

*We thank the referee for valuable and useful comments and remarks.*

While the final results and comparisons are reasonable the description of the spectral analysis and retrieval process is deeply limited and flawed and have be improved before acceptance for publication is conferred.

*We rewrote some parts of these sections, see new version of our manuscript.*

‘Major’ section

L 106: “The difficulties of the freons TCs retrievals are caused, first of all, by small values and a smoothed spectral dependency of the radiation absorption by these gases which lead to the low information content of the FTIR measurements with respect to the freons TCs.” This statement is vague, and poorly worded. While it may be colloquially expressing a practical opinion of someone doing retrievals it could be and would be more useful to readers if filled out more technically.

*We rephrased the text:*

The informativeness of the FTIR spectra with respect to target gases abundancies is low due to several reasons. First, the absorption of target gases is small. Therefore, for example, even for low sun (solar elevation of about  $15^\circ$ ), the transmission of solar radiation due to CFC-11 absorption is greater than 90 %, due to HCFC-22 absorption is close to 75 %, and only for CFC-12, the transmission is close to 50 %. Secondly, there is an absorption of interfering gases in the spectral range considered.

Thus, the CFC-11 absorption band is overlapped with several strong water vapor absorption lines and HNO<sub>3</sub> absorption band, each of the CFC-12 and HCFC-22 absorption bands is overlapped with a wing of water vapor absorption line. Finally, the CFC-12 and at a larger extent the CFC-11 bands have smoothed spectral dependency of absorption that requires to use wide micro-windows for retrieving their abundancies,  $2\text{ cm}^{-1}$  for CFC-12 and not smaller than  $30\text{ cm}^{-1}$  for CFC-11. These factors cause the difficulties in halocarbons retrieval from FTIR spectra measurements.

L128 – 136: “The analysis of the Inverse Problem Solution Process (IPSP)”, This apparent procedure is not described, therefor the methodology to determine the characteristics of the QHN, its full effect on spectra and subsequent retrievals, perhaps straight forward or perhaps more sophisticated is unknown. It likely would be of wide interest. Consequently the reader does not know how the author came to the exclusion of some 450 spectra.

*We described in detail the process of preliminary analysis of the spectra quality and slightly corrected the quantities.*

*New text:*

The period of the channeling is caused by the material and the thickness of the filter, and in the spectral region  $800\text{-}900\text{ cm}^{-1}$  it is close to  $1.1\text{ cm}^{-1}$ , while the amplitude of the channeling varies from zero to a few percent value depending on random filter positioning parameters. To analyze the presence and the amplitude of the channeling, we performed the Fourier analysis in the most transparent spectral range  $892\text{-}905\text{ cm}^{-1}$  for harmonic components with periods in the intervals of  $1\text{-}1.25\text{ cm}^{-1}$ . The channeling amplitude was calculated relative to the mean signal value in this spectral range.

It is worth mentioning that the SFIT4 software supports the accounting for channeling and its compensation in a spectrum. However, our estimations showed that when the channeling amplitude exceeded 2%, this compensation became unsatisfactory due to a significant increase of scatter in the results of retrievals. Thus, we excluded such spectra from the further processing. In addition, we analyzed the autocorrelation coefficient of a dark noise in the range of  $660\text{-}680\text{ cm}^{-1}$  (except a slope) and excluded the spectra with the averaged autocorrelation coefficient greater than 0.1. A large autocorrelation coefficient of the dark noise indicates the presence of

external influences on a measurement process. Moreover, we excluded from the further processing the spectra that were measured at the time when a haze or cloudiness was observed in any part of the sky, since the use of these spectra also noticeably increases the scatter of the results.

As a result of the described filtering, 2901 from 3523 (i.e. 82%) spectra measured before February 2016 were selected for further processing. In February 2016, the F3 filter was replaced by the standard IRWG NDACC filter f6 which wedge-shaped design eliminates the channeling. Thus, the quality of measurements was improved, and 1903 from 1958 spectra were selected, giving in a sum 4804 spectra for 2009-2019 period.

L145-150: “the main criterion for choosing the optimal values of setup parameters was the stability of the target gas TCs during a day. More precisely, the root mean square value (RMS) over all days for SD of the gas TCs per a day was minimized. Along with the daily variability of the TCs, the mean value and the SD of the information content of measurements (degrees of freedom for signal, DFS) (Rodgers, 2000, p. 19) as well as the estimates of the systematic and random measurement errors and the spectral residual  $\hat{\chi}^2$  the RMS difference between measured and calculated spectra for the retrieved state of the atmosphere ( $\chi^2$ )” Listing these does not explain how they are used. This section requires a thorough explanation.

*We extended the discussion giving clarification of the criteria used by Polyakov et al., 2019a,b, 2020b :*

In previous studies, Polyakov et al. (2019a, b, 2020a) determined a number of parameters of the retrieval strategy using the SFIT4 code for deriving the TCs of the target gases from the FTIR measurements at the St. Petersburg site: the boundaries of microwindows, the mean/apriori profiles of the measured gases, the magnitude and variability of the zero level, periods for taking into account (or excluding) the channeling, and the background shape of a spectrum (BSS). The criteria used for optimization of retrieval parameters are briefly described below. As we mentioned above, the lifetime of the target gases in the atmosphere is more than 10 years. In addition, CFC-11 and CFC-12 have no known active sources of emission. Therefore, we expect the stability of their retrieved columns during both the each day and the whole period of measurements (excluding trend). At a lesser extent due to its continuous production, the same criterion is valid for HCFC-22, at least for intraday variability. Thus (Polyakov et al., 2019a, b, 2020a) used the stability of the retrieved total columns in terms of minimal root-mean-squared (RMS) SD of the TCs for all days of measurements as the main criterion in selecting the retrieval parameters. Another important retrieval parameter is the number of degrees of freedom for signal (DFS) (Rodgers, 2000, p. 19)

for target gases. As a criterion for optimization, the SD of DFS was minimized. The estimate of total systematic and random measurements errors was also considered. Finally, the residuals (differences between spectra measured and calculated with the retrieved atmospheric state), for estimating of which the RMS residuals normalized to the unit, calculated in the SFIT4 software, and denoted as  $\chi^2$ , were analyzed. It should be noted that without additional analysis, the listed criteria do not unambiguously determine the optimality of the retrieval technique. The estimate of the TCs measurement errors can be considered as one of the main criteria, but the adequacy of the measurement model also should be taken into account. Thus, for example, by including during spectra analysis the additional unknown parameter as channeling, we increase the measurements errors, however, if we exclude it and residuals get larger, it will indicate that the parameters used are inadequate for real measurements, i. e. the actual presence of channeling in the spectra.

L150: “Table 1 presents the main optimized parameters obtained in previous studies.”  
Table 1 does not specify any of the mention retrieval parameters.

*The retrieval parameters corresponding to the criteria optimized (lines 145-150) are given in Table 1, as it is defined above.*

L152 – 154: “Target gas absorption is calculated based on pseudo–lines (see [mark4sun.jpl.nasa.gov/pseudo.html](http://mark4sun.jpl.nasa.gov/pseudo.html) for pseudo–lines), interfering gases absorption is calculated based on spectroscopic information from the HITRAN database”, this is not true please see the list of interfering species.

*We corrected the text.*

Target gas and COCl<sub>2</sub> absorption is calculated based on pseudo–lines (see [mark4sun.jpl.nasa.gov/pseudo.html](http://mark4sun.jpl.nasa.gov/pseudo.html) for pseudo–lines), other interfering gases absorption is calculated based on spectroscopic information from the HITRAN database which is described in detail by Polyakov et al. (2019a, b, 2020a).

L163: “The main factor that determines the shape of the SBL is the filter spectral transmission function (STF).” The spectral baseline is typically 0% transmission line. It typically is not affected by the optical filter transmission or envelope.

*We used a term SBL for denoting the spectra envelope corresponding to the unit transmission. We corrected the text avoiding using the term SBL.*

L163 – 169: Several points require more detail. Does the water vapor continuum effect the artificial light source spectra? The solar spectra? or both? “contribution in the considered spectral region under conditions of the St. Petersburg site can significantly exceed 50 %.” This 50% of what exactly?, “For a 30 cm<sup>-1</sup> window, the selectivity of continual uptake is sufficient to influence the IPSP results.” Completely unclear what this statement refers to. If this this continuum is a feature of the spectra and well modeled then some plot should be shown to prove it has been resolved.

*The continuum absorption of radiation, in this case, by water vapor in the transparency window of the spectra, is observed in a very humid atmosphere (see Mlawer, et al, 2012). In the Fig. A1 of Appendix A, we presented an example of transmission due to absorption by water vapor continuum for a day with a large water vapor content. In dry atmosphere, this absorption can be neglected. Moreover, we estimated the contribution of water vapor continuum numerically by analyzing spectra with and without it considering in the most humid days and gave the results in the text. The ignoring of continuum absorption in these days led to overestimates of the CFC-11 TCs by an average of 3%. Although this value is smaller than the measurement error, it is systematic and depends on the water vapor content. Therefore, it is necessary to consider the water vapor continuum. All these considerations are valid only for observational sites near the sea level and cannot be applied to the measurements at high altitude sites (like Jungraujoch).*

*New text:*

Water vapor continuum makes a significant contribution to radiation attenuation by the atmospheric water vapor (Mlawer et al., 2012). Our calculations have shown that radiation absorption by water vapor continuum in the considered spectral region under conditions of the St. Petersburg site can significantly exceed 50%. For a 30 cm<sup>-1</sup> window, the selectivity of continual uptake is sufficient to influence the spectra processing results. To calculate the water vapor continuum, we used a free–distributed computer code (MT\_CKD, 2017) and the daily profiles of water vapor independently derived from the FTIR measurements (Virolainen et al., 2017). In the first approximation, the contribution of the water vapor continuum to absorption is proportional to the water vapor partial pressure squared, and it can be detected only in a very humid atmosphere. We estimated the contribution of water vapor continuum numerically by analyzing spectra with and without it considering on the most humid days in 2018: July 29, August 2 and 9. The neglecting continuum absorption in these days led to overestimates of the CFC-11 TCs by an average of 2.9 %.

Although this value is less than the measurement error (see Table 4), it systematically depends on the water vapor content. Therefore, it is necessary to consider the water vapor continuum. In Fig. A1 of Appendix A, we present an example of transmission due to absorption by water vapor continuum.

#### Appendix A: Water vapor continuum

Figure A1 depicts the transmission functions of water vapor continuum. Although sometimes a value of precipitable water (PW) above St.Petersburg reaches 50mm, in clear-sky days, when FTIR measurements are performed, maximum value of PW is about 30-40mm.

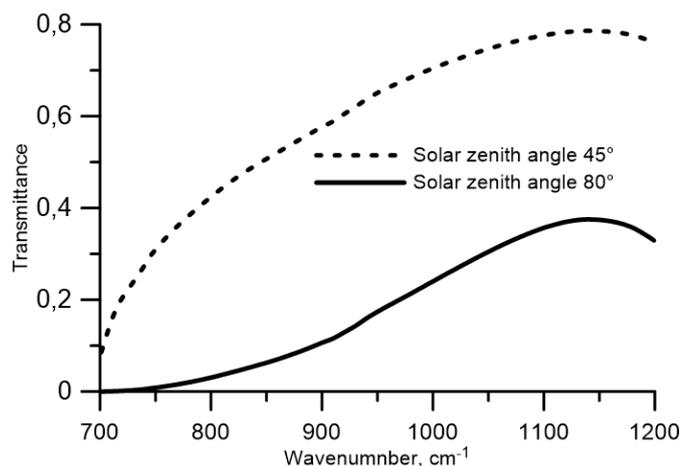


Figure A1. Transmission function of water vapor continuum for July 29, 2018 - one of the humid days of measurements in 2018 (PW totals 30 mm).

L170 – 183: This paragraph tries to explain the process of modeling the ice. Its is still not clear where the ice is in the optical path. But the mention of LN2 assumes its at the detector. This should be made clear. Was the WV continuum modeling used in the retrieval? Appears not but its not clear. It appears the a simple quadratic background was used as is standard in many retrievals. The term cryo-sediment does not seem appropriate for the feature.

*The ice is located directly on the cooled receiver (HgCdTe detector). The measurements of radiation absorption by ice were carried out using an artificial source of light, therefore the continuous absorption by atmospheric water vapor is not involved in these measurements. The quadratic background shape of the spectra can, of course, be used to simulate various phenomena, but its use must be justified and limited in accordance with the physical nature of the simulated phenomenon. The fact is that the quadratic component of the SBL in the case of a smoothed spectral dependence, like CFC-11, is poorly separated from the gas content, therefore, additional “freedom” (a priori uncertainty of the coefficient) can significantly distort the results obtained.*

*We replaced the term "cryo-sediment" with the term "amorphous water ice" – AWI.*

*New text:*

Repeated measurements of the STF showed that over time they exhibit a specific spectrum of absorption by amorphous water ice (AWI) formed on the HgCdTe detector at the temperatures that has a detector cooled by liquid nitrogen (e.g. Hudgins, et al., 1993; Lynch, 2006). The absorption of radiation by AWI depends on its thickness which increases during the measurement period and decreases during the period of inactivity of the instrument when the receiver is not cooled. In addition, the water vapor from the atmospheric air gradually (on a monthly scale) seeps into the evacuated zone of the instrument and also leads to an increase of the AWI thickness. To compensate for its variability, the BSS was refined with the second-degree wavenumber polynomial implemented in the SFIT4 code. With turning on one more variable, the correction of the BSS curvature specified by the coefficient at the second

power of the wavenumber (hereinafter – curvature value) can lead to “overfreedom” of the solution. To avoid this, the a priori curvature value uncertainty was limited. The parameters for compensating the BSS due to absorption by the AWI were selected in two steps. We minimized the intraday variability of the CFC-11 TCs in a series of spectra processing and, on the first step, got the a priori thickness of the AWI (0.3  $\mu\text{m}$  for F3, 0.9  $\mu\text{m}$  for f6 filter) with the a priori curvature value of 0. On the next step, we optimized the value of a priori curvature uncertainty as  $10^{-6}$  for both filters.

L205 – 214: The author should explain the large difference in the curves F12 versus F11 & F22  
*We added the explanation in the text:*

It is worth mentioning that the difference between the curves for different halocarbons in Fig. 1 is meaningful. The presence of a pronounced minimum for CFC-12 is due to a larger information content of spectral measurements with respect to CFC-12 abundancies compared to CFC-11 and HCFC-22 (DFS for CFC-12 is 1.2, CFC-11 - 1.05, HCFC-22 - 1.0, see Table 3). The reason for this is a weak absorption of interfering gases in the spectral range for CFC-12 retrievals. Thus, for CFC-12, an increase of the regularization parameter, which tightens the requirement for spectrum smoothness, leads to the suppression of useful information on the elements of the vertical gas distribution which is contained in a spectrum. Consequently, the intraday variability of retrievals is increasing. For CFC-11 and HCFC-22, the informativeness of spectral measurements is less, DFS is close to 1, therefore, large values of the parameter and the corresponding requirements for smoothness do not contradict the information contained in a spectrum.

L211: if the author is referring to a profile scaling procedure it should be clearly stated so e.g. “first guess profile multiplier.”

*Corrected:*

This can be interpreted as the complete absence of the information on the vertical profile of HCFC-22 in spectral measurements, i.e. only the information on the first guess profile multiplier (profile scaling approach). We performed the retrieval for both the profile scaling and the T–Ph approach with ....

L215-239: and Fig’s 2-4: This section seems to compare a single (per species?) constraint called ‘OE’ with an alpha optimized T-P constraint. First the OE a priori ( $S_a$ ) is not given or described. Further since it is only one of a large possible array of constraints the comparison is in no way of general significance or value to the reader as well as mis-labeled. This section is so lacking in information as to mis lead the reader. This section needs significant redress before re-submission. To wit the final statement regarding the contradicting conclusion found in Prignon is not explained.

*We rewrote this section:*

It should be noted that the target parameter of the retrieval, TC, is calculated from the initially retrieved vertical profile of the gas. Therefore, it is important to control the retrieval of trace gases profiles. For all three target gases, figures 2–4 depict the sensitivity functions of the TCs to relative variations in the gases profile at different heights (left) (see about averaging kernel (AK) area (Rodgers and Connor, 2003, section 2.1)) and the examples of initial (first guess) and retrieved volume mixing ratio (VMR) profiles (right). All curves are shown for two typical measurements: in a fall–winter season with a low Sun elevation and a low humidity, and in summer, with a high for the site latitude Sun elevation and a wet atmosphere. All parameters in

the figures are given for a regularization of both the OE and the T-Ph methods. Although the T-Ph regularization parameter was optimized, the covariance matrices of the OE method were taken from (Polyakov et al., 2018), where they were selected from general considerations. Figures 2–4 demonstrate that the sensitivity, which for the ideal case should be equal to 1 at all heights, turns out to be noticeably lower (from 0.5 to 0.8 for different gases, seasons and methods) at the surface. Then sensitivities increase, reaching a maximum at heights of 8–12 km for CFC-11 and CFC-12, and at heights above 12 km for HCFC-22 which is due to a higher stratospheric content of HCFC-22. Above, the sensitivity decreases which can no longer be significant due to the fall of the VMR of the target gases. As seen from Fig. 2–4, the measurement conditions have a significant effect on the sensitivity functions. In winter, when the Sun elevation is low corresponding to a thicker atmosphere in the solar beam path and low water vapor content, the information content of measurements is higher than in summer. For all three gases, the sensitivity is far from the unit to a greater extent in the lower troposphere in high humidity conditions in summer. Using T-Ph approach and choosing the regularization parameter based on minimizing the intraday variability of TCs, we obtained DFS = 1 for HCFC-22; the DFS value of other two gases is close to 1 (1.05 and 1.20, see Table 3). Prignon et al. (2019) reported the higher values of DFS (D = 1.97) caused by T-Ph regularisation with parameter  $\alpha = 9$  and a low atmospheric water vapor content above the mountain (3580 m a.s.l) site Jungfraujoch.

Table 3 & 4: Table 3 and the upper section of table 4 should be combined into one table and the lower part of table 4 should stand alone as table 4 giving the uncertainties of the retrievals

*Done*

L245 – 250: Please add to the tables how many spectra were removed in each step to remove outliers. This would be instructive on ‘far’ versus ‘near’ outliers were removed & overall data quality.

*New table was added as appendix B:*

Table B1. The criteria used and the percentage of data discarded after their application.

Criterion	CFC-11		CFC-12		HCFC-22	
	Value	Excluded, %	Value	Excluded, %	Value	Excluded,%
Sys err	7.96	2.9	2.58	2.5	5.91	0
Ran err	4.77	5.0	4.42	4.7	9.50	4.8
$\chi^2$	1.45	0.5	1.54	0.3	0.97	4.1
DFS	0.89	0	1.10	1.3	0.90	0
S/N	50-600	7.9	50-600	3.7	60- 600	4.2
Not conv	Yes	0	Yes	8.1	Yes	3.7
Div	Yes	3.8	Yes	0	Yes	3.0
No result	No files	4.2	No files	1.2	No files	0
Total excluded		19%		18%		16%
Spectra/Days	before filtering	4773 / 720		4768 / 718		4585 / 714
	after filtering	3864 / 678		3912 / 664		2855 / 663

Rows of Table B1:

- 1) systematic error (mean plus 2 SD)
- 2) random error (mean plus 2 SD)

- 3) residual ( $X_i$ ) (mean plus 2 SD)
- 4) DFS (mean minus 2 SD)
- 5) To exclude noisy spectra and possible non-linearity in measurements, we use only measurements with SNR values ranging from 50 (60) to 600
- 6) Not converged
- 7) Divergence warning
- 8) SFIT did not present results

L260: “spectral residuals vary from 0.34 to 0.52 % depending on the gas; it corresponds to the SNR values of 209,280, and 327” please explain (equation?) how these correspond?

*We corrected the text, adding word “mean”. Since the spectrum in residual calculations is normalized to unit, SNR and residual there are reciprocal values,  $SNR = 1 / \text{residual}$ . As we are talking about mean values, this equation is approximate.*

*New text:*

Ideally, the spectral residual should be equal to the measurements noise level. For target gases, the mean values of the spectral residuals vary from 0:34 to 0:52% depending on the gas; it corresponds to the SNR values of 209; 280, and 327 for CFC-11, CFC-12, and HCFC-22, respectively. Since the spectrum in residual calculations is normalized to unit, SNR and residual there are reciprocal values,  $SNR = 1 / \text{residual}$ . Comparing these values to the preliminary determined mean SNR in the opaque spectral range (364;351 and 324, in the same order of gases), we see that for CFC-11 and CFC-12 they are slightly less and for HCFC-22 are nearly the same. This means that for CFC-11 and CFC-12 the radiative transfer model and a set of parameters used, although satisfactory, but not ideally describe the absorption of radiation by the atmosphere and the observational system, whereas for HCFC-22 the retrieval technique works in the best way.

L265 – 272: Earlier the authors state they have a modeled covariance. This could be a reasonable estimate and consequently the calculation could be performed and would be informative.

*We do not have true covariance matrices available. The calculation based on the model matrix depends significantly on this matrix.*

L293 – 297: There is no explanation of why or how the optical filter could have such an effect on the variability as it is a static or passive component. Some explanation is required to support this statement.

*We added an explanation in the text:*

Due to channeling, the F3 filter (used before February 2016), leads to a large scatter in retrieval results owing to larger errors (see section 2.1).

L310: “which does not have a systematic component during a day, to the random error.”

This is not clear, What is a systematic component to a random error?

*Apparently, the reviewer kept in mind line 301.*

*We removed the phrase below:*

~~This is due to the largest contribution of the measurement noise (row 17 of Table 4), which does not have a systematic component during a day, to the random error.~~

Minor

L57 “Since Molina and Rowland (1974) have reported that CFCs accumulated in the Earth’s atmosphere lead to an increased rate of ozone depletion, the attention of both scientists and policymakers to the ozone hole problem has been increasing. “ This statement may have been true in the 1990’s but not so today.

*Corrected*

After Molina and Rowland (1974) reported that CFCs accumulated in the Earth’s atmosphere led to an increased rate of ozone depletion, the attention of both scientists and policymakers to the ozone hole problem had been increased.

L66 – Use of atmospheric content is not standard, often atmospheric burden when referring to the total column is used.

*Corrected*

Based on the 2015–2017 data, Montzka et al. (2018) showed that the rate of change in the CFC-11 atmospheric concentration decreased by .....

L73 & Amendments should be added after Montreal Protocol

*Added*

In the 2000s, the production and consumption of HCFCs in developed countries have decreased as a response to the Montreal Protocol and its amendments and adjustments.

L86-95 This review is not thorough. Certainly, any review of FTIR CFC efforts needs to include Rinsland 2010 and references therein.

*We studied the papers suggested and added them in overview and in reference sections. Thank you for the useful information.*

First FTIR measurements of atmospheric HCFC-22 were performed from the balloon in early 80-s (Goldman et al., 1981). Spectral resolution of these measurements did not allow halocarbons to be measured from the surface. Later, with the appearance of high-resolution instruments, halocarbons started to be derived with ground-based FTIR spectrometers. In last decades, TCs of halocarbons are measured by ground-based FTIR method more actively (e.g. Notholt, 1994; Rinsland et al., 2005, 2010; Zander et al., 2005; Mahieu et al., 2010, 2013, 2017; Zhou et al., 2016; Prignon et al., 2019).

L111: “the Tikhonov–Phillips (T–Ph) approach which is more suitable for long–lived gases with a pronounced trend.” – this statement is obvious or well known and requires a reference.

*We corrected the text and added the following reference.*

Tikhonov–Phillips (T–Ph) approach which allows more stable results to be derived than the optimal estimation (OE) method (e.g. Senten et al., 2012).

C. Senten, M. De Mazi`ere, G. Vanhaelewyn, and C. Vigouroux Information operator approach applied to the retrieval of the vertical distribution of atmospheric constituents from ground-based high-resolution FTIR measurements, Atmos. Meas. Tech., 5, 161–180, 2012, doi:10.5194/amt-5-161-2012

L124: “The observational system is based on a Bruker FS125HR Fourier spectrometer, but some of the equipment is non–standard.” Doe the author mean in an NDACC-IRWG sense?

*We explained it in the text:*

The observational system is based on a Bruker IFS125HR Fourier spectrometer, but some of the equipment is non-standard. In particular, before February 2016 a non-standard (for the IRWG-NDACC sites) spectral filter (hereinafter F3) was used for measurements in the spectral region with target gases absorption bands. Since this filter was plane-parallel, a parasitic interference arose in it, leading to the appearance of an effect of the optical resonance (“channeling”), see (Blumenstock et al., 2020). Moreover, a home-made solar tracking system is used.

L125: “a non-standard spectral filter F3 was used for measurements in the spectral region with considered freons absorption bands.” There is no reference for ‘F3’. If a local name it should be referenced as such.

*Yes, this notation is now introduced, see previous response.*

L128 – 136: The author should also refer to this as ‘channeling’ its more common name and insert the in press [Blumenstock AMT 2021] for a reference.

*Corrected, see previous response.*

L137: “For a preliminary assessment of the signal to noise ratio (SNR), the standard deviation (SD) of the signal”, Its not clear but presumably the SD of the SNR, Please clarify.

*No, as it is indicated in the text, the SD of the signal is considered, not of the SNR.*

L146: “More precisely, the root mean square value (RMS) over all days for SD of the gas TCs per a day was minimized. “ This not clear at all, please re-phrase.

*We corrected the text.*

At a lesser extent due to its continuous production, the same criterion is valid for HCFC-22, at least for intraday variability. Thus (Polyakov et al., 2019a, b, 2020a) used the stability of the retrieved total columns in terms of minimal root-mean-squared (RMS) SD of the TCs for all days of measurements as the main criterion in selecting the retrieval parameters

L150:  $\chi^2$  is within the nomenclature of SFIT is a normalized part of the convergence criteria. Is it being used here in that capacity or of simply renaming  $RM = \chi^2$ ? This needs clarification.

*Here  $\chi^2$  indicates that this is the SFIT estimated value, the normalized residual.*

*Considered:*

*the residuals (differences between spectra measured and calculated with the retrieved atmospheric state), for estimating of which the RMS residuals normalized to the unit, calculated in the SFIT4 software, and denoted as  $\chi^2$ , were analyzed.*

Table 2: Not readable needs to be reformatted with clear rows and columns

*We removed the table and rewrote text:*

The using of a wide spectral window for CFC-11 retrieval ( $30 \text{ cm}^{-1}$ , see Table 1) is unusual for deriving the information on the gas content from the high resolution IR spectra and requires the non-standard approach for considering the base spectra shape (BSS). This approach was described in detail by Polyakov et al. (2020a); the main features of this approach are listed below.

The constant and important factor that determines the BSS is the filter spectral transmission function (STF). We have measured the STF in a special experiment using an artificial light source.

Repeated measurements of the STF showed that over time they exhibit a specific spectrum of absorption by amorphous water ice (AWI) formed on the HgCdTe detector at the temperatures that has a detector cooled by liquid nitrogen (e.g. Hudgins, et al., 1993; Lynch, 2006). The absorption of radiation by AWI depends on its thickness which increases during the measurement period and decreases during the period of inactivity of the instrument when the receiver is not cooled. In addition, the water vapor from the atmospheric air gradually (on a monthly scale) seeps into the evacuated zone of the instrument and also leads to an increase of the AWI thickness. To compensate for its variability, the BSS was refined with the second-degree wavenumber polynomial implemented in the SFIT4 code. With turning on one more variable, the correction of the BSS curvature specified by the coefficient at the second power of the wavenumber (hereinafter – curvature value) can lead to “overfreedom” of the solution. To avoid this, the a priori curvature value uncertainty was limited. The parameters for compensating the BSS due to absorption by the AWI were selected in two steps. We minimized the intraday variability of the CFC-11 TCs in a series of spectra processing and, on the first step, got the a priori thickness of the AWI (0:3  $\mu\text{m}$  for F3, 0:9  $\mu\text{m}$  for f6 filter) with the a priori curvature value of 0. On the next step, we optimized the value of a priori curvature uncertainty as  $10^{-6}$  for both filters.

Water vapor continuum makes a significant contribution to radiation attenuation by the atmospheric water vapor (Mlawer et al., 2012). Our calculations have shown that radiation absorption by water vapor continuum in the considered spectral region under conditions of the St. Petersburg site can significantly exceed 50%. For a  $30\text{ cm}^{-1}$  window, the selectivity of continual uptake is sufficient to influence the spectra processing results. To calculate the water vapor continuum, we used a free-distributed computer code (MT\_CKD, 2017) and the daily profiles of water vapor independently derived from the FTIR measurements (Virolainen et al., 2017). In the first approximation, the contribution of the water vapor continuum to absorption is proportional to the water vapor partial pressure squared, and it can be detected only in a very humid atmosphere. We estimated the contribution of water vapor continuum numerically by analyzing spectra with and without it considering on the most humid days in 2018: July 29, August 2 and 9. The neglecting continuum absorption in these days led to overestimates of the CFC-11 TCs by an average of 2.9 %. Although this value is less than the measurement error (see Table 4), it systematically depends on the water vapor content. Therefore, it is necessary to consider the water vapor continuum. In Fig. A1 of Appendix A, we present an example of transmission due to absorption by water vapor continuum.

Thus for CFC-11 processing, we took into account STF, AWI variability and water vapor continuum.

L190: “by a priori information of the Tikhonov–Phillips” T-P is an ad hoc constraint not actually a priori information.

*Corrected by removing this expression.*

L192: “Unlike the OE, the T–Ph approach does not “pull” the solution to the mean profile”, The OE does not pull, the retrieved profile retains the a priori value when there is no new information from the spectra. Also not to the ‘mean’ rather the a priori.

*When there is no information on target gas in spectral measurements, the retrieval surely equals a priori mean profile. If there is a little information, the solution pulls the a priori mean. The Gaussian distribution is described by the mean and the covariance matrix; in this study, we use an OE with a priori Gaussian distribution of the retrieved vector. We added the word a priori.*

L195: “the OE approach requires the use of the covariance matrices for describing the variability of the target gases profiles,” not so, an array of ad hoc constrains can be applied within the OE context.

*We corrected the formulation of the method used.*

...OE approach, with the apriori information given in a form of normal distribution of the target gas profile, requires the use of the covariance matrices...

L202: choice (sp)

*corrected*

L209: ‘and the both’ is awkward maybe should be ‘and both’

*corrected*

L211: if the author is referring to a profile scaling procedure it should be clearly stated so e.g. “first guess profile multiplier.”

*corrected*

This can be interpreted as the complete absence of the information on the vertical profile of HCFC-22 in spectral measurements, i.e. only the information on the first guess profile multiplier (profile scaling approach).

L216: specify section and / or page of appropriate discussion in Rodgers & Connor 2003.

*corrected*

Section 2.1

L251: “geographical latitude”, is redundant.*corrected*

L248: does “not provide a solution” mean not converge or other issues, or both?

*Both. Either a logical parameter indicates the absence of convergence or divergence of iteration (when the number of iterations reaches the maximum), or there is no file with the results (the software processing is interrupted for unknown reasons).*

L310: “which does not have a systematic component during a day, to the random error.”

This is not clear, What is a systematic component to a random error?

*Apparently, the reviewer kept line 301 in mind.*

*In intraday measurements, there is a shift in the retrievals in this day (the reasons may be different). The magnitude of this shift is different for different days. Therefore, when considering the whole data set, this shift is a component of a random error, but when considering one day of measurements, this is a component of a systematic error. We corrected the text.*

EQ2 sin ()

*corrected*

L406: ‘belt’ might better be ‘range’

*corrected*

L418: “a noticeable seasonal variations” rather: “a noticeable seasonal variation”

*corrected*