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Atmospheric
Chemistry
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Discussions

Interactive comment on "Simultaneous atmospheric measurements using two Fourier transforminfrared spectrometers at the Polar Environment Atmospheric ResearchLaboratory during spring 2006, and comparisons with the AtmosphericChemistry Experiment-Fourier Transform Spectrometer" *by* D. Fu et al.

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Thank you to Dr. Sussmann for the thorough and detailed list of comments and suggested improvements to our paper. We were able to address all of his points and believe that with his aid we were able to improve the paper.

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In the following, we present the original comments in italics and our responses below. Quotes from the revised manuscript are in quotation marks. References cited can be found in the revised manuscript if they were not given in this response letter.

General comments:

This paper deals with partial column measurements of ozone relevant stratospheric species. Three related issues are covered, namely intercomparison of two different ground-based FTS instruments, validation of a new atmospheric chemistry satellite mission (ACE), as well as a geophysical analysis of the stratospheric chemistry and dynamics evolution over Eureka in spring 2006.

The paper is generally put on a good scientific basis and the covered issues fit well into the journal scope. The following general and specific remarks are intended to help along the process towards final publication in ACP.

Side-by-side intercomparisons of ground-based remote sounding instruments are of very high interest within the NDACC (Network for the Detection of Atmospheric Composition Change) community. There have been earlier studies (e.g., Murphy et al., 2001; Wunch et al., 2007) and there are planned future intercomparisons (e.g., Sung et al., to be submitted to ACP, 2008). It would be desirable to make it more transparent to the reader what the complementary information of this paper is relative to these others. Also the relation of this paper compared to other ACE validation papers could certainly be made more obvious.

The message of the paper could be presented more efficiently. The subsequent specific remarks are intended to provide some hints where to shorten or restructure the wording, and correct details. In addition, since co-authors are involved who have shown many times before their ability to write excellent papers, it would be highly

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desirable if they would actively contribute to this process.

Specific comments:
5305/1: Title is lengthy. Probably it could be shortened

This has been changed.

“Simultaneous trace gas measurements using two Fourier transform spectrometers at Eureka, Canada during spring 2006, and comparisons with the ACE-FTS”

5307 <Abstract>:

The abstract as is tells in a qualitative way what has been done: FTS-FTS comparisons, FTS-ACE comparisons, time evolution of ozone relevant trace species. This could be said in one or two sentences at the beginning. But what then is missing is the outcome of what has been done, which would be the main content of the abstract. Give a very short summary of all important results of your paper. It is certainly more than the statement that your intercomparisons are consistent with earlier studies. Give numbers or at least quantitative wording for all important new findings.

The abstract has been updated as suggested. The new version is given below.

“The 2006 Canadian Arctic ACE (Atmospheric Chemistry Experiment) Validation Campaign collected measurements at the Polar Environment Atmospheric Research Laboratory (PEARL, 86.42 °W, 80.05 °N, 610 meters above sea level) at Eureka, Canada from 17 February to 31 March 2006. Two of the ten instruments involved in the campaign, both Fourier transform spectrometers (FTSs), were operated simultaneously, recording atmospheric solar absorption spectra. The first instrument

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was an ABB Bomem DA8 high-resolution infrared FTS. The second instrument was the Portable Atmospheric Research Interferometric Spectrometer for the Infrared (PARIS-IR), the ground-based version of the satellite-borne FTS on the ACE satellite (ACE-FTS). From the measurements collected by these two ground-based instruments, total column densities of seven stratospheric trace gases (O_3, HCl, $ClONO_2$, HF, HNO_3, NO_2, and NO) were retrieved using the optimal estimation method and these results were compared. Since the two instruments sampled the same portions of atmosphere by synchronizing observations during the campaign and used consistent retrieval parameters, the biases in retrieved columns from the two spectrometers represent the instrumental differences. Mean differences in total column densities of O_3, HCl, $ClONO_2$, HF, HNO_3, and NO_2 from the observations between PARIS-IR and the DA8 FTS are 2.8%, -3.2%, -4.3%, -1.5%, -1.9%, and -0.1%, respectively. Partial column results from the ground-based spectrometers were also compared with partial columns derived from ACE-FTS version 2.2 (including updates for O_3) profiles. Mean differences in partial column densities of O_3, HCl, $ClONO_2$, HF, HNO_3, NO_2, and NO from the measurements between ACE-FTS and the DA8 FTS are -5.9%, -8.5%, -11.8%, -0.9%, -6.6%, -7.6%, -21.6%, respectively. Mean differences in partial column densities of O_3, HCl, $ClONO_2$, HF, HNO_3, NO_2 from the measurements between ACE-FTS and the PARIS-IR are -5.2%, -4.6%, -2.3%, -4.7%, 5.7%, -11.9%, respectively. This work provides further evidence of the reliability of ACE-FTS measurements from the first three years of on-orbit observations. Column densities of O_3, HCl, $ClONO_2$, and HNO_3 from the three FTSs were normalized with respect to HF and used to compare the time evolution of the chemical constituents in the atmosphere over Eureka during spring 2006."

Also, more quantitative information has been added to the Summary and Conclusions.

"Mean differences in partial column densities of O_3, HCl, $ClONO_2$, HF, HNO_3, NO_2, and NO from the measurements between ACE-FTS and the DA8 FTS are -5.9%, -8.5%, -11.8%, -0.9%, -6.6%, -7.6%, -21.6%, respectively. Mean differences in partial column densities of O_3, HCl, $ClONO_2$, HF, HNO_3, NO_2 from the measurements between ACE-FTS and the PARIS-IR are -5.2%, -4.6%, -2.3%, -4.7%, 5.7%, -11.9%, respectively."

5308 <Introduction>

Some reorganization/grouping would help to improve readability. Usually, in the introduction the state of the art would be described, then some deficiency or lack within this, and then how the present paper intends to improve upon this deficiency, i.e., the focus or goal would naturally be developed towards the end of the introduction. Currently, something about the focus/goal is said already in the first paragraph (5308/16) and then again and again at several places throughout the whole introduction.

To improve readability, the introduction has been reorganized and some material has been moved to Sec. 2. This rewrite also included the recommendations from Dr. Hase. The new version of the introduction is given below.

"As part of the validation program for the Atmospheric Chemistry Experiment (ACE), a series of springtime measurement campaigns have been held in Eureka in northern Nunavut (Kerzenmacher et al., 2005; Walker et al., 2005; Manney et al., 2008; Sung et al., 2007; Fraser et al., 2007). The primary objective of these campaigns is to provide a multi-year, high-latitude data set for validating satellite data obtained under the perturbed conditions present in the springtime Arctic stratosphere.

ACE, also known as SCISAT, is a Canadian-led satellite mission for remote sensing of the Earth's atmosphere from a circular, low Earth orbit (altitude 650 km, inclination 74°).

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This high inclination orbit provides global measurements over each season with a primary focus on the Arctic and Antarctic regions. An infrared Fourier transform spectrometer (ACE-FTS; Bernath et al., 2005) together with a dual, ultraviolet(UV)-visible-near-infrared spectrophotometer (Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation (ACE-MAESTRO); McElroy et al., 2007) are the scientific instruments onboard the satellite. The primary goal of the ACE mission is understanding the chemistry and dynamics of ozone in the upper troposphere and stratosphere. Thus SCISAT measures the concentrations of more than 30 chemical constituents, including many that influence the distribution of stratospheric ozone (Bernath et al., 2005; Bernath, 2006).

Validating data products from satellite-borne infrared FTSs, such as ACE-FTS, can be challenging because of the wide range of atmospheric species that are measured. Ground-based FTSs, such as those that are part of the Network for the Detection of Atmospheric Composition Change (NDACC, http://www.ndsc.ncep.noaa.gov/), can contribute significantly to these efforts because they cover a similar spectral range with high spectral resolution (e.g., Vigouroux et al., 2007; Cortesi et al., 2007; Wetzel et al., 2007). Some information on the altitude distribution of the atmospheric trace gases can be retrieved from the shapes of the spectral lines obtained from these ground-based FTSs (e.g., Pougatchev et al., 1995; Rinsland et al., 1998). This allows partial column[1] densities to be calculated from the ground-based measurements and it is these quantities that are compared with partial columns derived from the satellite observations.

As reported in other papers in this Special Issue, measurements from ground-based FTSs at 12 NDACC stations around the world have been employed in the ACE val-

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[1] Throughout this paper, the term "total column" is used to indicate that the column amount was calculated from the full altitude range of the ground-based measurement (typically from the ground to 100 km) whereas the term "partial column" is used for results obtained over a narrower altitude range. Depending on the type of ground-based measurement, the total column retrieved may be more sensitive to one region of the atmosphere than another (for example, NO_2 retrievals from FTSs are only sensitive to the stratospheric part of the total column; Sussmann et al., 2005).

idation program (e.g. Clerbaux et al., 2008; De Mazière et al., 2008; Dupuy et al., 2009; Kerzenmacher et al., 2008; Mahieu et al., 2008; Strong et al., 2008; Wolff et al., 2008). This paper presents complementary work focusing on comparisons of ground- and satellite-based FTSs during Arctic springtime and on results for trace gas species that play an important role in ozone depletion processes that occur each spring in the polar vortex, including O_3, the chlorine and nitrogen reservoir species (HCl, $ClONO_2$ and HNO_3), NO_x (NO and NO_2), and a stratospheric tracer (HF) (e.g. Solomon, 1999).

The 2006 Canadian Arctic ACE Validation campaign was the third in this series of campaigns. It took place at the Polar Environment Atmospheric Research Laboratory (PEARL, 80.05° N, 86.42° W, 610 m above sea level) in Eureka, Nunavut, Canada, from 17 February to 31 March 2006. PEARL is located in a region of large stratospheric variability in the Arctic (Harvey and Hitchman, 1996), where there is a higher probability of making measurements both inside and outside the polar vortex. Ten scientific instruments were used during the campaign. These included PARIS-IR (Portable Atmospheric Research Interferometric Spectrometer for the Infrared, a terrestrial version of the ACE-FTS) (Fu et al., 2007), a high-resolution Bomem DA8 FTS that is part of NDACC (Donovan et al., 1997; Wiacek et al, 2006; Farahani et al., 2007), and balloon-borne ozonesondes (Davies et al., 2000; Tarasick et al., 2005). These instruments were used to derive total columns, partial columns, and vertical profiles for most of the ACE target species, as well as temperature and pressure.

Herein, we describe the 2006 campaign observations made by PARIS-IR and the Environment Canada (EC) DA8 FTS and discuss comparisons both between the ground-based instruments and with the ACE-FTS results. To compare the ground-based and satellite results, this work first investigates the differences between the retrieved columns obtained by PARIS-IR, which is a relatively “new” campaign instrument, and the DA8 FTS, which is a permanently installed instrument and has been making long term observations at PEARL since 1993. To focus on the differences in the vertical columns that arise from the instrument performance, PARIS-IR and the

DA8 FTS were configured to measure atmospheric absorption spectra simultaneously. Details on the two ground-based FTSs and their observation strategies are illustrated in Sect. 2. The ground-based FTS retrieval method and measurement characterization are described in Sect. 3. The ACE-FTS measurements and retrievals are described in Sect. 4. In Sect. 5, total columns, partial columns and column ratios obtained from simultaneous atmospheric remote sensing measurements using PARIS-IR and the DA8 FTS at Eureka are reported. The results are used to compare measurements from PARIS-IR and the DA8 FTS, to investigate the quality of ACE occultation measurements, and to examine the consistency of the time evolution of the chemical constituents in the atmosphere over the Canadian high Arctic during spring 2006 obtained from these different data sets. ”

5309/5-17: This section, describing instrumentation which is not exploited within this paper, could be cancelled or at least shortened.

This section provides the general background information on the campaign. As shown in the following revised text, it was shortened by removing the instrumentation that was not directly involved in this work. Since the ozonesonde measurements were used in the comparisons, they are retained in the revised text, along with the FTIRs.

“Ten scientific instruments were deployed during the campaign. These included PARIS-IR (Portable Atmospheric Research Interferometric Spectrometer for the Infrared), a terrestrial version of the ACE-FTS (Fu et al., 2007) a high-resolution Bomem DA8 FTS (Donovan et al., 1997; Wiacek et al, 2006; Farahani et al., 2007), and balloon-borne ozonesondes (Davies et al., 2000; Tarasick et al., 2005).”

5311/5: <2 Ground-based instrumentation and observations>

This section could be shortened since both PARIS-IR and the DA8 at Pearl have been *described in earlier papers. Restrict to changes/amendments relative to what has been said in earlier papers.*

We would prefer to include the updated information for the PARIS-IR instrument instead of shortening this section since it provides basic information such as spectral coverage and resolution. The following three sentences are added at the end of the first paragraph in section 2, "Ground-based instrumentation and observations".

"Although PARIS-IR was deployed at Eureka in 2004 and 2005, there were major instrument changes and improvements for the 2006 campaign. In the 2004 campaign, undesirable spikes in the interferograms recorded by PARIS-IR resulted in spectral channelling. The temperature control system in the metrology diode laser of PARIS-IR was unstable in the 2005 campaign resulting in degraded spectral resolution. In addition, PARIS-IR was realigned by the manufacturer in mid-2005 and its performance was improved."

5314/13: <. . . the DA8 FTS used a Hamming function>

Explain: why not boxcar?

Since 1993, the DA8 FTS interferogram processing has been done routinely using the Hamming function. Originally, this was done to reduce spectral noise. It was found to have no impact in the SFIT1 retrievals. The observations made during spring 2006 also used the Hamming function, to maintain consistency with the approach that has always been used for the instrument.

5314/22: <. . . the ILS parameters obtained from the cell measurements were used as a priori inputs and values were retrieved from the atmospheric spectra . . .>

Specify: which parameters exactly did you retrieve?

What would be the difference if you retrieve ILS parameters using ideal ILS parameters as a priori?

Explain: why do you additionally retrieve the ILS parameters from the atmospheric spectra and not just input the ILS parameters you retrieved from the cell measurements as fixed parameters into the forward model?

To answer the first question (what parameters are retrieved), we have rewritten the sentence to include the phrase Empirical Apodization Parameters.

"The ILS parameters obtained from the cell measurements were used as the a priori values for the SFIT2 Emperical Apodization Function (EAP). The EAP is defined as a polynomial and SFIT2 allows for the subsequent retrieval of the polynomial coefficients as part of the state vector. We chose to retrieve third-order polynomial coefficients and a first-order polynomial coefficient for PARIS-IR and the DA8 FTS, respectively."

Second question regarding using ideal ILS parameters as a priori:

The nominal ILS would use a boxcar function defined by its maximum optical path difference, along with a correction for self-apodization due to off-axis rays arising from the finite field of view. An ideal ILS would be a pure sinc function (i.e., no off-axis rays). The EAP function in SFIT2 is used to account for additional self-apodization or modulation efficiency effects (i.e., beyond the effect of the finite field of view). The values of these parameters for the nominal ILS are (by definition) zero. It might be fine to use zeroes as the a priori values for the DA8 FTS, where only one order of the empirical function is required to achieve a good match for the ILS. Although the ILS was improved for PARIS-IR in 2006, we observed that the modulation efficiency was decreased by 20%. PARIS-IR requires 3 orders in the empirical function, and using zeroes for the a priori values is not really appropriate. The cell measurements are better than the ideal case.

Third question regarding why you don't simply use the parameters derived from the cell measurements:

Values for the empirical apodization parameters from the cell measurements were derived from the analysis of N_2O lines in the wavenumber range 2400-2800 cm^{-1}. Note, however, that the parameters required to properly model the PARIS-IR ILS vary with wavenumber. Fixing the parameters to the values derived for one spectral region will lead to errors in retrievals that employ other spectral regions. To reliably fix the parameters for atmospheric retrievals to the results from the cell measurements, we would need to generate parameters at several points across the PARIS-IR wavenumber range from the cell measurements, and then either use interpolation for the points in between or fit the variation of the parameters to some function of wavenumber (linear, quadratic,. . .). Unfortunately, with the cell measurements available, it is not possible to achieve sufficient coverage of the ACE-FTS wavenumber range to use this approach. Therefore, we retrieved the EAP function each microwindow individually and this significantly decreased the spectral fitting residuals. Errors associated with the uncertainties in the EAP function were provided in Table 2. This was found to be one of the major error sources for the PARIS-IR and DA8 FTS measurements.

5315/13: <Smoothing error, also known as null space error in the Rodger's OEM formalism, arises from the limited altitude resolution of the observing system. . . >

Rodger's – > Rodgers

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I know this is a frequently made statement, which is not correct, however. Limited altitude resolution is not the only contribution to smoothing error. There are other possible inherent physical contributions, e.g., the loss of sensitivity above certain altitudes (this is an effect in addition to the limited altitude registration/resolution, it is, e.g., due to line saturation), or non-physical secondary contributions like the smoothing effect from the (mathematical) regularization imposed upon the retrieval to reduce (trade off against) retrieval noise error. These things might be difficult to become both detailed and correct here. I suggest to skip any hand waving explanation, and just give a specific literature reference instead.

Changed to "Smoothing error, also known as null space error in the Rodgers OEM formalism, mainly arises from the limited altitude resolution of the observing system (Rodgers 1990, 2000)."

5315/25: <. . . (DOFS), which are the number of independent quantities obtained from the observations>

you probably mean: obtained from the retrievals; or: retrieved from the observations.

Changed to "As shown in Table 2, the results from the DA8 FTS typically have higher values of Degrees Of Freedom for Signal (DOFS), which are the number of independent quantities retrieved from the observations, than those from PARIS-IR since the DA8 FTS has a spectral resolution 10 times higher than that of PARIS-IR."

5315/26: <. . . (the vertical partial columns in a specified altitude range, in this case)>

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This is a frequently made miss-interpretation: DOFS gives the number of independent pieces of information; this is not equal to a number of partial columns that can be retrieved independently: you can easily check this out by looking at your n=DOFS different partial column averaging kernels: they are never rectangular functions, which your statement implies, however.

Deleted the phrase “(the vertical partial columns in a specified altitude range, in this case)”.

5316/5: <According to the Rodger’s OEM theory, the averaging kernel is the derivative of a derived parameter with respect to its a priori state value . . . >

Rodger’s $->$ Rodgers

First of all: don’t talk about a derived parameter, talk about a retrieved state vector instead (averaging kernel for a retrieved scalar parameter is a very special issue of rare practical relevance).

Secondly, the averaging kernel matrix is the derivative of the retrieved state vector with respect to the true state (not with respect to the prior state).

Thirdly, be more correct in wording, please distinguish: i) averaging kernel matrix, ii) averaging kernels (which are the rows of the matrix), and iii) total column column averaging kernel.
All in all, I suggest to avoid any self-made explanations of elsewhere described issues, just talk about total column averaging kernels here.

Please note that the manuscript only refers to total column averaging kernels. The term “averaging kernel matrix” did not appear in the text at all. The text is revised

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to:
“According to Rodgers OEM theory, the averaging kernel is the derivative of a derived vector with respect to the true state vector (Rodgers, 1990, 2000).”

5316/6: <. . . when this normalized derivative is small (nearly 0) all of the information comes from the a priori and when it is large (near 1) then the information in the retrieval comes mainly from the measured spectra (Rodgers, 1976, 1990, 2000).>

The derivative for calculating averaging kernel matrices is per definition unitless - in this sense it is trivial that it is normalized and this must not be mentioned. Confusion arises from the fact that the state vector quantity can be normalized (or not): you can calculate an averaging kernel matrix for some state vector quantity (e.g., profile given in VMR) or for a normalized state vector (e.g., scaling factors for each layer of a profile, which is the case with SFIT2). In both cases the averaging kernel matrix is unitless as said above. The rows of the matrix of the second case are sometimes regrettably called normalized kernels, although it is meant that the underlying state vector is normalized. Anyway, it is important to be aware of this difference since the resulting averaging kernel matrix will look different in general.

Again, I suggest to just talk about total column averaging kernels (since this is what is shown in Fig. 1, cancel <normalized> in the Figure caption), and avoid all unnecessary explanations. Your wording <normalized derivative> causes also some additional confusion, since total column averaging kernels can per definitionem not be directly derived from a retrieval using a normalized state vector like SFIT2.

Finally, since the wording is not absolutely clear, I like to ask this question on the underlying procedures: how do you exactly calculate total column averaging kernels shown in Fig. 1, knowing the fact that SFIT 2 is internally using a normalized state vector? Detail the individual steps of your procedure, please.

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We have removed ‘Normalized’ in Figure 1 caption and also deleted ‘normalized’ in 5316/6

Dr. Clive D Rodgers provided the formalism to calculate the averaging kernel matrix $\mathbf{A}$, i.e.,

$$\mathbf{A} = \mathbf{S_a K}^{\mathsf{T}} \left(\mathbf{K S_a K}^{\mathsf{T}} + \mathbf{S}_{\epsilon} \right)^{-1} \mathbf{K}, \tag{1}$$

where $\mathbf{S_a}$, $\mathbf{S}_{\varepsilon}$ and $\mathbf{K}$ are the a priori uncertainty covariance, the measurement noise covariance, and the weight function, respectively. SFIT2 program outputs the $\mathbf{S_a}$, $\mathbf{S}_{\varepsilon}$ and $\mathbf{K}$ for each retrievals. Then, each element in the averaging kernel matrix at the ith row and jth column, $\mathbf{A_{i,j}}$, can be determined using equation (1).

We would like to obtain averaging kernel matrix $\mathbf{A}$ in terms of relating the retrieved profile to the “true” profile in column density units, so we need to transform $\mathbf{A}$ accordingly. The detail derivation of the mathematical forms of averaging kernels for partial/total column is available in the presentation given by Dr. Steve Wood in NDSC Infrared Working Group during the NDSC 2004 meeting at Queenstown. The following is the basic derivation for the each element in column averaging kernel matrix from Wood’s presentation.

The retrieved column density at the ith layer, $\mathbf{\hat{c}_i}$, can be written as

$$\mathbf{\hat{c}_i} = \mathbf{\hat{x}_i N_i}, \tag{2}$$

where $\mathbf{\hat{x}_i}$ and $\mathbf{N_i}$ is the retrieved volume mixing ratio for the target species and the air column density at the ith layer, respectively.

Similarly, the real column density at the ith layer, $\mathbf{c_i}$, can be written as

$$\mathbf{c_i} = \mathbf{x_i N_i}, \tag{3}$$

where $\mathbf{x_i}$ is the real volume mixing ratio for the target species.

The each element at the ith row and jth column in the column averaging kernel matrix, $A_{i,j}$, can be written as

$$\mathbf{A^c_{i,j}} = \frac{\partial \mathbf{\hat{c}_i}}{\partial \mathbf{c_j}}. \tag{4}$$

Substituting equations 2 and 3 into equation 4 gives

$$\mathbf{A^c_{i,j}} = \frac{\partial \mathbf{\hat{c}_i}}{\partial \mathbf{c_j}} = \frac{\partial (\mathbf{\hat{x}_i N_i})}{\partial (\mathbf{\hat{x}_j N_j})} = \frac{\partial \mathbf{\hat{x}_i}}{\partial \mathbf{\hat{x}_j}} \left(\frac{\mathbf{N_i}}{\mathbf{N_j}} \right) = \mathbf{A_{i,j}} \left(\frac{\mathbf{N_i}}{\mathbf{N_j}} \right) \tag{5}$$

Combining equations 1 and 5, each element in the column averaging kernel matrix was calculated and the row of this matrix is the column averaging kernel.

We have developed our calculation code using IDL code provided to NDSC stations by Dr. Steven Wood. Further details on the total column averaging kernel method used for PARIS-IR are available in Section 4 of Sung et al (2007). The following is the citation of references noted above.

Error Group Summary: S. Wood, NDSC IRWG, Queenstown, November 9-12, 2004.

Sung, K., Skelton, R., Walker, K. A., Boone, C. D., Fu, D. and Bernath, P. F.: N_2O and O_3 arctic column amounts from PARIS-IR observations: Retrievals, characterization and error analysis, J. Quant. Spectrosc. Radiat. Transfer, 107, 385–406, 2007.

5316/9 <. . . (Rodgers 1976, 1990, 2000)>

Averaging kernels are not treated in Rodgers (1976).

Deleted this reference.

5316/9 <. . . averaging kernel profiles . . .>

unusual and never defined term, use: total column averaging kernels

Changed "The typical averaging kernel profiles" to "The typical total column averaging kernels"

5316/13-14 refer to Table 1 here for the first definition of (MW 1120)

Changed to: "For example, the averaging kernel values from the O_3 retrievals using spectral segments near 1120 cm^{-1} (MW1120 listed in Table 1) are close to 1 from 10 to 60 km for both FTSs."

5321/12-16 < For all six of the species investigated, the comparisons using daily mean values show larger differences (by up to 13%) between two FTSs than those using the individual observations recorded simultaneously. Variation in the measured total column densities during each observation day, as shown in Figs. 3, 4, and 5, arises from the temporal and spatial differences in Arctic atmospheric composition.>

I agree with the overall tendency but I do not strictly see this from Table 3: e.g., the difference is much higher for ClONO2 (17.62/4.28) than for HCl (7.84/3.22). Why?

Wouldn't another/additional explanation be a possible zenith-angle dependency of the retrieval/raytracing - could you try to check/exclude this?

Optional remark: you have a highly valuable a data set at hand. Some further *exploitation could allow a step forward in ClONO2 spectrometry. This would be of very high interest to the ground-based community: Discuss the potential reasons for the discrepancies (line shape, zero line, interfering species, resolution?) and an estimate of their relative contribution to the discrepancies observed. Have you checked these instrumental things for the two different instruments? What do you find? Did you play with different implementations of the Reisinger dual micro-window approach (fitting interfering species as profiles or only via column scaling? What is the impact on the discrepancies between the two instruments?)*

The significant discrepancies in the daily mean values between PARIS-IR and the DA8 FTS might arise from the different coverage of observation time and the different portion of the Arctic atmosphere measured by two instruments. PARIS-IR measured all of species from sunrise to sunset (because of its spectral range of 3800 cm^{-1} in each spectrum) and the DA8 FTS only measured $ClONO_2$ in the morning (since NDACC FTS filters only cover a spectral range of about 500 cm^{-1} in each measurement). In addition, as mentioned in our text, the variations of column density changed for different species. The $ClONO_2$ presented the largest variations. Please note the fact that the discrepancies in the total columns between two ground-based FTSs significantly decreased to 4.28% and 3.22% for $ClONO_2$ and HCl, respectively, and showed the same level of agreement between two species, when directly compared individual observations between the DA8 FTS and PARIS-IR instead of their daily mean values.

The different signal-to-noise ratios (SNRs) can also be a reason causing the slightly larger discrepancies in the retrieved column density of $ClONO_2$ than that of HCl. The absorption features of $ClONO_2$ and HCl are located near 780 cm^{-1} and 2725 cm^{-1}, respectively. These two spectral regions have different SNRs. The SNR of $ClONO_2$ is about 100:1 and the one for HCl is near 300:1. Yes, we did play with different implementations of the Reisinger dual micro-window approach. But these

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approaches decrease the degrees of freedom for the retrievals when fitting interfering species as profiles. The results were not desirable and were not used in this work. We also tried to investigate the zenith-angle dependency of the retrieval/raytracing. However, it is unfeasible to draw a realisable conclusion on the zenith-angle dependency of the retrieval/raytracing since the variations of solar zenith angle were small (about 1 to 2 degrees for each day) during the 2006 campaign at Eureka. In addition, all of the observations made at Eureka 2006 campaign are within a narrow range of solar zenith angle, i.e., generally between 85° and 90°.

5323/11 I suggest you cancel eq. (3) because it is a trivial and well known relation.

The equation was stated here to provide clear description since it is much easier to describe this information than using plain text.

5326/26 <The largest variation in the ratios is for ClONO2 for which the standard deviations of the mean difference are 28.3% and 14.2% for DA8 FTS and PARIS-IR, respectively. To some degree, this reflects the challenge in retrieving this molecule from the ground-based spectra.>

What do you mean by the second sentence? Please explain in some detail.

The second sentence was trying to say that the comparison between measurements from the ground-based FTSs and the observations of satellite-borne ACE-FTS were not straightforward. The difficulty arises from the following reasons: (1) the absorption features of stratospheric species in the spectra that measured from ground stations are subject to interference from the strong absorption features of other species. For example, the $ClONO_2$ spectra near 780 cm^{-1} measured at the ground have interfererence features due to H_2O, CO_2 and O_3. (2) The different viewing

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geometry between the ground-based FTSs and the satellite-borne ACE-FTS mean that the observations can not always sample the exact same portion of the atmosphere.

5326/29 <In general, the results from PARIS-IR show better agreement with ACE-FTS than the DA8 FTS results. This is most likely due to better temporal coincidences.>

I am lost here: Are you still talking about ClONO2 only at this point? I ask because the same holds true mainly for the Table 5 numbers for O3 and HCl, but it does not hold true for NO2.

This sentence begins the discussion of the comparisons of partial column densities between ground-based FTSs and the ACE-FTS for all of investigated species. We have made this clearer by making this a new paragraph. Indeed, we saw that for the all of the species investigated, PARIS-IR shows better agreement with ACE-FTS than does the DA8 FTS, both in terms of individual comparisons and the Mean Percentage Difference. To make the expression clearer, the sentence was revised to:

"In general, for the investigated atmospheric species, PARIS-IR shows better agreement with ACE-FTS than does the DA8 FTS."

5332/21 <. . . PARIS-IR and the DA8 FTS, these differences can be taken to be due to instrumental differences such as spectral resolution.>

I understand it is just an assumption that resolution effects dominate. You could easily try to verify this by artificially degrading the spectral resolution of the DA8 spectra and show whether the discrepancies decrease or remain.

The resolution effects in the column densities between PARIS-IR and two FTSs

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with higher spectral resolutions have been investigated by our previous work (Wunch et al. 2007) and were cited in this paper. Wunch et al. indicated that the PARIS-IR (MOPD = 25 cm) and University of Toronto FTS (MOPD = 50 cm) can retrieve total column densities of O_3, HCl, N_2O and CH_4 using the SFIT2 program with percent differences from the DA8 FTS at University of Toronto (MOPD = 250 cm) generally better than 4%. Total column amounts of the stratospheric species (O_3 and HCl) have larger differences than those of the tropospheric species (N_2O and CH_4). Instrument line shape (ILS) information is found to be of critical importance when retrieving total columns of stratospheric gases from the lower-resolution instruments. Retrieving EAP parameters from SFIT2 significantly improves the column comparisons of the stratospheric species for the PARIS-IR and U of T FTS. The remaining errors for stratospheric species total column amounts can be attributed to the lower sensitivity of the lower-resolution FTSs to the stratosphere.

Wunch, D., Taylor, J. R., Fu, D., Bernath, P. F., Drummond, J. R., Midwinter, C., Strong, K., and Walker, K. A.: Simultaneous ground-based observations of O_3, HCl, N_2O, and CH_4 over Toronto, Canada by three Fourier transform spectrometers with different resolutions, Atmos. Chem. Phys., 7, 1275–1292, 2007.

5341 Table 1, footnote c: <Several spectral ranges from individual spectra were used in the retrievals simultaneously> Which ones exactly were fitted simultaneously? What about ones that were not fitted simultaneously? Were the results averaged together (via some weighted mean?);

Those fitted simultaneously were marked by the lower case “c”, i.e., MW 1120 and MW 2725. We need not average the results since we have the results of each MWs from both instruments and compared them directly.

5341 Table 1, footnote c <simultaneously; also known as "multi-microwindow"

fitting.> can cancel this without loss of information

Changed to " c Several spectral ranges from individual spectra were fit simultaneously in the retrievals."

5342 Table 2, footnote c < Total Error= sqrt ((Smoothing Error)2 + (Retrieval Noise Error)2 + (Model Parameter Error)2).>

Just one optional remark: we have recently shown that an additional class of errors, namely interference errors can become significant, i.e., comparable to the smoothing error (Sussmann, R. and Borsdorff, T.: Technical note: Interference errors in infrared remote sounding of the atmosphere, Atmos. Chem. Phys., 7, 3537-3557, 2007). Implementation of this interference error quantification may be difficult at this time for all the different retrievals of your paper, but perhaps this and also the possible minimization of the interference errors might find your interest within future work.

Thank you for providing this reference. We will take it into consideration in our future studies.

5351 Figure caption: cancel <normalized> without loss of information

Deleted 'normalized'.

"Fig.1. Total column averaging kernels for PARIS-IR (blue circles) and the DA8 FTS (red squares) for 2006 Canadian Arctic ACE Validation Campaign."

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 5305, 2008.