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Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on “Technical Note: New ground-based FTIR measurements at Ile de LaRéunion: observations, error analysis, and comparisons with independentdata” *by* C. Senten et al.

C. Senten et al.

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Final author comment

We reply to all specific comments first. In the end, we address the questions raised by Referees #2 and #1 about whether the paper deserves publication in ACP. We remind the referees of the fact that the paper has not been submitted as part of the ACE Validation Special Issue.

Answer to Referee #1 (RC S51-S52)

1) Indeed, retrieval methods, theoretical recipes for error budget evaluations etc. have been published before. However, in addition to the (short) theoretical description, we provide the details about the implementation in practice, and the resulting error estimates. We also provide a discussion of the obtained error budget results for both particular locations at Ile de La Réunion (St-Denis and Maïdo). Especially for the temperature and interfering species error it is interesting to detail the method of evaluation, because this has seldomly been published before, and never in the same conditions.

2) For the visibility of the figures showing the spectral fits, we did not explicitly mark the wavenumber ranges of the micro-window(s) on the x-axis, because of space limitations. As the referee points out, it is not practical to go back to Table 1 to get the information about the figures' x-axes. Therefore, we have now included this information in the figure captions.

Answer to Referee #2 (RC S705-S707)

1) Error characterization:

Subsequent to the referee's comment, we have made various tests to make the best evaluation of the errors due to interfering species uncertainties, taking into account the paper by Sussmann and Borsdorff (ACP, 2007) that the referee evokes. These tests have shown indeed that we underestimated the concerned errors, as we represented each interfering species by a scalar parameter. This was especially the case for molecules for which water vapour is a strong interferer. This is in line with the remarks of T. Borsdorff (see comment SC S71-S73). We therefore excluded the interfering species component from the forward model parameter error, and calculated

the error on the target gas retrieval due to each interfering species by considering the uncertainties on the interfering species' profiles.
It turned out that only H_2O and HDO have a non-negligible impact on the target species, and therefore we took only these interfering species into account in the error calculations. The resulting reduced forward model parameter error and the interfering species error are included in the revised version of the manuscript, as well as an explanation of how we calculated the error budgets.

The above also answers the comments made by T. Borsdorff, (SC S71-S73). We thank Referee #2 for having pointed out this mistake to us and T. Borsdorff for his constructive remarks in this respect.

We essentially answered the other comments in our reply S47-S50 by C. Senten et al., dated 27/01/2008.

Answer to Referee #3 (RC S33-S35)

We thank Referee #3 for his useful comments that helped us to improve the manuscript substantially.

1) In the former version of the paper the measurement error was calculated by using a diagonal noise covariance matrix, erroneously defined by the reciprocals of the square of the signal-to-noise ratio (SNR) of the fitted micro-windows of our spectra. When the SNR is very small - due to the relatively weak signal of the target molecule in a region with high noise - this approach leads to very large measurement errors for the trace gases retrieved from those micro-windows. In particular, the effective SNR for the spectral window used for the HNO_3 retrievals is about 10, whereas for example the SNR for the O_3 retrievals is about 100. Therefore the measurement noise for HNO_3 was extremely large. For C_2H_6 the same discussion holds.

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We now recalculated the measurement error by using the noise to define the diagonal elements of the noise covariance matrix, instead of the reciprocal of the SNR! This corrected approach results in smaller, more realistic measurement errors for all molecules!
We have also corrected and completed the discussion of the error budget table.

2) The measurement location is indeed very humid, i.e., the mean H_2O column amount at St-Denis during our campaigns is about 7.5x10^{22} molecules/cm^2. Comparing the interfering species error values for Maïdo with those for St-Denis confirms that the mountain site Maïdo is much less affected by the high humidity in the tropics than the near sea level site St-Denis; and even more important, it allows us to quantify this difference. The other error components are similar for both measurement locations. This extended discussion of the error budgets at both sites is now added to the revised manuscript.

3) In the former version of the paper the relative differences between ground-based FTIR and correlative partial columns were defined by $(PC_{COR} - PC_{FTIR})/PC_{FTIR} * 100$ or $(PC_{FTIR} - PC_{COR})/PC_{COR} * 100$, depending on which instrument was considered as the reference. We have now adopted the following definition for the relative difference: $2 * (PC_{COR} - PC_{FTIR})/(PC_{COR} + PC_{FTIR}) * 100$, making no distinction between both data sets. Therefore the random error on this relative difference is $4 * [PC_{COR} * PC_{FTIR}/(PC_{COR} + PC_{FTIR})^2] * \sqrt{\sigma_{COR}{}^2 + \sigma_{FTIR}{}^2}$, where σ_{COR} and σ_{FTIR} are the relative random errors (in %) on the correlative and FTIR data, respectively.

4) As mentioned in our comment S47-S50, it is not possible to directly compare our O_3 results with results found by Schneider and Hase (ACP, 2008), because their hardware (i.e., a Bruker IFS 125HR, with a photo-voltaic MCT detector, and a precise solar tracker) allows a higher SNR of the measured spectra as well as a more stable

instrument line shape, both resulting in a higher precision of the data. Also, they have a more sophisticated retrieval algorithm that allows simultaneous fitting of the atmospheric temperature profile.
On the other hand, comparing to the results found by Barret et al. (JGR, 2002), is not straightforward either, because those observations are made at the non-tropical high-altitude station of the Jungfraujoch (46°N, 8°E, 3580 m a.s.l.), in the Northern Hemisphere, strongly differing from the Réunion station as to meteorological conditions.

5) Considering both measurement campaigns, in 2002 and 2004, we have used all six NDACC optical filters, namely filters 1, 2, 3, 5 and 6 during the first campaign and filters 1, 2, 3, 4 and 6 during the second one. So during the second campaign bandpass filter 5 has been replaced by filter 4 together with the InSb detector, improving the signal-to-noise ratio in the overlapping spectral domain covered by both filters.

6) Except for CO and C_2H_6, we did not include extensive discussions about the obtained time series of the retrieved molecules, because the measurement periods are too short to make any relevant geophysical interpretation.
When considering the total column amounts of O_3, CH_4, N_2O, CO, and C_2H_6, retrieved from the St-Denis spectra, it is reassuring to see that they all agree well with the values measured with a Bruker 120M FTIR spectrometer during the ship cruise across the Atlantic Ocean onboard the German research vessel Polarstern, in October 1996 (Notholt et al., JGR, 2000). In particular, the values we used to compare with are averages from 15 to 20°S in the case of CO, and from 20 to 25°S for the other gases.
For HCl and HF, our total column amounts observed at Maïdo agree quite well with the values found by long-term FTIR measurements performed at the sub-tropical site Izaña (28°N, 16°W, 2370 m a.s.l.) from 1999 to 2003 (Schneider et al., ACP, 2005). The same conclusion can be drawn for the O_3, N_2O and CH_4 stratospheric columns.

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Finally, our HCl total column amounts agree well with the values found at Mauna Loa (19°N, 156°W, 3400 m a.s.l.), measured with a Bruker 120HR FTIR spectrometer from 1995 to 2001 (Rinsland et al., JGR, 2003).
This information is added in the revised paper.
The fact that we find few correlative data to compare with, shows again the importance of performing measurements at a location like Ile de La Réunion.
Besides the error values on the total / partial columns of the target species, we now also provide the column amounts themselves, as well as the corresponding natural variability thereon. For all species the natural variability on the total (and partial) columns exceeds the total random error, except for the upper two partial columns of O_3 at Maïdo. This implies that we can effectively extract useful information from the obtained time series.

7) We have found quite a good correlation between the total column amounts of HCl and HF at St-Denis, namely 0.78.

8) Unfortunately, only very few measurements could be made on exactly the same day at both sites, because of meteorological conditions. Consequently, correlation plots are not very informative in this case. But we do see that for the stratospheric species HCl, HF and HNO_3, the total column amounts measured on the same day at both sites coincide within the error bars, and that for O_3 there is a small offset, confirming its non-zero concentration in the boundary layer. For the tropospheric species there is a significant offset in their total columns, corresponding to their abundances in the atmospheric layer in between both observatories. Based on the differences between the column values at Maïdo and at St-Denis we managed to estimate the surface volume mixing ratios of CO, N_2O, CH_4 and C_2H_6, which are also included in the revised paper.

Arguments for publication of the paper, with reference to the ACP evaluation guidelines cf. Referee #2 comments (RC S115-S117)

1) Does the paper address relevant scientific questions within the scope of ACP?

Scope of ACP:

"Atmospheric Chemistry and Physics (ACP) is an international scientific journal dedicated to the publication and public discussion of high quality studies investigating the Earth's atmosphere and the underlying chemical and physical processes. It covers the altitude range from the land and ocean surface up to the turbopause, including the troposphere, stratosphere and mesosphere.

The main subject areas comprise atmospheric modelling, field measurements, remote sensing, and laboratory studies of gases, aerosols, clouds and precipitation, isotopes, radiation, dynamics, biosphere interactions, and hydrosphere interactions (for details see Journal Subject Areas). The journal scope is focused on studies with general implications for atmospheric science rather than investigations which are primarily of local interest."

The present paper addresses remote sensing measurements of the Earth atmosphere, in particular of the tropo- and stratosphere, for the first time using FTIR solar absorption measurements at a tropical Southern Hemisphere site, Ile de La Réunion. The challenge is that this technique is very vulnerable to high local humidity, as is the case at a site like St-Denis. The purpose is to demonstrate that, despite the high local humidity, the FTIR technique is still capable of measuring a vast number of tropospheric and stratospheric species, with a precision that allows the detection of the atmospheric variability. The latter aspect is proven by a full error budget analysis.

It is also demonstrated that the technique is capable of providing some information about the vertical distribution of some of the species.
Ile de La Réunion is a complementary NDACC station: a dedicated infrastructure is being built at Maïdo. Therefore the paper also highlights the differences between the measurement capabilities at Maïdo (2200 m a.s.l.) and at the near sea level site St-Denis. These measurements are of great interest for filling a gap in the ground-based observation network in order to address satellite and model validation on a global scale.

2) Does the paper present novel concepts, ideas, tools, or data?

New data are presented. Also, to our knowledge, a detailed discussion of a full error budget estimation for ground-based FTIR remote sensing has not been published thus far. The paper by Kagawa et al. (ACP, 7, 3791-3810, 2007) is similar to ours in its concept, but its error budget evaluation is less complete. No new scientific ideas are presented; therefore the paper has been proposed as a Technical Note.

3) Are substantial conclusions reached?

As said above, our conclusions are that FTIR solar absorption spectrometry is feasible at tropical locations, even at sea level. Higher precision measurements can be performed at higher altitude, with the disadvantage that the boundary layer is not sampled, and therefore, that the measurements are less sensitive to local sources.
The precision of the measurements allows the detection of atmospheric variabilities at both sites.

Another conclusion that has been drawn from the limited time series is that the atmospheric composition above Ile de La Réunion is impacted by biomass burning on the African continent and Madagascar, especially in the months September and October. This has been confirmed by FLEXPART simulations.

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4) Are the scientific methods and assumptions valid and clearly outlined?

We have given all technical details that allow verification our results. An underestimation of the error due to interfering species uncertainties that was present in the ACPD version of the paper and that was outlined by Referee #2 and T. Borsdorff, has been corrected in the revised version. We thank both persons for having pointed this out to us.
Else, the validity of our method has not been questioned by the referees.

5) Are the results sufficient to support the interpretations and conclusions?

We believe that the answer to this question is yes, without any doubt.

6) Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists?

As said under 4), we have given all technical details that allow verification of our results.

7) Do the authors give proper credit to related work and clearly indicate their own new / original contribution?

In the ACPD version we forgot to take into account the publication by Sussmann and Borsdorff (ACP, 2007) regarding the interfering species error. This has been corrected for in the revised version. Additional references have been added, to better place the results in the context of other observations.

8) Does the title clearly reflect the contents of the paper?

None of the referees has made any comments in this respect. The title stresses that this is a technical paper.

9) Does the abstract provide a concise and complete summary?

None of the referees has made any comments in this respect. Still, we reformulated the abstract to reflect the revised contents.

10) Is the overall presentation well structured and clear?

11) Is the language fluent and precise?

12) Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

13) Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

14) Are the number and quality of references appropriate?

None of the referees has made any comments regarding questions 10 to 14, except for one remark concerning wavenumber identifications on the x-axes of the figures showing spectral fits. This is taken care of in the revision.

15) Is the amount and quality of supplementary material appropriate?

This is not applicable to this paper.

In conclusion, we believe that we comply with all evaluation criteria and therefore recommend publication of this paper in ACP.
We also remind the editor of the fact that all three short comments (SC S113-S114 by K. Hocke, SC S549-S550 by D. Feist, and SC S697-S698 by T. von Clarmann) and Referee #3 support publication of the paper.
Moreover, it is important to note that some of the FTIR data that are presented in this paper are used for the validation of ACE and are therefore mentioned (but not discussed) in the corresponding ACE validation papers (ACP - Special Issue: Validation results for the Atmospheric Chemistry Experiment (ACE), Editors: A. Richter and T. Wagner, Atmos. Chem. Phys., 2008). This does not preclude a full discussion of the characteristics of the data in a dedicated paper. It is very useful that, when using data for a specific application like satellite validation, a reference can be made to an original paper that provides all technical details about these data, which cannot be included in the application paper. Therefore, this is in no sense a double publication!

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 827, 2008.