Atmos. Chem. Phys. Discuss., 6, S6084–S6098, 2007
www.atmos-chem-phys-discuss.net/6/S6084/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simultaneous ground-based observations of O_3, HCl, N_2O, and CH_4 over Toronto, Canada by three Fourier transform spectrometers with different resolutions" *by* D. Wunch et al.

D. Wunch et al.

Received and published: 10 January 2007

Anonymous Referee #1. Specific comments:

Section 3, page 10889: "generated by FSCATM, a nonlinear forward model ... ." To my knowledge, there are no linear forward models around for the radiative transfer problem encountered here. The Lambert-Beer law introduces significant nonlinearity for all but the weakest (optically thin) spectral features. Section 3, page 10889: "... a nonlinear forward model that uses an a-priori state estimate ... ." The term a-priori refers to the retrieval process. In the context of the descrip-

tion of the forward model, I suggest to avoid this term.

This wording has been changed from “generated by FSCATM, a nonlinear forward model," to “generated by FSCATM, an atmospheric model that uses climatological VMR profile estimates..."

Page 10889: It might be advantageous to describe the forward model first and to discuss the retrieval procedure afterwards.

We have added more information about OEM to the section on SFIT2, and hopefully this clarifies this problem.

Page 10891: Please add details on gas cell used (pressure, length).

The gas cell parameters have been added to the text.

Page 10892: Concerning the set of simulated spectra, I do not fully understand which parameters generate the sample of 16 spectra for each choice of MPD. As far as I understand, noise has been added to each spectrum to generate the error bars given in Figs. 3 to 7.
However, it is not necessary to generate a manifold of spectra to determine the noise error in the retrieved column. Just generate a single noise-free synthetic spectrum and specify the assumed SNR in the retrieval process. The retrieval of this single spectrum will then yield the mean column, and the noise error covariance reported by the retrieval code can be used to calculate the noise error of the total column (this approach neglects the nonlinearity of the inversion problem, but is certainly feasible here).
Which conclusion can be drawn from the current simulation? I assume that the intention of the authors is to show that the ability to detect variability in total column decreases when MPD becomes too small. However, the outcome of the exercise is quite arbitrary, as it depends on the selection of the “same atmospheric conditions" (which are left unspecified?). If the climatological mean is

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selected here which is also used as a-priori in the retrievals, all retrievals will perfectly recover the original column. If a certain unique disturbance is chosen, the outcome might depend significantly on its assumed shape.
To make the simulation conclusive, I suggest to generate a set of spectra in accordance with the estimated variability of atmospheric conditions. The simulation would then lead to a definite conclusion: one would be enabled to specify which MPD (+ SNR combination) is required to follow the true variability of the total column to a certain extent. (Alternatively, there is a shortcut to reach this result by application of partial column sensitivities to the expected natural covariance of the trace gas profiles.)

Noise-free spectra were generated using the SZAs and ZPT profile from September 1st, 2005, adding white noise to achieve the 250 SNR. Our wording has been changed. The error bars on the columns in Figs. 3-7 are the standard deviations of the columns retrieved from the 16 spectra, and are not a direct result of the error on the spectra themselves. Our wording has been adjusted accordingly.

We have assumed throughout the paper that the total column amounts of these molecules do not change significantly throughout the day. We do not measure near dawn or dusk (mostly due to the location of our observatory), and so our simulated spectra are generated with profiles reflecting a typical difference from the a priori that we see in our daily total column averages. The point of Figs. 3-7 was to show that for a single daily column measurement, there is a significant effect from MOPD below about 50 cm, especially when ILS effects are ignored.

Section 4, general: The results from the instruments are put together and are compared, but not too much effort is undertaken to shed light on the remaining discrepancies. As an example, with respect to CH4 results it is speculated that the current spectroscopic description generates the unexpected large discrepancies. This is probably right, but can be proven to some extent: e.g. cutting down the resolution of both the TAO-FTS and the U of T FTS spectra to PARIS-IR

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resolution should allow to retrieve compatible columns from all instruments.

Cutting down the TAO-FTS OPD and resolution does result in larger and more structured residuals. However, this does not necessarily have to result in significantly different column amounts, since those could also be dependent on the ILS, which, for a truncated TAO-FTS interferogram would be much closer to the ideal than either the U of T FTS or the PARIS-IR ILS.

Page 10898: The assumed SZA dependence of retrieved columns is rather significant. For example, according to Fig. 13, the TAO-FTS results differ by about 2.4 % between the two subsets. Let us assume the tracker offset amounts half the apparent diameter of the sun. Even this quite significant deviation would generate an airmass inconsistency of only 0.4 % at 45N SZA. Given the high solar elevations of the measurements, the observed discrepancies can hardly be ascribed to an SZA error. Are there other possible explanations?

The assumed SZA dependence is significant. The solar tracker at TAO was unable to track across solar noon - it slowly drifted off the sun completely and did not recover until someone manually moved it back in place. This is why we thought it best to exclude all data with SZA of less than 40. At the very least, the system was unstable during those times, and since our goal was to measure the same air mass simultaneously, this seemed to lie outside the optimal measurement times.

Page 10898: I assume that x denotes the vector which collects the volume mixing ratios of the target gas in each model layer. Please clarify.

Yes, x denotes the vector which contains the VMR profile. This has been clarified in the text.

Page 10899: The vector rho should not be referred to as the atmospheric density or airmass. Its components are the partial columns of air molecules in each model layer.

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The vector ρ now has been better described in the text.

Fig. 18: The figure caption is not a concise description of what is shown. I assume that the graph shows the response of the retrieved total column wrt a unity perturbation of volume mixing ratio in each model layer.

The figure caption has been changed.

Fig. 1: to which wave number region these results refer to?

The figure caption has been changed.

Table 1 should also list the SNR achieved by each instrument (for typical spectra used in the analysis, in the spectral range of 2500 to 3000 cm-1).

The signal-to-noise ratios have been added to Table 3, instead of Table 1, since they are retrieved quantities.

Anonymous Referee #2. Specific comments:

Section 3 - SFIT2 and "forward model" description. The description/definition of "forward model" is confusing and inconsistent The term refers to FSCATM on p 5, but to something quite different in 3.1 on the next page. FSCATM is only part of the forward model - it is the algorithm used to calculate the ray path through the atmosphere, based on assumed pressure-temperature-concentration profiles and the solar zenith angle. The "forward model" is the procedure used to calculated the observable quantity - in this case the IR absorption spectrum - from an assumed set of input parameters. This includes not only the calculated ray path from FSCTM, but also the layered spectrum calculation, which is based on Hitran line parameters. SFIT2 is the "inverse model", which calculates the most probable set of input parameters to the forward model (including trace gas concentrations and ILS parameters) which would lead to a given observed

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spectrum.

Yes, you are correct. However, SFIT2 also contains an instrument forward model that computes the expected absorption line for a spectrum given the instrument's field of view, apodization function and ILS (if a LINEFIT tabular function or polynomial coefficients are provided). It can also retrieve the ILS coefficients as part of the inversion process. This has been clarified in the text.

Section 3 - resolution, lineshape and profile retrieval. It is not clear exactly how SFIT2 is being implemented with respect to total column retrieval vs vertical profile retrieval and ILS retrieval, and this needs to be spelled out clearly. This is important because the observed lineshapes in measured spectra are determined by both the ILS and the assumed (a priori) vertical profile, and there is significant correlation between the two effects. I am not sure from the text if the vertical profile is retrieved in ALL cases as part of the SFIT2 retrieval, even when only the total column is reported. But there is an important difference between retrieving the total column with profile retrieval and column retrieval with the profile fixed at the a priori. If the profile is fitted, how realistic is it to also fit ILS parameters (6 of them) from the same spectrum? Is there enough information in the spectra (especially for narrow stratospheric lines and/or lower resolutions where there are only a few data points to define each measured lineshape)? The use of a separate window to fit ILS parameters in the PARIS instrument (3.1, p7 column 1) is also related to this issue - how much did the ILS retrievals vary over a day, and what is the author's definition of "success" in fitting the ILS from the same or different windows. All these effects question the validity of the approach, and solid justification is needed.

SFIT2 always retrieves a profile and the column is computed from the integrated profile–this has been shown to be the preferred method of retrieving total column amounts (see, for example, Rodgers, §4.3). This has been clarified in the text. The degrees of freedom are listed in Table 3 for the case when the extra 6 parameters are

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retrieved, and these values are >1 for most microwindows (except for 2925 and 3040 cm^{-1} for PARIS-IR). But the lower number of spectral points in the microwindow is a concern, and something we are trying to show here. It is clear that for resolutions <50cm OPD, we cannot properly retrieve total columns using this method and in these spectral microwindows - there is not enough information.

PARIS-IR attempted to circumvent part of this problem by using a grid containing only 29 layers, and a separate microwindow for EAP/PHS retrievals, which did not completely solve this issue for the 2925 and 3040 cm^{-1} microwindows.

The ILS changes by <10% (2σ) over the four days of measurements for the U of T FTS and PARIS-IR.

Our definition of “success" in this case is a reduction in residual error from the standard retrieval.

The results of 3.3 suggest that fixing the ILS parameters at the LINEFIT values, and retrieving them from the spectra do give similar results, but are the profiles fitted in the exercise of 3.3? It isn't clear. (By the way, which spectrum/spectra were used in 3.3? The text does not say, but the caption to Fig 8 implies spectra of 2 Sept.)

Yes, the profiles are retrieved from SFIT2, this has been clarified in the text. The text now states that the data used were recorded on Sept. 2nd.

The “spurious oscillations in the profile" observed for CH4 may well be due to overfitting rather than lineshape parameters errors if both ILS and profile are being fitted and the inversion is under determined. Both N2O and CH4 show systematic residuals in the fitted lineshapes regardless of the ILS fitting method used, which suggests that the vertical profile is wrong. With so many free parameters to fit the observed lineshape, the residuals should be better than this.

True, there are many free parameters, and a cause of the oscillations may be in part

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due to an a priori with an unphysical shape. Our methane a priori profiles, however, are generated from climatological means, and there have been other studies that have shown that spectroscopic parameter errors can account for the problems in methane retrievals (e.g. Rothman et al., Worden et al., and Brown et al.). Methane has been identified as not having well-defined spectroscopic parameterization (see, for example `http://cfa-www.harvard.edu/hitran/HITRAN_conf06_presentations/Session2/2.2-Boudon.ppt` slide 23, Pentad intensities, noting that the differences can be as much as 20%). Current issues related to methane line mixing are being addressed by Tran et al., but have not yet been incorporated into HITRAN formatted parameters.

While there are structured residuals for N_2O as well for the U of T FTS, these residuals are much smaller (the U of T FTS average SNR for the N_2O microwindow is 600, and the average SNR for the CH_4 microwindow is 350). Oscillations are also seen in the TAO-FTS profile retrievals (where no ILS fitting is used). Oscillations are not seen in the N_2O retrieved profiles.

Overall, the presentation of this issue of lineshape and resolution effects (which is the core of the paper!) is unclear - resolution is treated in 3.2 with simulated spectra, while LINEFIT vs PHS/EAP is treated separately in 3.3 from measured spectra. It seems obvious to take the measured TAO spectra in 3.3 and reduce their resolution to those of the other two instruments and examine the effects. Was this done? It may do away with the need for 3.2 altogether. So in summary, I would like to see 3.2 and 3.3 reworked into a more coherent presentation.

"It seems obvious to take the measured TAO spectra in 3.3 and reduce their resolution..." Yes, we agree that this would be the obvious thing to do, and we have done it. However, two complications arise. The first is that the SNR of the spectra increases significantly as the TAO interferogram is truncated, masking the effects of the decrease in resolution. The second problem is that the ILS improves significantly as the TAO interferogram is truncated, also masking the effects. These problems could be solved by adding noise to the interferograms and applying an effective apodization to the inter-

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ferograms, but it seemed to us that simulating the spectra with SFIT2 would be a better choice: giving us more control over these parameters. The text has been changed to outline these problems. We also ran the same simulation, but for a SNR of 100. The difference in total column amounts retrieved from the PHS/EAP Retrieved case to the Standard Retrieval are much larger than the difference in total column amounts retrieved from the PHS/EAP Retrieved cases between the SNR 100 and SNR 250 runs. As would be expected, though, the higher SNR gives results that are closer (by $<$1% between 100 and 250 SNR) to the “truth".

4. Results The comments about tracker errors for zenith angles less than 40 degrees need justification. Generally tracker errors are more critical at low sun angles where the airmass is a stronger function of sza.

The solar tracker at TAO was unable to track across solar noon - it slowly drifted off the sun completely and did not recover until someone manually moved it back in place. This is why we thought it best to exclude all data with SZA of less than 40. At the very least, the system was unstable during those times, and since our goal was to measure the same air mass simultaneously, this seemed to lie outside the optimal measurement times.

5. Conclusions Given comments on 3.2 and 3.3 above, I am not convinced that the final conclusion is true - ie that using SFIT2 to retrieve ILS parameters as part of the state space can be used in place of an independent determination, eg with LINEFIT. Desirable it may be, but requires better justification.

This conclusion is supported by the simulations in Figs. 3-7 and the data that generated Fig. 8. While we believe that probably the best method of including ILS information in the SFIT2 retrieval is through careful characterization with LINEFIT (which has the added benefit of pointing out where alignment errors in the instrument occur), in situations where gas cell measurements are not feasible (say, balloon flights, or when reanalysing historical data), the SFIT2 PHS/EAP method is an acceptable method.

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Further points Sect. 3, p5 “Every attempt was made to ensure the TAO FTS incurred a minimal loss of signal ...". Was this realized? Is the beam from the solar tracker so large that the beams for the other two instruments could be picked off the side without blocking the main beam? If so, what is the impact on the effective solar viewing angle for the two beams which view only the edge of the solar beam?

The wording has been changed from “Every attempt was made to ensure that the TAO FTS incurred a minimal loss of signal, and that its signal-to-noise ratio was reduced by less than 10%." to, “Every attempt was made to ensure that the TAO FTS incurred a minimal loss of signal, and as a result, its signal-to-noise ratio was reduced by less than 10%."

The U of T FTS and PARIS-IR field of views were filled completely, so while there may be a small reduction of intensity due to acquiring solar radiation from the edges of the solar beam, this should be of minimal importance. The TAO suntracker significantly overfills its input mirrors, and so while the pick-off mirrors did capture some of the TAO-FTS solar beam, the effect was small.

P5. “The only difference between the three retrieval methods ...". There is only one retrieval method, but 3 cases, maybe “cases" should replace “methods".

The wording has been changed to “procedures".

3.1. Why did the authors not use a consistent version of LINEFIT to fit the ILS of the three instruments. Is there any risk of systematic differences due to LINEFIT algorithm changes between versions?

We checked for any systematic differences between lineshapes computed from LINEFIT 9 and 11 and found none. The main difference between the two programs is that LINEFIT 11 takes the temperature of the gas cell into account as part of the retrieval, whereas the temperature is fixed in LINEFIT 9.

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Section 3. last sentence before 3.1. .“... the bulk of the discrepancies can now be attributed to differences in instrument resolution." I suggest add “ ... and instrument lineshapes" since these clearly also play a large part.

Done.

P7 col 1 .“ ...from a very broad N2O band ..." should read “N2O line".

The PARIS-IR uses multiple lines in the N_2O band. The wording has been corrected accordingly.

Apart from the suggestions for 3.2 and 3.3 above, theses sections could be substantially shortened, in that the text largely duplicates what is clear from the figures and is repetitive.

The text is somewhat repetitive, however, there is more precise numerical information in the text than in the figures, so we feel that it is important to keep the text.

Anonymous Referee #3. Specific comments:

1) On section 2.2, 3rd paragraph. Please explain, why the spectra of the U of T FTS are apodized with a triangular function. Would it not be better to use the boxcar apodization and taking the ILS into account during the retrieval?

We ran a series of tests to determine whether boxcar or a triangular apodization would be best for the U of T FTS retrievals, and the triangular apodization provided better fits and more stable retrievals. Because of the poor nature of the U of T FTS modulation efficiency, our reasoning was that the triangular apodization would not significantly affect the resolution of the U of T FTS spectra, but it would reduce noise from the ends of the interferogram.

2) The aim of the paper is to compare instruments of different resolution. Please explain, why different resolutions could lead to different results, especially how

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errors in the spectroscopic line list could influence the results for different resolutions.

The resolution of an instrument affects the number of spectral points that describe a microwindow. This limits the number of vertical pieces of information that can be retrieved from the spectral line. There must, then, be a limiting resolution at which the resolution of the instrument will be too low to retrieve any information from the atmosphere from a particular microwindow. We wanted to investigate, in the five microwindows that we chose, what that limiting resolution is. This has been added to the paper.

The spectroscopic line parameter errors affect the spectral fitting procedure performed by SFIT2. Spectroscopic errors can come from errors in the line position, line intensity and line width. The last of the three might produce systematic differences in column amounts for instruments of different resolution. Line width errors manifest themselves as "W"-shaped residuals (which are clearly visible in the methane fits for the U of T FTS (i.e. Fig. 12)), but which do not necessarily cause a bias in the retrievals. Without a more detailed spectroscopic study, we cannot say definitively by how much the columns would change, and if there should be a bias seen under our specific conditions.

Methane, in particular, has been identified as not having well-defined spectroscopic parameterization (see, for example `http://cfa-www.harvard.edu/hitran/HITRAN_conf06_presentations/Session2/2.2-Boudon.ppt` slide 23, Pentad intensities, noting that the differences can be as much as 20%). Current issues related to methane line mixing are being addressed by Tran et al., but have not yet been incorporated into HITRAN formatted parameters.

3) Please define modulation efficiency and phase error and how they parameterise the ILS.

Modulation efficiency is the real part of the FFT of the instrument line shape. The phase error is the angle between the real and imaginary parts of the FFT of the instrument line shape. This has been clarified in the text.

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4) The imperfect ILS is described for the U of T FTS, but what is the reason for the misalignment of the TAO-FTS?

This is not an atypical modulation efficiency for this type of spectrometer. There is a minor misalignment of the instrument that is currently being investigated.

5) Pick-off mirrors: We assume that the Doppler shift resulting from the relative rotation of Earth-Sun system is taken into account in the retrieval. In my opinion the use of the pick-off mirrors has the consequence that different instruments look at different locations on the solar disk. If this is correct, please quantify the error resulting from the Doppler shift due to the rotation of the sun.

Our suntracker does not image the sun - it only contains flat mirrors that deflect solar radiation down into our lab, so each pick-off mirror collects light from all parts of the solar disk. Each instrument images the solar disk onto their detectors, and, as you correctly mention, any minor wavenumber shifts from off-centre detector alignment are taken account in the retrieval for each instrument individually. Only the 2775-cm^{-1} microwindow contains solar CO lines, and these are modeled individually.

6) Please define EAP and PHS. Is it possible to retrieve the ILS from these parameters? If these parameters give an indication for the stability of the ILS, I would like to see how these parameters vary over time. This would allow to check the statement that the ILS is constant over a few months for ground-based measurements. Short-term fluctuations are often important and should be quantified, if possible.

The EAP and PHS are polynomial coefficients describing the modulation efficiency and phase error. The modulation efficiencies derived from the EAP vary by less than 10% over the four days of measurements. A note has been added to the text.

7) It would be interesting to know if the authors considered recording spectra from the TAO FTS, downgraded to the resolution of the two lower-resolution in-

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struments and evaluating/comparing the results. I think that this would be very useful.

We did consider this, however, two complications arise. The first is that the SNR of the spectra increases significantly as the TAO interferogram is truncated, masking the effects of the decrease in resolution. The second problem is that the ILS improves significantly as the TAO interferogram is truncated, also masking the effects. These problems could be solved by adding noise to the interferograms and applying an effective apodization to the interferograms, but it seemed to us that simulating the spectra with SFIT2 would be a better choice: giving us more control over these parameters. The text has been changed to outline these problems.

8)Why were comparisons using the averaging kernels according to the formalism described in Rodgers and Connor, (2003), not applied here? I think that doing this would have been very useful for this study.

We did look at comparing total column amounts of the three instruments by smoothing the results with averaging kernels, as in Rodgers and Connor, 2003. This does improve the comparisons. However, in this study, we are interested in showing how well the lower resolution instruments can retrieve total column amounts of O_3, HCl, N_2O and CH_4. We compare the results to the TAO-FTS in order to compare the total column values to an instrument we deem to be closest to the “truth."

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