Improved FTIR retrieval strategy for HCFC-22 (CHClF2), comparisons with in situ and satellite datasets with the support of models, and determination of its long-term trend above Jungfraujoch

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Authors' response to Anonymous Referee #2 (https://doi.org/10.5194/acp-2019-73-RC2)

We use blue text for referee's comments and black text for authors' response to these comments.

General Comment

The manuscript provides a clear and straight-forward description of the work. I recommend publication in ACP after the minor comments below are addressed.

> Thank you for your comments that help to improve this manuscript. Here follows our response to your specific and technical comments:

Specific Comments

Page 5, lines 36-39 – Is this the filtering referred to on page 7, line 26? Explain on page 7 the difference between the filtered and non-filtered in situ time series. This is the first time filtering is mentioned.

> Yes it is. These baseline data are directly provided by the AGAGE teams in charge of the in situ measurements using the method briefly explained in our manuscript and fully described in, e.g., O'Doherty et al. (2001) or Cunnold et al. (2002). In this work, we only compare our results with the AGAGE baseline data (i.e., filtered). The comparison between AGAGE Mace Head and AGAGE Jungfraujoch is done in order to emphasize that these series are in excellent agreement and that therefore we can also compare our results with Mace Head data. Finally, the difference between the AGAGE filtered and non-filtered series is shortly discussed in page 5 lines 36-39 of the original manuscript. The filtering is applied in order to produce time series representative of broad atmospheric regions, meaning that measurements directly influenced by regional pollution are filtered out.

Page 6, Section 4.1.2 – ACE-FTS HCFC-22 measurements are mentioned in the Introduction, so why aren't they included in the comparisons with the FTIR retrievals? Briefly explain why in this section. ACE data are also mentioned in the Data Availability section – is this because they was used in determining the systematic component of the Sa matrix?

> We decided to drop ACE-FTS HCFC-22 retrievals for the time series comparison because we wanted to keep an identical partial column height definition (i.e., 11.21-30 km as defined by the

information content in Section 3.3) between the datasets. Indeed, only about 20% of ACE-FTS measurements were remaining when selecting this altitude range. Nevertheless, following your comment, we decided to include ACE-FTS HCFC-22 data for the lower stratospheric decadal trend comparison. Section 5 and Table 4 have been thus modified to include the ACE-FTS 2005-2014 trends.

Page 6, lines 19-24 – Clarify what the lower boundary conditions are for – all trace gases in BASCOE? "only [a] few global observations are available . . ." – a few observations of what? Make clear which lower boundary conditions are derived from MLS and which from HGGC.

> Section 4.1.3 was modified following your comment in order to clarify what are the lower boundary condition and the simulation initial state.

Page 7, Section 4.3 – Some additional explanation should be provided regarding the comparison between the FTIR mean tropospheric mixing ratio and the in situ surface mixing ratio. Why compare the FTIR tropospheric mean mixing ratio rather than the FTIR surface value or lower tropospheric mean (I assume due to the information content)? How representative is the mean tropospheric mixing ratio of the surface mixing ratio? What error does this introduce? Figure 4 suggests that there are differences, since the FTIR mean values exhibit more seasonality than the in situ values.

> This comment is similar to the Major Comments of Referee #1. Firstly, we have modified the beginning of Section 4.3 in order to explain more clearly how the tropospheric mean mixing ratio series is build. As described in the information content and error budget Section (3.3), the characterization of the averaging kernel matrices showed that only two pieces of information could be extracted from the entire total column / profile retrieved by the inversion algorithm. The height of these partial columns / pieces of information are defined by the eigenvector of these averaging kernel matrices. Right hand side of Figure 2 further demonstrates that the separation between the two partial columns / pieces of information is located near 11 km, defining two broad ranges above and below that altitude, without any more vertical resolution. Therefore, it is not relevant nor correct to only take the mixing ratio retrieved for the surface layer.

Please refer to our response to Referee#1's comment for the discussion on the tropospheric cycle retrieved in our time series. As a short discussion was already in the original manuscript at the end of Section 4.4 (Comparison of lower stratospheric columns), we decided, for clarity, to switch Section 4.3 (tropospheric time series) and Section 4.4 and to more elaborate on the cause of the FTIR tropospheric cycle at the end of the tropospheric Section (i.e., 4.4 in the revised manuscript).

Page 8, lines 35-37 – Add these 1988-2017 total column trends to Tables 2-4.

> We added the 1988-2017 trends to Tables 2-4.

Page 10, lines 9-10 – Explain more explicitly how the improved retrieval strategy developed for HCFC-22 is transferable to other gases

> By this last sentence, we mean that we would be able to improve retrieval strategies of some other chlorine-bearing source gases (e.g., CFC-12) in order to retrieve partial columns, allowing then to independently characterize tropospheric and stratospheric trends. But for each case or species, a specific retrieval strategy has to be defined and optimized.

Page 18, Figure 4 – Why does the AGAGE vs. FTIR scatter plot have a layered structure, with a relatively constant AGAGE value for a range of FTIR values?

> As you pointed it out, FTIR tropospheric time series shows a significant annual cycle while in situ series do not. As a result, each year compared in the scatter plot are regrouped in "layer" (see Figure 1c hereafter).

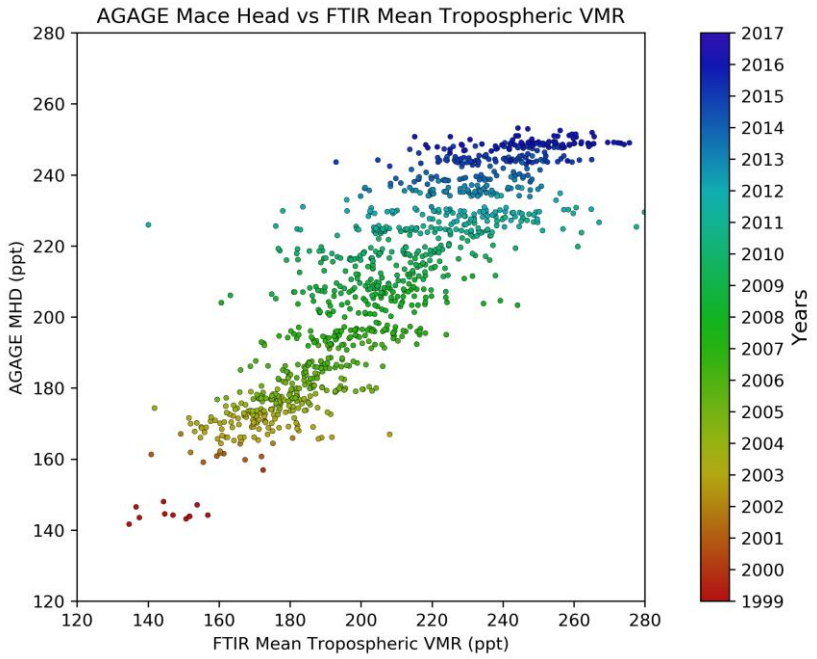

Figure 1c: Daily coincidences between AGAGE Mace Head (MHD) and Jungfraujoch FTIR mean tropospheric VMR time series. Points are coloured according their year of measurement (see right colour bar).

Page 21-22, Tables 2-4 – Why are different time periods used for calculating the trends in the total columns, tropospheric columns, and lower stratospheric columns? This makes it difficult to directly compare them. The choices should be more clearly explained in the text.

> The periods used for calculating trends are selected in order to take the best from the compared time series but also to have consecutive trend periods of 10 years to highlight the changing trend values in the three last decades.

Technical Corrections

The manuscript should be reviewed carefully for grammatical and typographical errors. For example, there are many missing commas and hyphens, unnecessary or missing "s" on words, and other errors. Some are identified below, but this list is not exhaustive.

Thank you for all these technical corrections, the large majority of them have been included in the revised manuscript, here follows the exceptions:

Page 2, line 36 – not clear what Fohn refers to – delete?

> The authors referenced in the manuscript refer Föhn events to atmospheric situations as depressions over the Bay of Biscay bringing air into the Southern rim of the Alps and thus uplifting polluted air from surface to Jungfraujoch.

Page 6, line 17 – age-of-air study

> "age of air" is used in the literature.

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