

Final author comments on “Spectrometric monitoring of atmospheric carbon tetrafluoride (CF₄) above the Jungfraujoch station since 1989: evidence of continued increase but at a slowing rate”, Atmos. Meas. Tech. Discuss., 6, 7535-7563, 2013.

We are very grateful to the referees for their useful constructive comments and numerous suggestions for improvement of our manuscript. We provide hereafter a detailed point-by-point answer to the various questions raised.

Interactive comments by Referee #1 (see RC [C2381](#))

1. Pg 7538, line 16: “(e.g. Zander et al., 1992,...)” As “e.g.” means “for example,” a comma should be placed after the “e.g.” so it reads “(e.g., Zander et al., 1992, ...)”.	Commas have been added where appropriate.
2. Line 27: Same as above. Should be “(e.g., Rinsland...”	Idem
3. Pg 7539, line 4: Please provide a reference regarding emission factors for Chinese smelters.	A reference to the International Aluminum Institute 2012 survey and the corresponding citation have been added
4. Pg 7541, line 18: Suggest changing “badly” to “strongly.”	“badly” has been replaced by “heavily” (strong is already used twice in the same sentence)
5. Pg 7541, line 15 – Pg 7542, line 9: I don’t think this long explication of why they used a “split” bandpass is necessary, and should be considerably shorter. I suggest just writing that using the entire bandpass from 1283.73 to 1285.15 was impractical because of H ₂ O and HDO interference, so it was split into two windows and reference Figure 1. The interference is obvious from the residuals, and the long discussion of Run 1 vs Run 2 is not needed.	This section has been shortened and reworded; some elements have been moved to the caption of Figure 1
6. Pg 7542, line 18-20. The line “As a result of the poor information content...” could be more simply stated as “As there is poor vertical discrimination in the spectra, the constant a priori profile was scaled for fitting.”	This sentence has been changed to “As there is poor vertical information content in the individual spectra, the constant a priori profile was simply scaled during our fitting procedure.”
7. Pg 7542, line 27. Suggest replacing “adopted within the frame” by “used in.”	Done
8. Pg 7546, line 7-10: Suggest a graph or table instead of writing in all those numbers.	The six-year time averaged trends and corresponding global emissions are now listed in a table (as Table 2).
9. Pg 7546, line 23: Suggest replacing “combined with the assumption” with “assuming.”	Done.
10. Pg 7546, line 23 – Pg 7548, line 15: This is a long, wordy comparison of this work with	We believe that the comparative data sets need to be introduced in the text

previous measurements. I think that some elements of the comparison might be better put into a table, and other elements into the caption of Figure 3.	and the comparisons discussed. Distribute the various elements between an additional table and the caption of Fig.3 does not seem to be the most appropriate way to us and we would prefer keeping the text as is.
11. Pg 7548, line 19: "21st millennium" should be "21st century."	Done
12. Pg 7550, line 3 "...one should keep in mind that..." The meaning of this phrase may not be obvious to some that are not fluent in English. Suggest replacing this with "note that".	The beginning of this sentence has been changed. It now reads as "Nonetheless, it remains that..."
13. Pg 7550, lines 9-13. "Since the 1980's...regulation." This has already been covered in the introduction, and does need to be repeated.	The summary section has been written such as to consist in a stand-alone part of the text, putting the research done into perspective. Some repetitions are therefore unavoidable.
14. Pg 7552, line 4: "it is worth mentioning that" is a cliché that better used in conversation rather than writing.	The sentence has been reworded to "Finally, we expect that the new in situ..."
15. Figure 1: For clarity, suggest colorizing the chemical formulae on the upper right as well as the lines connecting them. For example, "HNO ₃ " could be in green writing, "CO ₂ " in pink writing., etc.	A version accounting for the suggested changes has been prepared.
16. Figure 2: It's confusing to include the linear fit information. It would be more illustrative to include the fitting function and derived coefficients, and having a second graph in the Figure showing the rate of CF ₄ increase as a function of time.	Dr A. Goldman also suggested providing the various coefficients. Therefore, we have included this information in the caption of Fig. 2, for the two linear regimes as well as for the parabolic (black line) fit to the complete time series.
17. Figure 3: "It is important to note that..." is a cliché. Just write "Data sets can be..."	Done.

Interactive comments by Dr A. Goldman, referee #2 (see RC [C2384](#))

1. For the use of the formal Hitran 2004, combined with the preliminary online Hitran 2006 update as reported in the paper (correctly excluding the cf ₄ line parameters of Boudon et al.), it will be useful to verify that this is equivalent to using the formal Hitran 2008.	In order to select the linelist to combine with the CF ₄ pseudolines, we performed some tests using all available spectra from 2009. Runs performed with the formal Hitran 2008 compilation provide the less satisfactory fits to the observations, with 'Run2' spectral residuals significantly and consistently deteriorated. The retrieved columns show more scatter but no systematic bias with respect to the final data set retained here (i.e. Hitran 2004 incl. the Aug. 2006 updates and the CF ₄
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	pseudolines).
2. Furthermore, it will be useful to check the formal Hitran 2012, available since June 2013, which shows some changes in the line parameters of H ₂ O and CO ₂ in the spectral interval used (1284.73 - 1285.15 cm ⁻¹).	Using again the 2009 spectra, we performed a first evaluation of the new Hitran 2012 linelist, after combination with the CF ₄ pseudolines based on Nemtchinov and Varanasi cross-sections. In this case, we obtain very comparable fitting residuals. The CF ₄ total columns were only slightly lower, by (-0.3±0.1)%, when compared to our standard run.
3. In addition to the detailed numerical values reporting of CF ₄ column increases, it will be useful to provide the actual equations of the solid lines in Fig. 2, as well as of the two linear fits shown in boxes inside the Fig.	This was also suggested by Referee #1. We now provide the coefficients in the caption of Figure 2.
4. Technical Corrections: the text is mostly free from typos and language errors, but there are still a few that can be corrected. For example: p. 7541, parag. 3, line 1, the "consists in" can be "consists of" . p. 7541, parag. 4, line 9, need a space before the nu3. p. 7543, line 4 below the section 2.2 header, we see Miller et al. (2010), but in the references list this is given as 2008, which is the correct one.	These have been fixed.

Interactive comments by Referee #3 (see RC [C2456](#))

1. Several analysis runs are described: run 1 / run 2 / run 2 left / run 2 right. It did not become fully clear to me which recipe is the one finally selected for the analysis – I guess you use a sequential procedure using run1 in the first step (for adjusting the interfering species), followed by run 2 for adjusting CF ₄ - please clarify. It would be informative to provide the discrepancy between the run1 and run2 CF ₄ total columns (discussion on page 7542).	Indeed, we use a sequential approach: after a pre-fit of HNO ₃ in two dedicated windows, the major interferences are fitted in run 1. Thereafter, CF ₄ is further adjusted during run2. We have shortened the corresponding discussions and also removed the reference to the run2 left and right (line 9-12, pg 7542). As to the comparison between 'run1' and 'run2' total columns, we added (line 8, pg 7542): "This RMS decrease was confirmed over the entire database, while the related CF ₄ columns reduced by less than 0.7% on average."
2. In the random error budget of table 1, the H ₂ O and HDO profile slopes are listed two times?	Table entries have been edited to clarify this. The first one intends to evaluate the impact of an inappropriate slope (with respect to the actual atmospheric conditions) of the assumed vertical distributions for water vapor, the second accounts for the

<p>It would be interesting to compare the estimated random error with the empirical scatter between adjacent data points in the time series (the scatter of the data points recorded with the homemade spectrometer seems larger?).</p> <p>It would be instructive to present the partial column sensitivity of the FTIR retrieval.</p> <p>The annual cycle seen in the CF₄ total column is a bit worrying. It might be worth to investigate this anomaly further (e.g. correlation with solar elevation, water vapour column, etc).</p>	<p>impact of the modeling of the local continuum on the retrieved CF₄ columns.</p> <p>We have performed this comparison. The average relative standard deviation around daily mean total columns (for days with at least 4 independent Bruker measurements available) amounts to less than 1% on average, and is always lower than 3%, i.e. significantly lower than the estimated random errors of 7%. Indeed, the homemade instruments provide more scattered results (see comment on pg 7544, line 9-13). Due to the lack of vertical sensitivity, we only performed a simple scaling of the a priori CF₄ vertical distribution. Scatter plots indicated no correlation between the CF₄ total columns and either the solar elevation or the water vapor total columns. Their contributions to the seasonal signal are therefore very unlikely.</p>
<p>3. I would suggest to introduce the line list provided by Boudon et al. earlier in the paper (e.g. in section 2.1). Then the discussion could be a bit more extensive and more information could be provided: e.g. please quantify "consistently and significantly larger". Does use of the line list or use of the pseudolines achieve lower residuals?</p>	<p>The Boudon et al. line-by-line parameters are now introduced earlier in the paper, in section 3. It reads as follows: "A 'quality-test' covering our entire database, performed by replacing the adopted CF₄ pseudo-lines by the synthetic line parameters of Boudon et al. (2011), as included in the formal Hitran 2008 compilation (Rothman et al., 2009), led to the following appreciations: (i) line positions good; (ii) average fitting residuals ~30% higher; (iii) retrieved CF₄ columns consistently larger by ~25%. We believe that this last observed difference may primarily result from the fact that the Boudon linelist, currently restricted to the ν_3 R-branch, is only based on one high resolution FTIR laboratory spectrum at 296 K and a low pressure of 0.17 torr, while the pseudo-lines were derived from a large set of spectra recorded at numerous typical pressure and temperature sets encountered throughout the atmosphere (Nemtchinov and Varanasi, 2003)."</p>

<p>4. The construction of an a-priori taking into account the age of air as function of altitude is an interesting exercise. Because the CF4 trend has been significantly variable over the course of the observation period, it might be worth to extend this investigation. (e.g. calculate two a-prioris, a "strong increase" a-priori and a "weak increase" a-priori. What is the effect on the stratospheric or surface mixing ratios deduced from the FTIR columns?)</p>	<p>We have built a "strong increase" a priori, assuming an annual rate of change of 1.08 ppt, as measured by the AGAGE network in the 1970s and 1980s. Using this profile and the same subset of spectra as in the paper, we now derive surface mixing ratio higher by +4.2 ppt with respect to the constant vertical distribution. This difference is in good agreement with the gap observed between the green and black curves of Fig.3, in the early 1990s.</p>
<p>5. Fig.2 / text page 7546: What is a 20% smoothing function, please give more details: e.g. 20% of data points? 20% of time interval covered? Is this a running mean? Why then does the red line in Fig 2 covers the complete time interval?</p>	<p>This is not a running mean. A local smoothing is applied, using 20% of the data points (i.e. not necessarily a fixed time interval) at once. The actual bandwidth progressively considers all data points along the x-axis.</p>
<p>6. Page 7538: "In the absence of..." -> "Due to the absence of..." Acknowledgements: "Görnergrat" -> "Gornergrat"</p>	<p>These have been changed.</p>