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Interactive Comment

Interactive comment on "Global stratospheric fluorine inventories for 2004–2009 from Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) measurements" *by* A. T. Brown et al.

Anonymous Referee #2

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In Brown et al., the authors derive an accounting of atmospheric changes in total F over a 6 year period. They accomplish this task by combining results from ACE retrievals for the majority of F-containing gases with those from a model calculation constrained by independent surface observations for the remaining F-containing gases. I interpret the goals of the work to be to 1) provide a measure of time variations in total atmospheric F as a function of latitude and altitude, 2) quantify the different contributors to total F, 3) determine if the amount and change in total F measured in inorganic gases is well accounted for by the observed and modeled emitted components (mostly organic F

+ SF6), and 4) provide some indication of how these results affect present or future climate.

Unfortunately, I find the paper to be unclear and imprecisely written, lacking appropriate citations, and missing many necessary details. The paper also contains a significant error related to the use of GWP. GWP is a metric that quantifies the future integrated climate influence that an emission of one gas has relative to an emission of CO2. As a result it is entirely inappropriate to weight mixing ratios by GWPs as they do, and they will need to thoroughly reconsider many sections of the paper related to this. To derive quantities relevant for climate forcing provided by a trace gas at a given mixing ratio, weight that mixing ratio by a radiative efficiency; to derive quantities related to the relative climate influence of emissions of different gases integrated over time, weight emissions by GWP.

On lacking clarity and precision, some examples: the paper is supposedly about construction and calculating a F "budget", but in my mind this causes confusion. The paper is not about sources and sinks but instead is an accounting of the F observed in the atmosphere. p. 16886, a vertical gradient in total F doesn't necessarily indicate increasing emissions, it only means that sources are larger than sinks at the present time. . . for long-lived gases it can take quite some time to reach steady state. Nowhere is the latitude range meant by "tropics" defined as used by the authors. P. 16897, is it necessary to cause potential confusion by using terms (organic and inorganic) in an unconventional manner? The Montreal Protocol actually doesn't ban any chemical; use for certain purposes was controlled and prohibited. P. 16897, why "loosely interpreted"? The last sentence of the conclusion section has no verb.

There is very little mention of extensive ground-based measurements of these organic Cl- and F-containing gases. One of the purposes of Section 2 seems to be to inform the reader about the accuracy or comparability of these retrievals to other methods; I'd argue that it fails to do this well without mention of ground-based results. Total F has also been derived from surface observations, yet no mention of those results is

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made in the discussion of total F trends (section 6.4). Citations are lacking in the text for lifetimes, GWPs (the latest of which should be retrieved from the latest WMO report cited as recommended in that report).

Uncited assertions are also made early in section 5. These seem to be unrelated points that might best be removed. It seems quite unusual to remove outliers at some arbitrary level (at MAD>2.5) and then calculate a standard deviation with the remaining data and propose that it can be called an error that then become combined in a linear fashion (p. 16898, not in quadrature...)?

I'm surprised that anyone ever thought that F could be used as a proxy for understanding atmospheric Cl (introduction). It seems to me more accurate to suggest that measures of atmospheric F provide an independent means to assess our understanding of the atmospheric chemistry (and transport) of ozone-depleting substances containing both of these halogens.

I found interesting the expectation of a correlation coefficient near 1.0 in F from reservoir species compared to source gases (section 6.2). It seems that the timeframe represented by reservoir and source gases are not the same, such that one would not expect a correlation coefficient of 1.0. It would be interesting to understand how much this time-lag influences the expected value of the coefficient, and whether or not it has a spatial dependence that can be detected in your data given that I'd expect the largest time difference (and total F difference) in the high-lat NH compared to other regions. In this section it is not clear how the results for reservoir and source gases are being aggregated, this needs indicating. Tables and figure captions are also missing details: Table 5, are these sums of F from source gases and reservoir species? Table 7, are these column averages? Table A1-A4, are these an average for all years? State clearly in the caption of Figures 1 and 2 how the error bars were derived so that the figure stands alone.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 16885, 2013.