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Atmospheric Measurement Techniques
Discussions
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Interactive comment on "CFC-11, CFC-12 and HCFC-22 ground-based remote sensing FTIR measurements at Reunion Island and comparisons with MIPAS/ENVISAT data" *by* Minqiang Zhou et al.

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General Comments

This paper presents for the first time the time series of three important gases on the atmospheric chemistry, CFC-11, CFC-12 and HCFC-22, at two stations in Reunion Island. The strategic location of the ground-based station and the methodology proposed, using high-resolution Fourier Transform infrared (FTIR) solar absorption spectra and the well-established retrieval strategies, provide high confidence to the results obtained. This is also documented with a comprehensive comparison with other mea-

surement techniques and previous studies. The paper is well-written and concise. However, the presentation of the results and their discussion is a bit confusing and it is not well structured. For example, the authors first carry out a trend analysis comparing with other techniques, such as those from MIPAS sensor, and in the following section, a validation with coincident MIPAS is presented on a measurement-to-measurement basis, but the corresponding trends are not compared to coincident MIPAS database. Moreover, the overall treatment of the theoretical errors is imprecise, so the authors should improve its description, and discussion. Detailed explanations of the sections to be corrected are mentioned in the Specific Comments. I suggest this paper may be suitable for publication after addressing these issues and the specific ones listed below.

Specific Comments

Section 2.2.

Line 129: One of the key issues addressed by the paper is the presentation of the linear trends for CFC-11, CFC-12 and HCFC-22 as observed by ground-based FTIRs. To do so, it is very important to document/show that the spectrometers do not show any temporal drift that could affect the trend estimation. This could be done and discussed, for example, by including a Figure with the time series of Instrumental Line Shape (ILS) in Section 2.2 or of the retrieved DOFs in section 2.2.1 (line 204) or of the measurement noise.

Section 2.2.1.

Line 192: The retrieved HCFC-22 vertical profiles at St. Denis show a stronger variability in the troposphere/lower stratosphere than in Maido (see Figure 2). It would be nice if the authors include some explanation for this observed variability? Line 196: As it is stated by the authors, the a priori covariance matrix is crucial in the optimal estimation method as well as to estimate the FTIR smoothing error. Thereby, authors need to justify the use of WACCM model outputs to estimate the covariance matrices for the considered trace gases. It would be recommendable to state how well the WACCM simulations represent the real atmospheric states or provide references on works that show ability of the WACCM model to reasonably simulate vertical distribution of the target gases. Line 204: Are the FTIR retrievals filtered according to some specific quality filter or SNR? Please, clarify, because in Section 2.2.3 the authors mention that "although the retrievals use the same database of spectra, the numbers of successful retrievals for CFC-11, CFC-12 and HCFC-22 are different".

Section 2.2.2.

The basis for the theoretical error estimation is presented well, but the discussion of the error budget is a bit confusing and imprecise, and it must be clarified and completed. A detailed error discussion would be very useful because it indicates where a special effort should be paid to improve the precision of the CFC-11, CFC-12 and HCFC-22 FTIR products in the future. 1) St. Denis and Maido are two humid sites. To avoid the H2O interference, the authors follow the recommendable retrieval strategy of performing a pre-fit of H2O previous to the target gas retrievals. Nevertheless, the effect of humidity (cross-interference of H2O) should be included in the error estimation to analyse its remaining impact and support the two-steps strategy. 2) Other important error source that is not included in the error budget is the Instrumental Line Shape (ILS). According to Figure 1 (averaging kernels), the retrieved FTIR profiles of the target gases still show sensitivity in the lower/middle stratosphere, where the ILS' impact is not negligible. Please, consider including it into the error estimation. 3) Some details of the error estimation calculation are missed or imprecise. For example: a. How the error covariance Sb is exactly calculated (diagonal, inter-layer correlation)? b. How is the error of the interfering species estimated? c. For temperature, the Sb matrix is computed from the difference between NCEP database and balloon observations, but please state the assumed error values for reference. d. For the smoothing error, how are the systematic and random contributions computed? e. In the text the authors mention that for the retrieval parameters the systematic contribution is set to zero, but

in the Table 2 the authors assume that the zshift parameter has the same systematic and random uncertainty and the corresponding results are shown. Please clarify. Also, just to homogenise the format, considering including the systematic and random error values in Table 2 in the same way. For example, for SZA the systematic and random errors are given (I guess because they are different), but not for zshift. Please include even if they are equal. f. I guess that the error budget presented in Table 2 and 3 corresponds to the whole FTIR spectra database. Please state. g. Please specify the references for the assumed uncertainty, for example, for the spectroscopic parameters, the solar zenith angle, etc.

Section 2.2.3, 2.2.4 and 3.

The presentation and discussion of the results are mixed through the three subsections. For example, the intercomparison with MIPAS database is a validation study that experimentally quantifies the uncertainties of the new FTIR products. But, this is shown as the last section of the paper, when it should be the first part of the results section. Also, only a measurement-to-measurement comparison is carried out, without neither comparing annual cycles nor linear trends, when this analysis is presented and discussed in the previous section with other database (among them MIPAS observations from literature) for linear trend, but no references are given for seasonal patterns. Specific comments are listed below:

1) Consider re-organising these three sections by merging them into a unique one. This section could include a subsection describing the "comparison/validation" dataset (MIPAS and SMO), and three subsections for the comparison at the different time scales: measurement-to measurement, annual cycles and linear trends versus coincident MIPAS database (I would also suggest to include a more detailed comparison with SMO data). This implies that authors should carry out the comparison between the MIPAS and FTIR annual cycles and linear trends, not included until now in the paper. Regarding to SMO data the authors have different options to perform this comparison (total column-averaged dry air mole fractions of the different target gases or simply averaged

tropospheric VMR values) and there are multiple references in literature. In the trend analysis subsection, the authors could include the comparison with results from literature (FTIR at Junfraujoch, ACE-FTS and MIPAS) as already shown in section 2.2.4. 2) Unify the FTIR products. The paper presents and discusses four different products for the target gases: total column, partial columns between 2.155-100 km, vertical profiles and partial columns between 6 and 30 km, when the FTIR is only sensitive to one wide layer (typical DOFS about 1 for CFC-11 and HCFC-22, and 1.5 for CFC-12). So, the products will show systematic differences due to the different integration ranges, but the variability observed by them will be the same or very similar. 3) Section 2.2.3. This section only addresses the presentation of the total column time series and no significant results are discussed. Thereby, consider removing it and moving part of the text to other sections. For example, the explanation of the data density could be shown in the section 2.2 (FTIR retrieval), because a quality filter is applied in the spectra or retrievals (I guess the SNR is set in the SFIT retrievals). The authors suggest that this could be due to the fact that the microwindows used for the CFC-11 and HCFC-22 estimation lie in the edge of the spectrum with lower SNR. Have the authors checked if the number of successful retrievals improve for example by reducing the broad microwindows used for CFC-11 (only taking the part with higher SNR) or have they investigated other lines? 4) Section 2.2.4: a. Please consider including a more detailed comparison with SMO in-situ and flask data. b. Line 312: For CFC-12 a significant trend change is detected in 2004. Have the authors some idea of reasons of this change? It is natural or an instrumental artefact? According to the Figure 5, this trend change is also observed in the in-situ data, thereby it is likely due to natural variability of CFC-12. To take into account non-linear terms in the trend analysis, a Fourier term could be included in equation 10 to account for inter-annual trend variation (for example, refer to Gomez-Pelaez et al., 2006 and 2010). So, the trend analysis could be done with the whole time series. c. Line 322: Have the annual cycles for CFC-12 evaluated considering the whole FTIR time series because the linear trend is only estimated from 2009 onwards? Please state. d. Line 330: A significant seasonal variation is only observed for CFC-12, which

has a typical DOFS value of 1.5. Thereby, its annual cycle could be capturing the tropopause shift or part of the lower stratosphere information? As mentioned before, it would be interesting to include the annual cycles observed by MIPAS and SMO data to improve the discussion. Also, it would be nice to compare the seasonal patterns of the target gases with some references from literature. 5) Section 3: a. Line 348: Adding a brief explanation of the MIPAS products (for example, vertical resolution and sensitivity) and the expected uncertainty values will be useful for the comparison section. b. Line 365: As mentioned before, it will be interesting to have information about the vertical sensitivity of the MIPAS profiles, since smoothing the MIPAS profiles does not much sense if FITR and MIPAS vertical sensitivity are comparable and even it could produce unrealistic results. c. Line 397: Please state if the comparison of the monthly means is for the coincident MIPAS and FTIR database or considering the respective original time series (I guess it is the last option).

Technical Comments

Line 91: State the specific goals of the NDACC network similar to TCCON network. Line 173: Figure 2 and Figure 3 are in wrong order. Line 239: Replace “resp.” by respectively. Line 245: Replace “uncertaintyis” by “uncertainty is”. Line 246: Do the authors mean “but not in the model parameters uncertainties for CFC-12”? Line 257: Remove the full stop before “together”. Line 264: Replace “,” by “.”. Line 275: Include reference for the in-situ and flask uncertainty. Line 287: Correct the reference of Santer et al., (2000). Line 300, 305, 310: Although the period at which the trends are evaluated are included in Table 4, please include in the text for a better comparison for FTIR measurements at Junfraujoch, and SMO. Table 2 Line 589: Consider moving the sentence “When a relative. . .” to the end of the paragraph. Line 591: Replace “zshiftis” by “zshift is”. Figure 1: To homogenise the figure formats, please, set the limit of the abscissa axis for CFC-11 to -0.02.

References Gomez-Pelaez, A.J., Ramos, R., Perez-delaPuerta, J., "Methane and carbon dioxide continuous measurements at Izana GAW station (Spain)" in GAW Report (No. 168) of the "13th WMO/IAEA Meeting of Experts on Carbon Dioxide Concentration and Related Tracers Measurement Techniques (Boulder, Colorado, USA, 19-22 September 2005)", edited by J.B. Miller, World Meteorological Organization (TD No. 1359), 180-184, 2006. http://izana.aemet.es/publications/IzanaCH4CO2_ReportGaw168_13th_meeting_expert_CC

Gomez-Pelaez, A.J., R. Ramos, E. Cuevas, V. Gomez-Trueba, 25 years of continuous CO2 and CH4 measurements at Izaña Global GAW mountain station: annual cycles and interannual trends; Proceedings of the “Symposium on Atmospheric Chemistry and Physics at Mountain Sites (ACP Symposium 2010, June 8-10, 2010, Interlaken, Switzerland)”, 157-159, 2010 http://izana.aemet.es/publications/Abstract_25-year_CO2_and_CH4_Izana.pdf

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