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Atmospheric Chemistry and Physics Discussions

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.

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Global stratospheric fluorine inventory for 2004 - 2009 from Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) measurements and SLIMCAT model simulations

Response to Reviewers' Comments

We thank the reviewers for their comments. These comments are repeated below,

followed by our response.

1. "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."

The RE VMR-weighted total fluorine has now been calculated and the changes in this value between 2004 and 2009 have been evaluated. However, this weighting does not take the lifetime of the species into account. The lifetimes of CFCs and their replacement HCFCs and HFCs do not have the same lifetimes. The long-term climatological impact of replacing CFCs must include a consideration of the lifetimes of these species. The GWP of a species is dependent on the lifetime of the species. We therefore include both the RE and GWP- weighted trends. It is made clear that GWP-weighted values present the climatological influence of the emission of a hypothetical packet of gas with the relevant burden of fluorine containing species over a 20-year timeframe.

In the abstract:

"The calculation of radiative efficiency-weighted total fluorine allows the changes in radiative effect between 2004 and 2009 to be calculated. These results show an increase in radiative effect of between 0.23 $\pm$ 0.11 % per year and 0.45 $\pm$ 0.11 % per year due to the increase in fluorine containing species during this time. To account for the climate effect of these species with respect to their atmospheric lifetime, the atmospheric mass of each species used in this study was weighted by its global warming potential (GWP) to produce a GWP-weighted total fluorine trend. These GWP-weighted values present the climate influence of the emission of a hypothetical packet of gas with the relevant

burdens of fluorine-containing species over a 20-year timeframe. These trends show mean changes of 0.02 $\pm$ 0.08% per year in the NH, and 0.07 $\pm$ 0.05 % per year in the SH. Overall GWP-weighted fluorine remains roughly constant globally over a 20-year period."

In the Introduction: "In addition to this, the radiative efficiency-weighted trend in total fluorine is calculated showing the radiative effect of the changing VMR of fluorine between 2004 and 2009.The GWP-weighted trends in total fluorine are also presented and illustrate how changes in atmospheric fluorine have affected climate forcing over a 20-year time period."

In the Results and Discussion: "This is done in two ways. The immediate radiative effect of the total fluorine was evaluated by weighting the total fluorine VMR by the radiative efficiency of each species. The mean total fluorine mass was weighted by the 20-year GWP to evaluate the climate forcing impact of total fluorine, due to the varying atmospheric lifetimes of the fluorine-containing species, over a 20-year time period."

Section 6.5: "The errors on all of these GWP-weighted trends are larger than the reported GWP-weighted trends; these results suggest that it is likely that the GWP-weighted fluorine will remain roughly constant over a 20 year time period."

An entirely new section has been written on the changes in the radiative effect of fluorine (radiative efficiency weighted fluorine) during this time:

"The main environmental impact of the increased atmospheric fluorine will be on warming in the troposphere and lower stratosphere. In order to evaluate this effect the total fluorine was weighted using the radiative efficiencies (Solomon et al., 2007) of the individual species to calculate the change in radiative effect due to changes in fluorine VMR during this time. These changes in radiative effect for each species were calculated by removing VMR above the tropopause and multiplying the VMR (in ppb) of each species by its radiative efficiency. These values were summed and the mean was calculated for each year. A least squares fit to the means then allowed the changes

in radiative effect to be calculated. The results of this analysis can be seen in Table 8 and Figure 5. Both hemispheres exhibit a similar increase in radiative effect due to fluorine species. In the northern hemisphere the rates are 0.23 $\pm$ 0.11 % per year in the extra-tropics and 0.45 $\pm$ 0.11 % per year in the tropics. Similarly in the southern hemisphere radiative effect due to fluorine-containing species appeared to increase by 0.29 $\pm$ 0.20 % per year in the tropics and 0.45 $\pm$ 0.09 % per year in the extra-tropics. Once more the same calculations were carried out without including CFCs and halons. These results are shown for comparison in Table 8. Without CFCs and halons the increasing radiative effect of fluorine containing species would be significantly higher ranging between 3.84 $\pm$ 0.02 % per year in the extra-tropical southern hemisphere and 4.45 $\pm$ 0.02 % per year in the extra-tropical northern hemisphere. Thus at the present time the climate impact of fluorine is increasing."

In addition to this there is a new figure and table included.

2. "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."

The gradients give an indication to the changes in the VMR of fluorine-containing species. It is now made clear in the paper that the gradients represent the changes in VMR of total fluorine rather than changes in the emissions of fluorine containing species.

3. "Nowhere is the latitude range meant by "tropics" defined as used by the authors."

This has been clarified in the paper in the following way:

"For this analysis the globe was divided into 4 latitude bands (70° N to 30° N – Northern hemisphere extra-tropics, 30° N to 0° N – Northern hemisphere tropics, 0° N to 30° S – Southern hemisphere tropics - and 30° S to 70° S – Southern hemisphere extra tropics)."

4. “P. 16897, is it necessary to cause potential confusion by using terms (organic and inorganic) in an unconventional manner?”

As mentioned in the text, this notation was chosen as it follows the convention used in previous fluorine budgets.

5. P. 16897, why "loosely interpreted"?

The term loosely has been removed.

6. “The last sentence of the conclusion section has no verb.”

This has been corrected.

7. 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).

These references have been added c.f Table 1.

8. “Total F has also been derived from surface observations, yet no mention of those results is made in the discussion of total F trends (section 6.4).”

Total F from ground based FTIR measurements are mentioned in the introduction, in addition to this Kohlhepp et al. (ACP, 12, 2012) is also referenced.

9. “Uncited assertions are also made early in section 5. These seem to be unrelated points that might best be removed.”

This has been done.

10. “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?”

MAD filtering is simply carried out to remove outlying data points. The vast majority of the data points are retained and for the most part the median of the data is not significantly changed. Once the outliers are removed the mean and the standard deviation

can be calculated. 2.5 MAD has been chosen as it removes the majority of the outliers without removing significant amounts of “non-outlying” data.

11. “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.”

This point has been clarified in the text:

“Fluorine budgets are useful metrics for checking the atmospheric chemistry of fluorine-containing species. Differences between model and measured values would suggest that there are additional species contributing to the total fluorine budget which have not been considered or that our understanding of the chemistry and transport of these species was lacking.”

12. 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.

The method used to calculate the correlation has been better explained in the text:

“Correlations were calculated by summing the relevant species (see above) to produce source and reservoir species profiles. This produced a profile with a value for the VMR of reservoir and source species every 1 km between the tropopause and 53.5 km. These VMRs were plotted against one another for each altitude in order that a correlation could be calculated.” Using this method one would expect a correlation

between the source and the reservoir species to be roughly one since the air at each level of the stratosphere is by definition stratified and as such has a set age-of-air (all be it with a slight spread of ages in each air mass). Since fluorine atoms are so reactive the fluorine can only be present as either a source gas or a reservoir and thus a correlation should be present."

13. "Table 5, are these sums of F from source gases and reservoir species?"

This has been clarified in the caption

14. "Table 7, are these column averages?"

No, they are simply an unweighted mean of the stratospheric total fluorine. An explanation of the error on the values is now given in this caption.

15. "Table A1-A4, are these an average for all years?"

The captions have been changed to clarify this e.g:

"The percentage contribution of each species to the total fluorine budget in the latitude bands between 70° N and 30° N calculated from mean total fluorine profile for all years".

16. "State clearly in the caption of Figures 1 and 2 how the error bars were derived so that the figure stands alone."

"Figure 1: The fluorine budgets from 2004 to 2009 in latitude band 30oN-70oN and 0o-30oN (black). The inorganic (blue) and organic (red) fluorine profiles are also shown. The error bars are the result of combinations of standard deviations of the mean profiles of the relevant fluorine containing species calculated from the filtered data for each species. SLIMCAT VMRs were given a 5% error which overestimated the error in the ground based VMR and uncertainties in the transport."

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