Reviewer #1

The manuscript "Retrieval of Atmospheric CFC-11 and CFC-12 from High resolution FTIR Observations at Hefei and Comparisons with Satellite Data" by Zeng et al. describes the retrieval of atmospheric ozone depleting chlorofluorocarbons CFC-11 and CFC-12 from solar absorption spectra measured using a high resolution Fourier Transform infrared spectrometer at Hefei, China, and examines the resulting, multi-year timeseries, and compares it to satellite measurements and concentrations prognosed by a model.

The retrieval scheme builds on work carried out at the St Petersburg NDACC-IRWG (Network for the Detection of Atmospheric Composition Change – InfraRed Working Group) station. The novelty here is the retrieval of these species from the spectra measured at Hefei, representing one of the few measurements of its kind in China. The long-term monitoring of key atmospheric constituents such as these and the understanding of their evolution within a global context is important and the publication of these results should be encouraged.

The manuscript is generally well structured and written but would benefit from further development of several sections to provide more of a thorough description of some of the important concepts as described below under specific comments.

Subject to the incorporation of these changes and the corrections suggested under technical corrections below, publication of this manuscript is recommended.

Response: We appreciate your constructive and positive 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 manuscript presents retrievals of CFC-11 between January 2017 and December 2020 and CFC-12 between September 2015 and December 2020. The authors should explain why the two observing periods are different.

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.

The abstract states that comparisons are made to other NDACC stations. It should be made clearer whether Hefei is or is not an NDACC-IRWG station. Also make it clear that the comparison is with NDACC-IRWG stations, not other NDACC observations.

Response: Hefei is not an NDACC-IRWG station now, but is applying to join the NDACC-IRWG. We added this explanation on Line 121-122 in Section 2.1. We also make it clear that the comparison is

with NDACC-IRWG stations, not other NDACC observations.

The abstract also introduces the comparison to ACE-FTS satellite measurements and WACCMv6 model and presents quotative results of the comparison. It would provide important context here to define the spatial extent of the satellite/model data used i.e., global or coincident with Hefei.

Response: The spatial extent of ACE-FTS satellite data coincident with Hefei is centered at the Hefei site with latitude of ±5° and longitude of ±10°. The WACCMv6 simulated data consider the Hefei site (31.9°N, 117.17°E) as the center, with a horizontal resolution of 0.95°×1.25°. We added this explanation on Line 341 in Section 3.2.

In its current form, Section 2.2. does not provide sufficient information to allow the reader to reproduce the author's results. For example, how was the pseudo line list produced and how can it be obtained? Also, Table 1 lists zshift and beam as background retrieval parameters for CFC-11, but these are not described or explained in the text.

Response: The address providing the pseudo-line-lists is included on Line 143 in Section 2.2. Also, we added the description about zshift and beam correction on Line 147-152 in Section 2.2.

In Section 2.3, How is the measurement error used to refine the regularization strength determined? This may be covered in the cited article, but it is probably important enough to discuss within the manuscript.

Response: We modified the content of this part. According to Steck (2002), we can determine the unique value of α for the given mean retrieval measurement noise error σ_m, the measurement noise error can be selected by interest, but should be considered within a reasonable range (20% in citation). The smoothing error can also be considered to determine the value of α. Steck (2002) indicated that we can consider these two errors at the same time, and the method is to minimize the total error calculated by measurement noise error and smoothing error (because the dependence of the forward model parameter error on α is equivalent to the measurement noise error, the consideration of the forward model error is ignored). According to the posteriori error estimation method, we calculate the total error of the measurement noise error and the smoothing error $\mathbf{S}_{tot} = \sqrt{\mathbf{S}_m^2 + \mathbf{S}_s^2}$, and the results are list on Line 181-191 in Section 2.3.

It is unusual to see a column averaging kernel that contains as much structure and sharp transitions as the ones plotted in panel c of Figs 1 and 2. It would be good to include the layer averaging kernels, or a subset thereof, to see how this has come about.

Response: Following the suggestions from the two reviewers, we replaced the total column averaging kernels with layer averaging kernels in Fig. 2(c) and 3(c).

Not all sources of error listed in Table 3 are mentioned in the text of Section 2.5. These sources and the associated assumptions concerning their magnitude should be discussed.

Response: We added the other missing sources of error and their magnitude on Line 221 and 223 in Section 2.5, including the uncertainty of temperature dependency of line width, air widening of line width, H_2O spectroscopy and ILS.

At P10 L215, the sentence "The time series are fitted by a lowpass filtered fast Fourier transform (FFT) technology and a linear fitting to simulate the seasonal and interannual variation of CFC-11 and CFC-12 (Thoning et al., 1989)" may not accurately describe the timeseries decomposition process. It appears from Fig. 3 that a linear trend and multi-harmonic seasonal cycle have been fitted. The authors should consider revising this statement and state the number of harmonic terms that have been used to fit the seasonality.

Response: We used the second-order polynomial and the four harmonic terms to fit the seasonal periods of CFC-11 and CFC-12, where t is the time fraction in years, and the equation $F(t)$:

$$F(\mathrm{t}) = a + b \cdot t + c \cdot t^2 + \sum_{\mathrm{k}=1}^{4} \left(d_{2\mathrm{k}-1} \cos(2\pi k t) + d_{2\mathrm{k}} \sin(2\pi k t)\right)$$

where a is the intercept, b and c represent polynomial fitting term coefficient, and d_1 to d_8 represent sin/cosine harmonic term coefficient.

In the revised manuscript, we modified the fitting according to other sites, and reused first-order polynomial and three harmonic terms to simulate CFC-11 and CFC-12, the equation $F(t)$ become：

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

where a is the intercept, b represent annual trend, and c_1 to c_6 represent sin/cosine harmonic term coefficient. The annual trend is also obtained by the new fitting formula $F(\mathrm{t})$. We revised the statement, and describe the fitting formula in detail on Line 239-243 in section 3.1.

In section 3.1 two retrieval products are discussed: the total columns and near surface concentrations. These products should be introduced prior to their discussion. It would be helpful to do this as part of a discussion of the information content of the retrieval process possibly as its own sub-section in section 2. The error analysis should also state how the retrieval errors propagate into these two products.

Response: Following the suggestion of the reviewer 2, the discussion part about near surface concentrations was deleted, due to the insufficient sensitivity. The error analysis for retrieval of total columns is discussed in Section 2, and we added the error calculation formula in detail on Line 169-171 in Section 2.3.

In the conclusions, the statement that "ACE-FTS and WACCM data clearly overestimated the decreasing rate,.." doesn't appear to be justified in the context of the evidence presented given the spatio-temporal differences between the measurements. This should be revised.

Response: We modified the statement to "The decreasing trend of ACE-FTS and WACCM is significantly higher, while the corresponding value of FTIR total column is -0.47 ± 0.06 % yr^{-1}".

It would be good to see some stronger conclusions drawn, for example placing the findings of this work in the context of previously published findings and a comment on the differing types of emissions that lead to the difference between Hefei and St. Petersburg.

Response: There were few reports on CFC in Hefei and St. Petersburg before. St. Petersburg is close to

the industrially developed European part of Russia, and there are some sources of CFC-11 and CFC-12. At the same time, the temperature in St. Petersburg is mild in summer (about 17 °C), and the temperature in Hefei is about 29 °C in summer, the use of air conditioners in summer should be lower than in Hefei. Some studies in China indicate that the leakage of CFCs caused by waste treatment in municipal solid waste landfills and low leak tightness of automobile mobile air conditioning systems on hot and humid days may be the potential source of CFCs (Zhen et al., 2020; Zhang et al., 2017). Section 3.3 has modified to compare with more NDACC sites, as for St. Petersburg's research on emission types is insufficient, so we will not discuss emission at St. Petersburg.

Are there any plans to continue or update the dataset? It would be good to include this information.

Response: We will update our dataset later. At present, Hefei site is applying to join NDACC-IRWG. When we join it successfully, we will upload our data to the NDACC database and update the data. We added this description to the Data availability of the revised manuscript.

Technical corrections

P1 L25 in abstract remove % sign after -0.47 to be consistent with the rest of the abstract, elsewhere when expressing a value and uncertainty the parentheses are unnecessary.

Response: We have modified the contents in the revised manuscript.

P2 L56 citation should be Montzka et al., 2021.

Response: We checked the citation, and this citation is Montzka et al., 2018. Montzka et al. (2018) in Nature Letter, wrote in the 4 paragraph of the paper shows "The gap between expectations and observations widened substantially after 2012, when CFC-11 global mole fractions began decreasing even more slowly. In recent data (from mid-2015 to mid-2017), the mean rate of change for CFC-11 (-1.0 ± 0.2 ppt yr^{-1}, or $-0.4 \pm 0.1\%$ yr^{-1}) was about 50% slower than that observed during 2002–2012; it also was much slower than has been recently projected." The content is consistent with our citation, so there should be right here.

P2 L57 It might help the reader to know the type of atmospheric observations, in-situ or remote sensing.

Response: Observations in Gosan, South Korea, and Hateruma, Japan are in-situ observations. We added this explanation on Line 63 of the page 2.

P2 L60 insert a space between CFC-11 and and.

Response: We did it in the revised paper.

P2L60 check the units are correct for the emission rates (Gg not kg?) and use yr^{-1} to be consistent with the rest of the manuscript

Response: We corrected the unit for the emission rates.

P2 L64 "Study of the temporal-spatial distribution and variations of CFCs in the atmosphere is of great

significance to reduce stratospheric ozone depletion and greenhouse gas emissions." The study itself does not reduce the emissions, but it does improve understanding and suggest what needs to be done to facilitate reductions. Consider revising this sentence.

Response: We modified the statement to “Study of the temporal-spatial distribution and variations of CFCs in the atmosphere is of great significance for improving understanding and implementing policies to reduce stratospheric ozone depletion and greenhouse gas emissions”.

P3 L80 this sentence may need a change of emphasis, in that HIRDLS, ILAS etc are not mainly used for CFC measurements, but they may be the main instruments used for this type of measurement.

Response: We changed the sentence to “Satellite remote sensing techniques, such as high resolution dynamics Limb Sounder (HIRDLS), Improved Limb Atmospheric Spectrometer (ILAS), the collocated Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) and Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS), also play an important role in measuring the global distribution of CFCs”.

P3 L92 Throughout the manuscript there are sentences like this where the un-parenthesised citation is used at the beginning of the sentence with the parenthesised version at the end. It is unnecessary to include the citation twice in one sentence.

Response: We modified this kind of citation in the revised manuscript.

P5L 141 remove the word time and replace with either iteration or step, i.e., “iteration index i” or “step i”.

Response: We modified it to “iteration index i”.

P9 L207 Suggest starting the sentence introducing the error values from the Polyakov study with "At the St Petersburg site..." or similar, to avoid a little confusion.

Response: We modified the description here following this suggestion.

P9 L209 Last sentence should be elaborated.

Response: We modified this sentence to “Our error estimates are similar to those at the St. Petersburg station, and slightly smaller compared with the latter”.

P9 Table 3, This table is a little hard to read, consider more use of horizontal lines to separate items

Response: We added horizontal lines to Table 3.

P10 L219 Throughout section 3.1 trends are given the units %/yr-1 when they should be %yr-1 (to be consistent with the rest of the manuscript) or %/yr

Response: We modified all units %/yr-1 to “% yr^{-1}” in the manuscript.

P10 L225 Insert a space between -0.49 and %

Response: We did it in the manuscript.

Figures 3, 4 and 8: Consider using the same x axis scale for both timeseries to allow the reader to see the seasonal cycles aligned.

Response: Figures 4 use the same x axis scale for both timeseries. We deleted Figure 8, which shows timeseries comparison, but now added the comparison of monthly means of the two data in Figure 7 and 8.

P13 L254 In this discussion, are the seasonal amplitude and variability not the same? I.e., the amplitude in units of molecules per unit area or mixing ratio is also expressed as a percentage of annual mean or detrended mean?

Response: The seasonal amplitude and variability are not the same. We added the explanation of the seasonal amplitude and variability on Line 267-269 in Section 3.1.

P13 L256 consider starting a new paragraph to discuss near surface concentration seasonality

Response: The discussion about near surface concentration was deleted, according to the comments of reviewer 2, as the information content is not enough to obtain meaningful surface values from FTIR retrieval.

P13 L271 CFC-11?

Response: We modified it to "CFC-11".

P15 L304 It should be explained why the columns of dry-air mole fractions are being compared and not molecules per unit area that were discussed previously. Also, it is not apparent in the text how the dry-air column has been derived.

Response: Because the profile range measured by ACE-FTS satellite is inconsistent with the FTIR measurement range, the dry air mole fractions are calculated here for comparison. It is also facility to compare with the ground and upper air mixture ratios obtained from field measurements in other regions of China. We added the dry-air column formula in Eq. 8.

P16L333 Are global WACCM data used or the same spatial criteria as the ACE-FTS? This should be made clear in the text.

Response: the simulated data consider the Hefei site (31.9°N, 117.17°E) as the center, with a horizontal resolution of 0.95°×1.25°. We added this introduction on Line 340-341 in Section 3.2.

P17 Fig. 8. Check the y-axis label of panel (a)

Response: We deleted Figure 8, according to the comments of reviewer 2.

P17 L361 It would make life easier for the reader if the column differences were expressed as a percentage.

Response: We deleted Figure 9 and the discussion about Figure 9, according to the comments of reviewer 2.

P18 Fig. 9. Include the parameters of the linear regression

Response: We deleted Figure 9, according to the comments of reviewer 2.

P18 L380 The meaning of the last sentence is unclear. Perhaps: "This is one of the few..."

Response: We modified the sentence to “This is one of the few reports about the detection of CFC-11 and CFC-12 columns and their tempo-spatial variations in China”.

P19 L404 Start a new paragraph for the St. Petersburg comparison.

Response: We modified the part of conclusion.

P19 L407 It would be good to go on to describe the emission source differences

Response: We modified the part of conclusion.

References: There are some inconsistencies in formatting of the references, e.g. the use of capitalised journal and article titles, which should be rectified.

Response: We modified the format of all references.

References

Montzka, S. A., Dutton, G. S., Yu, P., Ray, E., Portmann, R. W., Daniel, J. S., Kuijpers, L., Hall, B. D., Mondeel, D., Siso, C., Nance, D., Rigby, M., Manning, A. J., Hu, L., Moore, F., Miller, B. R., and Elkins, J. W.: An unexpected and persistent increase in global emissions of ozone-depleting CFC-11, Nature, 557, 413-417. https://doi.org/10.1038/s41586-018-0106-2, 2018.

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