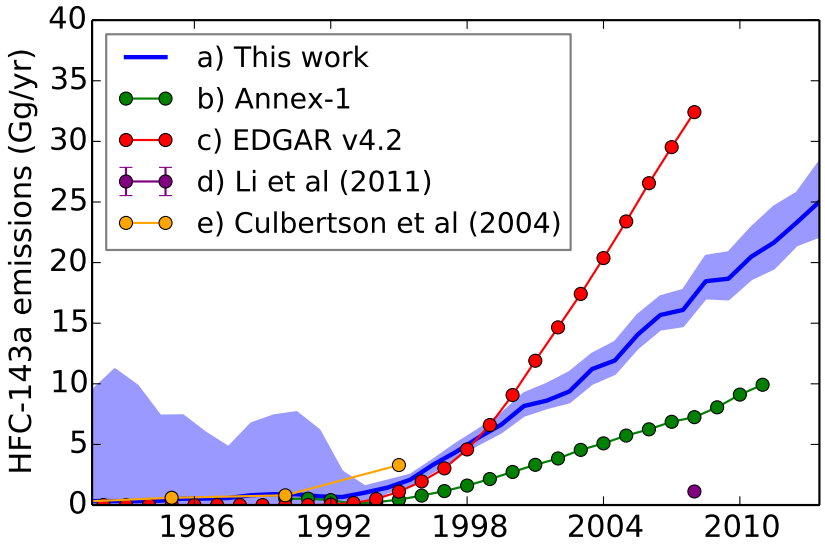

Fig. 1. Revised Figure 5