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Interactive comment on “Global emissions of HFC-143a (CH₃CF₃) and HFC-32 (CH₂F₂) from in situ and air archive atmospheric observations” by S. O’Doherty et al.

Anonymous Referee #1

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The paper discusses some high quality measurements made over an extended period from archived samples and in-situ instrumentation. The study is a useful and timely addition to our understanding of HFC global concentrations and how they are changing in the background atmosphere over time. The paper also supplies global emission rates derived from the observations with the use of an inversion model. The paper includes an interesting comparison to inventory-based emissions estimates and discusses implications of the apparent differences. The graphics are quite clear, although some additional information is needed. Also, some unlikely conclusions are drawn from the change in the ratio of mole fractions measured at a northern hemispheric site relative to

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a site in the southern hemisphere. Though this is a minor part of the paper, this section needs to be carefully reconsidered, along with the discussion of Australian emissions and Table 2, which don't add substantially to the conclusions provided by this study, in my opinion. Once these issues and the other details described below are rectified, I imagine that this paper would be acceptable for publication.

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 influenced by the inter-hemispheric distribution of emissions, it is also influenced 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 influence 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.

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.

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 influenced by

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the prior emission magnitudes and their uncertainty?

Details:

p. 6474 HFC-23 has a lifetime of »50yr. 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. L 23, projected for what year? Is this sentence and the following one a discussion of the past or future?

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

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

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)

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 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 specification. 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.

On emission estimates, it would be useful to know which of the processes listed as being considered in deriving the uncertainty in Section 3 are the main contributors to

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the derived uncertainty. 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.

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.

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

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

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

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