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Atmospheric Chemistry and Physics Discussions

Interactive comment on "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" *by* Maxime Prignon et al.

Anonymous Referee #2

Received and published: 28 April 2019

General Comments

This manuscript presents an improved retrieval strategy for deriving HCFC-22 from ground-based solar absorption spectra of the $2\nu6$ Q-branch recorded by two FTIR spectrometers at the NDACC Jungfraujoch station. The retrieval uses the SFIT-4 code with Tikhonov regularization to obtain independent tropospheric and lower stratospheric partial columns of HCFC-22, as well as total columns. The resulting time series extends from 1988 to 2017 and is used to derive trends in tropospheric, lower stratospheric, and total columns over the 30-year record and to examine differences between

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individual decades in the tropospheric and total column trends. The HCFC-22 growth rate in the troposphere is shown to have slowed in the last decade. The FTIR mean tropospheric mixing ratios agree well with AGAGE in situ data and the lower stratospheric columns agree well with MIPAS satellite data. The FTIR measurements are also compared with the BASCOE CTM and WACCM models, showing that WACCM lower stratospheric columns are biased low relative to the FTIR and BASCOE. All of the lower stratospheric datasets exhibit a seasonal cycle that is anti-correlated with the mean age of air derived from a BASCOE-CTM simulation.

The results of this study are of importance in monitoring the effectiveness of the Montreal Protocol and its amendments, particularly given the length and vertical sensitivity of the FTIR data record at Jungfraujoch. The FTIR retrieval strategy for HCFC-22 could be extended to other NDACC stations and could be adapted and applied to other chlorine-containing gases.

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

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.

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?

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.

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.

Page 8, lines 35-37 – Add these 1988-2017 total column 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.

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?

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.

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.

Abstract, Page 1, line 15 – HCFCs

Abstract, Page 1, line 15 – first, but temporary,

Abstract, Page 1, line 15 – change "as" to "for"

Abstract, Page 1, line 15 – ozone-depleting

Abstract, Page 1, line 16 – CFCs

Abstract, Page 1, line 17 – Layer,

Abstract, Page 1, line 20 – Fourier Transform infrared is used here, but Fourier Transform InfraRed on page 2, para 1

Abstract, Page 1, line 20 – high-altitude

Abstract, Page 1, line 24 – delete miscellaneous

Abstract, Page 1, line 25 – ozone-depleting substances (ODSs)

Page 1, line 30 – chlorine-containing

Page 1, line 35 – HCFCs

Page 1, line 35 – change "as" to "for"

Page 1, line 35 – CFCs

Page 1, line 40 – total HCFC emissions

Page 2, line 4 – satellite

Page 2, line 13 – space-borne and ground-based

Page 2, line 14 – delete "has"

Page 2, line 15 – ACE-FTS has been performing solar occultation measurements since

Page 2, line 16 – although

Page 2, line 19 – high-resolution

Page 2, line 19 – stations currently retrieve HCFC-22 abundance.

Page 2, line 22 – could also mention the Paris Agreement here

Page 2, line 23 – by the detection of an unexpected

Page 2, lines 26 and 28 – Here and throughout the manuscript, “section” should be changed to “Section” or “Sect.” following ACP formatting guidelines. Do a search and replace.

Page 2, line 31 – affiliated with the

Page 2, line 32 – conditions

Page 2, line 34 – experiences

Page 2, line 35 – This particular location also enables study of the

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

Page 2, line 40 – study,

Page 3, line 2 – detectors), and have been recorded at two spectral

Page 3, line 9 – well-isolated

Page 3, line 23 – National Centers

Page 3, line 24 – and extrapolated above

Page 3, line 34 – were tested to optimize

Page 3, line 37 – fitted alone gives poor

Page 4, line 3 – enables retrieval of more

Page 4, line 11 – spectral

Page 4, line 11 – signal-to-noise ratio (SNR), root-mean-square

Page 4, lines 19-21 – Here and throughout the manuscript, all vectors and matrices should be in bold font.

Page 4, line 24 – DOFS, the trace of the averaging

Page 4, line 25 – change “expresses” to “indicates”

Page 4, line 33 – four components

Page 4, line 34 – bold font for the matrix term

Page 4, line 38 – off-diagonal

Page 4, line 39 – set to 3 km

Page 4, line 39 – 5% relative

Page 5, line 6 – abundance, so the

Page 5, line 7 – as is obvious

Page 5, line 19 – outdoors

Page 5, line 32 – have been performed . . . since

Page 6, line 14 – nine heterogeneous

Page 6, line 17 – age-of-air study

Page 6, line 19 – Is LBC really needed as an acronym? Could just use “lower boundary condition” throughout for better clarity.

Page 6, line 22 – for the year

Page 6, line 23 – HCFC-22, the

Page 6, line 34 – (Kinnison et al., 2007)

Page 6, line 37 – horizontal grid and on a grid with 66 vertical levels, with the

Page 7, line 1 – LBC, [or lower boundary condition,]

Page 7, line 4 – resolutions

Page 7, line 4-5 – the different vertical sensitivities

Page 7, lines 9-10 – Use bold font for vectors and matrices.

Page 7, line 13 – kernel

Page 7, line 18 – add space before “being”

Page 7, line 19 – time series

Page 7, line 22 – with the AGAGE in situ time series

Page 7, line 23 – delete “here”

Page 7, line 25 – very well for

Page 7, line 27 – Unfiltered time series show similar results, with

Page 8, line 4 – Is this sentence intended to be a paragraph?

Page 8, line 5 – which is within

Page 8, line 6 – see Section 3.3)

Page 8, line 6 – BASCOE lower stratospheric HCFC-22 time series is slightly lower than these two

Page 8, line 8 – delete “of”

Page 8, line 11 – trend from the monthly mean lower

Page 8, line 19 – delete “as well”

Page 8, lines 20-21 – barely detectable in the MHD

Page 8, line 24 – height statistics

Page 8, line 24 – Could add the range of tropopause heights at JFJ over the year.

Page 8, line 30 – trend uncertainty

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Page 8, line 35 – column trend

Page 8, line 38 – 1980s and early 1990s

Page 8, line 41 – 1980s

Page 9, lines 3-4 – calculated using AGAGE data

Page 9, line 8 – The 2005-2012 . . . are compared to the

Page 9, line 22 – (66 +- 6) %

Page 10, lines 1-2 – agreement for the

Page 10, line 9 – this improvement in retrieval strategy . . . essential for monitoring

Page 17, Figure 2, line 10 – The first eigenvector

Page 17, Figure 2, line 11 – instruments, and their second eigenvector has

Page 18, Figure 3, line 2 – time series of HCFC-22 total columns

Page 19, Figure 5 legend – Bruker Fourier fit

Page 19, Figure 6, line 8 – Seasonal cycle of HCFC-22 columns

Page 20, Table 1, lines 1 and 2 – "Homemade" or "homemade"? The latter is used elsewhere.

Page 20, Table 1, line 2 – text (Sect. 3.3)

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