Reviewer #2

General comments:

The authors used Tikhonov regularization to retrieve CFC11 and CFC 12 at Hefei station, China. They looked at the trend and seasonal cycle during 3 years for CFC 11 and 5 years for CFC 12. Although they mention new retrieval method in the abstract, they followed a very well know method of Tikhonov regularization. They also compared trends and the averaged retrieved profile shape with the ACE satellite as well as one NDACC station in St. Petersburg, Russia. Overall, there is a great value in creating independent ground data. However, the presentation and discussion of results needs to be improved. The information content of the retrieved data is only adequate for retrieving the total column for CFC 11 and maybe two column layers for CFC 12. But authors investigated the surface level value which is not meaningful information from the retrieval. They discussed that optimal estimation method (OEM) is not able to truly retrieve CFC data from FTIR sensors which is not correct. Authors wrote in a way that there is no way to constrain the results in OEM, however using more complex covariance matrix this is very possible. Also, I think the manuscript would benefit from adding a few more NDACC station rather one NDACC station to have a more meaningful comparison and discussion of the results. I would recommend a major correction is needed before publication of this manuscript. More detail comments are provided below.

Response: We appreciate your constructive comments. The comments and proposed corrections have been taken into account and helped improving the paper. Each comment has been addressed as follows. There is an extensive discussion among the authors regarding how to revise the content. Line numbers refer to the revised manuscript (version without tracked changes).

Specific comments:

- The general motivation of this work needs to be improved. Retrieval data has one DOF for CFC 11 and two DOF for CFC12 which means they can provide information about total column (assuming that the sensitivity is up to dominant portion of the CFC profile) and two partial columns for CFC12 (using average kernels they should identify the most meaningful layers that can be retrieved). The current motivation assumes that FTIR retrieval can retrieval a detailed profile from surface to stratosphere, which is not possible based on the sensitivity of these measurement. The motivation of the study needs to be rewritten to clarify how the retrieved information adds to the satellite and in situ measurement and the value of data based on true sensitivity of the data.

Response: We revised the objective of this work on Line 111-113 in Section 1 as follows:

The objective of this paper is to obtain the CFC-11 and CFC-12 total columns from the solar spectra based on ground-based FTIR spectroscopy, and compare with the ACE-FTS satellite data, WACCM data and the data from other NDACC-IRWG stations (St. Petersburg, Jungfraujoch, and Réunion).

Retrieving CFC profile is named as of the main objective of the manuscript. However, considering the low DOF it seems one total column (or two partial columns for CFC12) can be retrieved.

Response: We replaced the CFCs profile with CFCs total columns as the main objective on Line 111-113 in Section 1.

- It is not clear why only 3 years of data is used for CFC 11 while 5 years for CFC 12. Authors should clarify this inconsistency in the periods and elaborate on how that could affect their conclusions. Moreover, trend analysis with only 3 and even five years of data is not a robust conclusion. If authors include more NDACC stations and use longer period for available data, then they could discuss the overall trend of all included stations, and how a few years of Hefei compares with recent years of other stations.

Response: The spectral range for retrieval of CFC-11 is 830-860 cm^{-1}, while the spectral range for retrieval of CFC-12 is 1160.2–1161.4 cm^{-1}. We replaced CaF_2 incoming light window with KCl window for FTIR spectrometer in December 2016, which increased the covering spectral range from greater than 1000 cm^{-1} to greater than 700 cm^{-1}. So we can retrieve CFC-11 since then. The explanation is included on Line 128-130 in Section 2.1. In addition, we added the data from two NDACC stations for comparison in Section 3.3, but the two NDACC stations do not provide long-period data.

Page 5, ln 146 to ln150. Authors mentioned that they did not use the optical estimation method because of the high fluctuation in their results. However, they can use more constrains in OEM by incorporating more a complex covariance matrix in the retrieval to achieve a more restricted result. It is fine to use the Tikhonov regularization, but the discussion of paper is in a way that OEM is limited which is not true in there are many ways to constrain your results to prevent high fluctuations. You can find some good examples of more complex covariance matrix to constrain the OEM results in these papers and many more online

1. Shams, Shima Bahramvash, Von P. Walden, James W. Hannigan, and David D. Turner. "Retrievals of Ozone in the Troposphere an Lower Stratosphere Using FTIR Observations over Greenland." IEEE Transactions on Geoscience and Remote Sensing(2022).
2. Turner, David D., and W. Greg Blumberg. "Improvements to the AERIoe thermodynamic profile retrieval algorithm." IEEE Journal of Selected Topics in Applied Earth Observations and Remote Sensing12, no. 5 (2018): 1339-1354.

Response: We read through the papers you provided, and think the discussion about that OEM is limited is not true, so we deleted this description in Section 2.3 of the revised manuscript.

- There are multiple NDACC stations, it is not clear why data is only compared with St Petersburg? It looks subjective, rather than rigorous research to find relevant and meaning full stations to compare. What is the impact of transportation and local sources. I would recommend authors use multiple station data for comparison to provide a more detail context for their comparisons. Specially that seasonal cycle has a lag time in their cycle. Having a clear discussion on different sources and sinks could cause these differences.

Response: Since CFC-11 and CFC-12 are not standard NDACC species, the data from most NDACC stations are not uploaded to the NDACC data archive. Following your suggestions, we contacted the researchers at the Jungfraujoch and Réunion stations to get the data from the two sites (for some reasons, only the recent-year data are provided). We added the data from the two NDACC stations for comparison in Section 3.3, and have a discussion on which cause the differences.

- Page 6, ln 146 to ln156. Your methodology is very similar to Polyakov et al, 2019. Please discuss if there is any difference in the method that you used. Otherwise, I would suggest to just reference their paper as the same methodology is used and there is no need to repeating the same information. Especially that authors did not show any of these matrixes in the plots. As suggested in later comments, adding plots of DOF, average kernel, and Jacobian matrix is a nice way to characterize the sensitivity of the measurements. You can add formulation of those variables to clarify their meaning. Instead use the formulation to elaborate the errors that used in the text.

Response: We deleted the repeated discussion in Section 2.3 as suggested, and added formulas of DOFs, average kernels and covariance matrices to explain their meanings on Line 169-175 in Section 2.3.

- Page 12, ln 250, authors investigate surface level CFC 11 and CFC 12 in multiple plots. The measured FTIR data has 1 DOF for CFC11, thus there is not enough information to extract the near-surface value. Because a profile is incorporated as a priori, there is a profile output, however, there is no meaningful information at all levels of the profile. That's why DOF and averaging are useful information to indicate the sensitivity of the retrieval and the vertical resolution of the results. All the investigation of surface level needs to be excluded. Instead, if the DOF and average kernel show that the measurement has adequate sensitivity to one tropospheric column, the authors can analyze that data.

Response: We deleted the discussions on near-surface value as suggested, due to the low DOFs.

Writing/presentational comment

- Hefei is not part of the NDACC. (Could be a great addition though)

Response: In the abstract and Line 120-121 in Section 2.1, we added the description that Hefei is a candidate NDACC-IRWG station now.

- page 3, ln 71 to 75. It is mentioned that Yi et al, 2021 used in situ measurement. Are these measurements still active? What is the in situ temporal resolution? Again, the text implies that the retrieval can provide surface-level information, which is not correct. The text needs to be updated. Moreover, authors can include those surface measurements in their plots to compare with local measurements. On page 3, ln 78 to 88 for each satellite, please include the vertical and spatial resolution of the retrieved CFC.

Response: There is no report shows that the in-situ measurements of Yi et al. (2021) are still active. The measurements of Yi et al. (2021) collected air samples continuously for 4 - 7 days each month. We added this description on Line 79. Because we deleted the discussion on surface concentration in the revised manuscript, we didn't make this comparison in the revised manuscript. The vertical spatial resolution of each satellite and airborne measurement were added on Line 83-97 in Section 1.

- Page 4, ln 108. Add solar before FTIR remote sensing site.

Response: We did it as suggested.

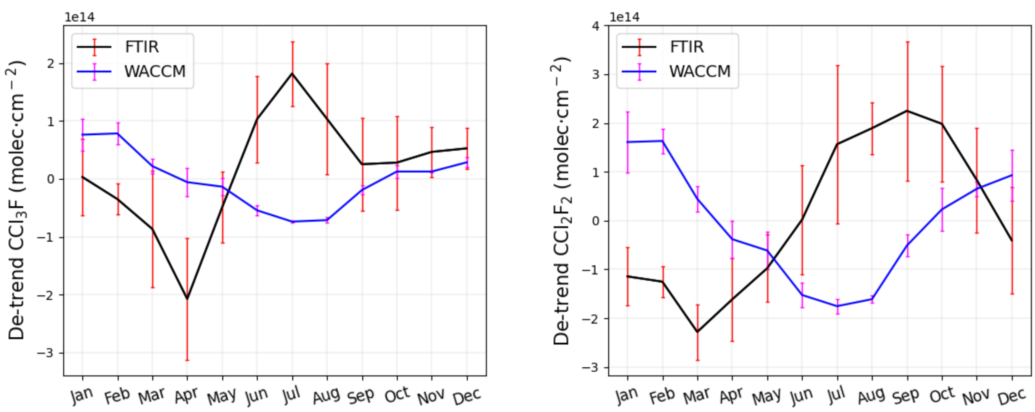
- Page 4. Ln 108 to 117: a map of the study area can be very helpful, to illustrate the location of Hefei and other NDACC stations will be added to the study.

Response: We added a map of the study area in Fig. 1. The map illustrates the location of Hefei, St. Petersburg, Jungfraujoch, St Denis and Réunion stations.

- Page 4, ln 123. It is not clear if the authors used the monthly climatology of CFC as the prior or if they used a specific prior for each month that they retrieved. (12 different profiles for each year of each gas.) also, it would be helpful to write the WACCM spatial resolution that is used in the study. I suggest the look at How the monthly variability and cycle of seasons of the received data is similar or different from prior information that is used.

Response: Here we used the averaged profiles calculated from the monthly profile of WACCM v6 at the Hefei station as the a priori profile. The WACCM v6 spatial resolution is 0.95 °×1.25°. We added the description on Line 139-140 and Line 341, respectively.

We ever made comparison between monthly variability and seasonal cycle of WACCM data and FTIR data, and the results are shown in the Figure as below. However, it can be seen that the monthly variability of FTIR and WACCM data is very different and there is no meaning information, so we didn't show this comparison in the manuscript.

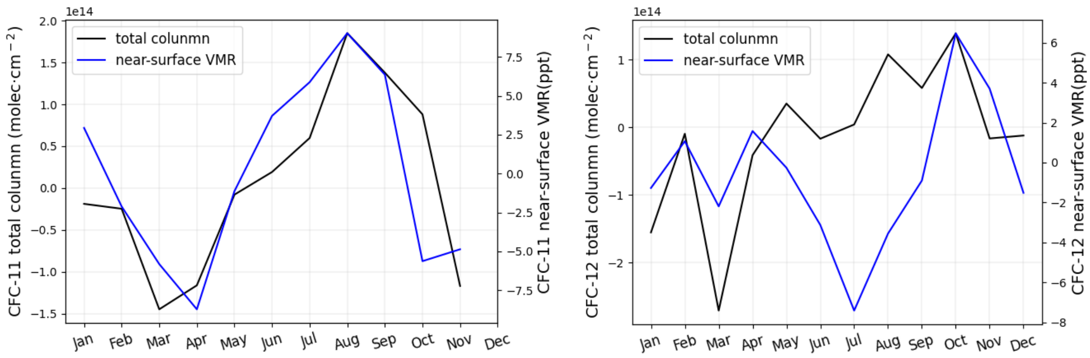


- Page 8, ln 225. De Maziere et al., 2018 did not talk about the trends. Clarify which citation is related to each part of statement in this line.

Response: We corrected this citation error.

- Page 13, ln 260. The reasons behind the one-month phase delay need to be clarified. Look at other datasets (grand measurements in particular) and investigate if this delay is persistent there.

Response: Besides Hefei station, only St. Petersburg station provided both total column and profile of CFC-11 and CFC-12 data. Therefore, we compared the difference between monthly variation of total column and near-surface (about 200m) VMR for CFC-11 and CFC-12 at the St. Petersburg station, as shown in the Figure below. It can be seen that the monthly variations of total column and near-surface VMR at the St. Petersburg site are not the same, especially for CFC-12. For CFC-11, the lowest near-surface VMR occurring in April, is one-month later than March, when the lowest total column occurs, but both the maximum occur in August. For CFC-12, both the maximum total column and near-surface VMR occur in October, but the minimum near-surface VMR is July and that of total column is in March.



Te et al. (2016) found that there was a time lag of two months between surface VMR and total column of CO in Paris and Jungfraujoch, and described this time delay might be caused by different emission patterns. Therefore, the inconsistency between the monthly variation of near surface concentration and the total column for the target gas is persistent. However, we deleted the discussion on near surface VMR for our FTIR measurements as suggested, so the discussion on this phase delay was deleted in the revised manuscript.

- Page 18, ln 365. The seasonal cycle in St. Petersburg happens in fall which contradicts your explanation of the seasonal cycle in Hefei on page 13, ln 265. “In addition, more use of air conditioning and other refrigeration equipment in summer, and foams releasing more CFCs 265 at high temperatures lead to high concentrations of atmospheric CFCs." Authors need to further explain the seasonal cycle and its subjectivity to locations especially by adding more stations to the study it would be interesting to see how they vary and if that could lead to an interesting conclusion.

Response: We added the comparison of the seasonal cycle at four stations in Section 3.3. For the seasonal cycle of CFCs, emissions are one of the influencing factors, so we modified the description of reasons for seasonal variations at the Hefei site on Line 274-283 in Section 3.1.

Fig

- Add a figure of study location as well as selected NDACC data stations.

Response: We added a map of locations of selected NDACC and Hefei stations in Fig. 1.

- Page 7. Fig 1. The total column average kernel is not very easy to comprehend. I would suggest including the DOF profile, mean averaging kernel profile, and the Jacobian matrix presentations to fully characterize the retrieval information.

Response: We added the mean averaging kernel profile and the DOFs profile in Fig. 2(c) (d) and Fig. 3(c) (d). We don't know how to plot the Jacobian matrix, so we didn't add the Jacobian matrix.

- fig 3, and fig 5 (must check all the plots) axis has ccl2f2 on their axis while the caption says CFC. The same acronym should be used.

Response: We made the acronym the same for all plots in the paper.

- Fig 5. Please include the averaged seasonal cycle from WACCM and ACE for the same period to show what information this study is bringing to the table.

Response: We ever tried to study the seasonal cycle of ACE data. Unfortunately, due to the lack of observations in tropical and subtropical areas, ACE data within 27 °N–37 °N, 107 °E–127 °E are concentrated in February and May, most data are missing in January, March, June, July, November and December. Therefore, it is difficult to do the seasonal cycle analysis for ACE-FTS data. The seasonal cycle of WACCM data is not displayed, because of the large difference with that of FTIR.

Fig 6 and fig 4. the information content of these measurements is not sufficient to have meaningful surface value from the FTIR retrieval to investigate the results.

Response: We deleted this part in the revised manuscript.

- Fig 8. It is not clear what information is depicted in this plot and what research questions are targeted here. There is so much subjectivity in two-point FTIR retrieval especially when they are located this far apart. I suggest removing this figure instead create some harmonic analysis of time series based on monthly CFC data for each station (as suggested before at lead 4 sites that are distributed in a different location) and discuss how the harmonic time series are similar or different.

Response: We deleted Fig. 8 as suggested, and added comparisons of monthly CFC-11 and CFC-12 for four stations, shown in Fig. 7 and Fig. 8 of the revised manuscript.

Other modifications:

Response: In Section 3.1 of the revised manuscript, we modified the fitting formula slightly, and used first-order polynomial and three harmonic terms to simulate CFC-11 and CFC-12 time seires, the equation $F(t)$ become:

$$F(t) = a + b \cdot t + \sum_{\mathrm{k}=1}^{3} (c_{2\mathrm{k}-1} \cos(2\pi k t) + c_{2\mathrm{k}} \sin(2\pi k t))$$

where a is the intercept, b represents annual trend, and c_1 to c_6 represent sin/cosine harmonic term coefficient. The annual decreasing rate is obtained by the parameter b.

References

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Yi, L., Wu, J., An, M., Xu, W., Fang, X., Yao, B., Li, Y., Gao, D., Zhao, X., and Hu, J.: The atmospheric concentrations and emissions of major halocarbons in China during 2009-2019, Environmental Pollution, 284, 117190. https://doi.org/10.1016/j.envpol.2021.117190, 2021.