

Black: referee's comments red: authors' answers

First, we want to thank the referee for the detailed analysis of our paper.

The paper by Zhou et al is the first study of the long term trends in the total column of the major atmospheric CFC's 11 and 12, and HCFC 22 from the southern hemisphere. The datasets are from Reunion Island's two NDACC stations using their high resolution FTIR spectrometers. The analysis technique uses the well-established SFIT4 software to give total columns abundances of all three species. The data is compared with collocated satellite measurements which compare quite well and agree within the error budget of the analyses. The trends are derived using a regression model with robust statistical uncertainties. These trends are compared with in-situ data which show qualitative agreement in the overall well established changes in the concentrations of these gases.

The paper is therefore technically sound using well established analysis procedures and with a team of researchers that are experts in this field. The paper is well written in general but there are issues with formatting of equations and a few minor typos (outlined below). There is also the need to explain further some areas of the text that are a bit confusing and unclear, but again the particular sections are mentioned below. The paper is suitable for publication in AMT subject to the authors satisfactorily addressing the questions/comments listed below.

Comments/Questions: 1. Page 2, line 74. While it is true that this is the first study of all three species, the authors have missed the only other study from the southern hemisphere of HCFC 22 total columns, namely that of Sherlock et al (1997). This mentioned only in the context that southern hemisphere measurements are so few and far between, which these authors will appreciate.

Indeed, we missed this very relevant study. Thanks to the referee for pointing that. We added this work in the introduction part, as well as the Zander et al. (1994) reference.

We also found the retrieval microwindow ($868.7-831.0\text{ cm}^{-1}$) in Zander et al. (1994) is larger than the one used in our study ($868.75-829.4\text{ cm}^{-1}$; from Jungfraujoch). As the background of this window has a large slope (see Figure 1 in the paper), we prefer to choose a larger microwindow to obtain a better fit of the background. Several microwindows have been tested, and $828.6-831.0\text{ cm}^{-1}$ is selected at the end, because it provides us more data with less scatter. What's more, the seasonal cycle of HCFC-22 from this microwindow is more close to the MIPAS result. Therefore, we updated the FTIR HCFC-22 retrievals both at St Denis and Maito in the revised paper.

2. Page 3, lines 109-112. This sentence is poorly constructed, rephrase.

The sentence is rephrased.

3. Page 3, lines 121-125. The history of the SFIT2/SFIT4 codes is not quite correct here. SFIT2 was certainly developed by the Institutions mentioned, but SFIT4 was largely developed by lead contributors from the University of Bremen and NCAR, with other contributions from several other Institutions within NDACC.

Thanks for your suggestion. We changed the sentence as: SFIT4, largely developed by lead contributors from the University of Bremen and National Center for Atmospheric Research (NCAR), with other contributions from several other Institutions within NDACC.

4. Page 4: general comment here, the formatting of the equations needs improving. Line numbers need to be right adjusted without an ending dot or comma.

All equations have been improved.

5. Page 4 line 144, remove extra 'a'

Corrected.

6. Page 4-5, lines 145-179. This is an interesting discussion on the use of channel spectrum fitting in SFIT4. There is little discussion in the literature on the use of this option in high resolution IR spectra so it is very useful here. However the authors discuss this without first explaining why it is necessary in the first place. Clearly this is an issue in some spectra for long wavelength channels. So first establish the problem (in a sentence or two), and then follow with the explanation of how this is treated in the analysis. We added the following sentences after equation (2): “Some low frequency oscillations of the baseline can occur in the spectra, resulting from the mirrors, filters or apertures. While this is not a problem for small retrieval windows (a slope and a curvature are sufficient to fit the baseline), it could be necessary in the case of wide window to include a so-called “beam correction” to fit these oscillations. In SFIT4, this is done by adding a zshift-like parameter z_b for the interferogram perturbation (IP) model”.

In line 161, the maximum number of beams is purely a software choice which can be changed to anything the user wishes.

Corrected.

7. Page 5, line 165, “are narrow, a linear fit is”

Corrected.

8. Page 5 line 170, here is the first mention that channel spectra are necessary to fit. Explain what an IP-type beam is if you think it necessary otherwise leave it out as this implies other choices and will only make sense to established SFIT4 users.

Since IP-type beam has been introduced in equation (3)-(5), we prefer to leave it out here but we added the reference to Eqs. 3-5 in the text.

9. Page 5, line 177, “As such an oscillation”

Corrected.

10. Page 5, line 192/193, So what you are saying is that for each specific gas and site, there is one respective a priori profile, yes?

Yes, we add this sentence in the text to make it more clear to readers. For each specific gas and site, there is one respective a priori profile.

11. Page 5, line 197, “values of the covariance matrix”

Corrected.

12. Page 5, line 199/200, this seems a little confusing contrasting the WACCM variability for the covariance (understood) but with a priori profile ensemble. It is not clear what the a priori ensemble is?

We want to express that the same dataset, 2004-2016 monthly data from WACCM, is applied to construct the a priori profiles and a priori covariance matrix. We changed the sentence to make it clear to readers like: “In our study, 2004-2016 monthly data from WACCM is used to provide the variability for the FTIR retrieval, which is the same dataset used for creating the a priori profiles.”.

13. Page 6, line 209, sensible => sensitive

Corrected.

14. Page 6, line 210-213. With such a relatively poor dofs, what advantage does the profile retrieval bring compared with simple scaling? That is, is there some tangible benefit (does it improve the error budget and other fitting statistics)?

Indeed, the profile retrieval shows a better fitting, but not a lot, compare with the column retrieval. The advantage of the profile retrieval is to give us more information on the profile retrieval error and more knowledge about the vertical sensitivities (total column averaging kernel).

15. Page 6, section 2.2.2, improve the editing of equations, equation numbering etc. Fix issues with symbols (KbEb), and the symbol on line 224.

Corrected.

16. Page 6 line 228, what specifically is ignored? The iteration error?

In this equation, we assume that $F(x_t, b) - F(\hat{x}, b) = \hat{K}(x_t - \hat{x})$, so the iteration error (2-order items) is ignored. We added this clarification.

17. Page 6, line 232/233, specifically put in each term symbol, that is , “ ..forward model error E_f , the.”

Corrected.

18. page 6, line 246, is there a “not” missing, that is, it should read “but not in the ”?

Corrected, not is added in the sentence.

19. page 7, line 257, insert comma after Mäido.

Corrected.

20. Page 7, line 261, it might be useful to put CFC-11 and HCFC-22 in brackets after the wavelength ranges for clarity.

Added in the sentence.

21. Page 7, line 264, can you say that the offset is actually quantitatively consistent with the pressure difference?

Figure 4 has been updated by adding the a priori total columns at St Denis and Mäido (green dash lines). The offset of the a priori total columns between St Denis and Mäido is mainly caused by the different altitudes (or pressures), which is consistent with the offset of the retrieved total columns between St Denis and Mäido. Therefore, we think this offset is quantitatively consistent with the pressure difference.

22. Page 7, lines 275-279. These sentences need some editing; “Our FTIR measurements capture the main but the scatter in the uncertainties of the FTIR measurements. The FTIR columns are also associated with the scatter in the air column”

Corrected.

23. Page 8, line 300, Jungfraujoch

Corrected.

24. Page 8, line 303, the end of this sentence makes it a bit unclear. If you do not perform analysis on the Mäido data, then remove the “only”. What you are trying to say here is that due to its short time span you only perform the trend analysis on the St Denis data.

This not exactly what we meant: we are not performing only trend analysis at St Denis, we also use Mäido but in combination with the St Denis partial columns. To clarify, we changed the text as follows: “Since the time range of Mäido measurements only covers about 3 years, we cannot perform trend analysis on Mäido data only. Therefore, we use the total columns at Mäido in combination with the St Denis partial columns calculated at the altitude of Mäido (2.155-100 km) to derive the trends of CFC-11, CFC-12 and HCFC-22 for the period 2004-2016”.

25. Page 8, lines 328, 329. This is an interesting dynamic in the data, the apparent cycle in the CFC-12. One might be tempted to point the finger at water but both CFC-11 and HCFC-22 have worse water interference. One would think that any dynamical effect would impact on CFC-11 as well while any chemistry is unlikely here. Do the authors have any ideas?

It is also not clear for us in this study: we found no obvious correlation between CFC-11 and water vapor interference. Figure 4 (updated in the revised manuscript) shows that the a priori total columns also has a seasonal cycle for CFC-12, which is caused by the air mass changes. However the amplitude of the variation for a priori total column is smaller than that for retrieved total column. We will look deeper into these seasonal variations when more data will be available. It would also help a lot to use a atmospheric chemistry model to figure out what is leading to such seasonal variation, but this was beyond the scope of this AMT publication. However, in the revised version, we added the seasonal cycle from MIPAS data: it shows similar seasonal cycle which gives confidence that this is not an artefact of the retrievals. The possible reasons will be investigated in the future.

26. Page 9, formatting of equation 13

Corrected.

27. Page 9, the MIPAS DOFS is clearly larger, and is therefore the one that is smoothed, but for completeness, can the DOFS for MIPAS be mentioned to show why this is so?

Added in the text.

28. Page 10, line 387, add "respectively" at the end of this sentence.

Corrected.

29. Page 11, line 418, remove extra comma.

Corrected.

30. Page 11, line 424, add "respectively" after HCFC-22

Corrected.

31. Page 18, table 4 caption, line 603, is there an extra % there?

Corrected.

32. Page 20, figure 1, CFC-11 graph legend, the colour scheme for each of the different gases has not worked, and in the caption, line 618, replace "with" => "by"

Corrected, and the figure has been improved to clarify the absorption lines of each species.

33. Page 24, figure 5, is there enough FTIR data to show monthly means rather than individual data? Since the profiles are uniformly mixed in the trop, how far off would a simple calculation of the mean mole fraction be, based on a partial column divided by an partial air column from the pressure (for qualitative not quantitative comparison)?

Thanks for your suggestions. We think it is better to keep the individual data since several gaps exist in the time series of the FTIR measurements. The quantitative comparison is shown below (In-situ vs Xgas), and we replaced in the manuscript (Fig.5) the FTIR columns in molec/cm2 by Xgas in ppt. A short discussion is also added in the text.

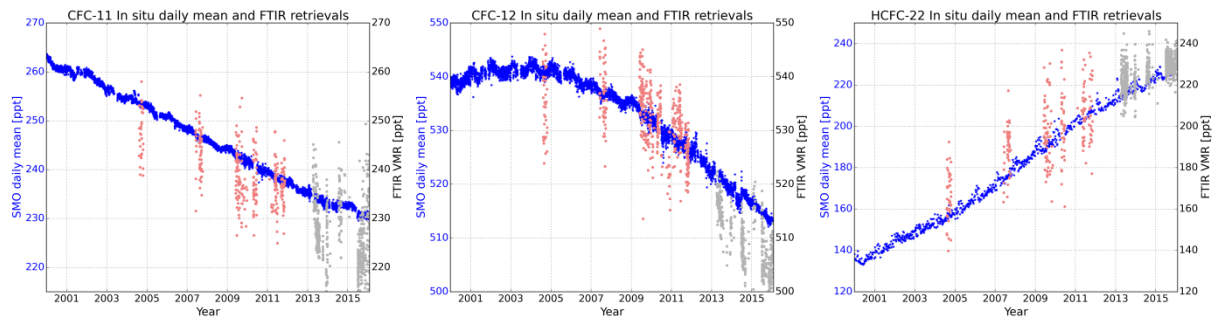


Figure 5. In-situ daily mean (CFC-11 and CFC-12) and flask pair measurements (HCFC-22) at SMO site (blue) and individual FTIR column averaged dry-air mole fractions at St Denis (light coral) and Maïdo (grey). Left: CFC-11; middle: CFC-12; right: HCFC-22.