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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of tropospheric and stratospheric ozone trends over Western Europe from ground-based FTIR network observations" *by* C. Vigouroux et al.

C. Vigouroux et al.

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Response to Referee Comments

The authors thank the referees for their helpful and constructive comments and questions. The manuscript has been clarified in many points. Therefore, the referees have contributed to a substantial improvement of the paper.

1. Referee 1

A. Main comments

Comments 1) to 3)

A general comment of the referee is that the analysis procedures are not "that consis-

tent"

The referee is right in a sense that the retrieval parameters are not identical at all sites. But one should consider that each individual station has its own limitations (the water vapour has a stronger influence at low altitudes sites, the ozone amounts and variabilities are different at different latitudes,...), thus it is not possible that a completely identical set of parameters leads to an optimal strategy at all the sites simultaneously. One could have imposed a single set of parameters that gives reasonable results at all sites - even if not optimal. This approach is considered for the future data archiving in the FTIR community in NDACC but has not been implemented yet during the UFTIR project, in which each partner optimized the retrieval parameters for his site. Only the parameters that are essential if one wants to avoid systematic biases between the various stations results have been homogenized: these are the retrieval micro-windows and the spectroscopic database. Only the Zugspitze station has chosen to shorten the micro-window but we do not expect that this would induce any systematic bias compared to the other stations. This has been clarified in the new manuscript, and some information has been added concerning the specific referee comments below.

Specific comment on interfering species

At all sites, the absorptions of all interfering species that appear in the micro-windows are included in the simulated spectra. Thus, the absorption lines due to CO2, C2H4, and O3 isotopes are also calculated at the stations where they are not retrieved. The only difference is that at these stations, the profiles of these molecules are fixed to their a priori profiles. The reason for that is that the absorption of the interfering gas can be so small that a simultaneous retrieval of the interfering species profile or column does not improve the target gas retrieval. The significance of the interfering species absorptions and associated spectral fit residuals vary from one station to another. Therefore again, it has been left to each individual PI to judge the usefulness of retrieving all interfering species in the retrieval optimization process.

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Specific comment on failure of water vapour retrievals at Harestua

Water vapour is indeed a species which is difficult to retrieve due to its profile shape (the volume mixing ratio values show very large gradients with altitude in the troposphere, with maximum values at the surface) and its high variability. Because of these high gradients of water vapour with altitude, the three lower altitude sites have more difficulties to retrieve water vapour. Each of these stations has implemented its own best solution: at Ny-Alesund H2O profiles are not retrieved but daily sonde data are used for the H2O a priori profiles, and at Kiruna, independent retrievals of the water vapour profiles (in separate micro-windows, before carrying out the target gas retrieval) are made using PROFFIT9 which makes the retrievals of H2O on a logarithmic scale thereby improving the retrieval quality (Schneider, M., F. Hase, T. Blumenstock: Water vapour profiles by ground-based FTIR spectroscopy: study for an optimised retrieval and its validation, ACP, Vol. 6, 811-830, 2006). At Harestua, none of these solutions could be implemented.

Specific comment on the choice of Sa at Zugspitze: "Why did the Zugspitze not use a Sa of 10%? Why different correlation lengths? Given access to climatological data, these lengths can also be estimated. Is this what was done for each station or something more arbitrary? "

As said above, there are enough differences between the stations (location, altitude) to justify that the regularization parameters, such as the a priori constraint, is let to each partner's appreciation. At Zugspitze, it was chosen to use the information coming from the CTM2 model to construct Sa, rather than a Sa with a constant 10% value on the diagonal as was done at all other stations. As CTM2 is a tropospheric model, the Zugspitze approach has the disadvantage that the Sa values for altitudes above the model altitude domain have to be chosen somehow. In this case, they have been decreased smoothly from 8% at about 27 km to zero at 70 km. These lower values at high altitudes explain to a large extent the smaller DOFS of 0.5 at Zugspitze compared to the other stations (1.0 to 1.2). The correlation lengths can indeed be estimated

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from climatological data. They are not constant within the whole altitude range of the retrievals (usually between 3 and 6 km). But since the SFIT2 algorithm allows only one correlation length, a typical value has been used. At Ny-Alesund and Kiruna, unrealistic values were used (a larger value of 8 km and no correlation, respectively), since this parameter can also be used as a regularization one, as the Sa matrix itself, and the effective SNR value (see answer to question 4) below).

4) On what basis were the SNR values chosen? Does this reflect the inherent noise in the spectra? The SNR values given in Table 2 are effective values, used in the retrievals. These are the values also from which S_ϵ (Eq. (2) of the paper) is constructed. They usually do not represent the inherent noise in the spectra, but are smaller. This is linked to the fact that the residuals in a spectral fit are not only coming from pure measurement noise but also from uncertainties in the spectroscopic line intensities or pressure broadening coefficients,... Therefore, if one uses too high SNR values, the retrieval process will try to retrieve information from this generalised "noise". Thus the effective SNR is used as a regularization parameter (together with Sa): larger values give larger DOFS, retrieved profiles closer to the real one, and the residuals (observed spectrum - calculated spectrum) should decrease. If the effective SNR is too high, the residuals will not decrease anymore and the retrieved profiles may start to oscillate. On the contrary, the real SNR, corresponding to inherent noise in the spectra, is used for the noise error calculation in part 2.4.1.

5) Neglecting phase error... The manuscript has been changed according to your remark.

6) Independent layering

Your remark has been taken into account in the new manuscript.

7) Total random error comparisons with Kagawa et al.

Indeed, it was not possible to compare both error budgets. In the new manuscript, we

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simply removed this reference, but also Schneider et al., 2005, and compared our total random error only with Barret et al., 2002.

8) Section 3.1.2: Why the Dobson at Harestua is so different to the one at Ny-Alesund?

Since the publication of the present paper in ACPD, we discussed the bias seen at Ny-Alesund between FTIR and UV-Vis comparisons with the UV-Vis data provider, F. Wittrock. He confirmed that his current data in the NDACC database have a bias 6 to 12% smaller depending on solar zenith angle. A new analysis has been made which solves this problem of bias, but the new data set are not yet available to be used in the present paper. Then, the only non understood biases are at Harestua. The fact that approximately the same bias is seen with both UV-Vis and Dobson comparisons suggests that the bias comes from the FTIR data, and that all Dobson data are consistent. We have added the information from F. Wittrock in Sect. 3.1.2 to clarify the discussion on biases.

9) Section 3.2.1: What does the value of 2*1.E 17 molec.cm-2 exactly explain?

We have given the mean value of the ozone amount in the 0-3 km layer at Kiruna, to illustrate that the differences in tropospheric ozone amounts between high-latitude and mid-latitude stations seen in Fig. 8, are also due to the different altitudes of the sites, and not only due to the different latitudes (on the contrary of the stratospheric partial columns amounts). As the partial columns are given in molec.cm-2 in Fig. 8, we thought that it was clearer to give the value of the 0-3 km layer ozone amount also in molec.cm-2. Looking at Fig. 8, we can then easily see that about half of the difference between high-latitude and mid-latitude stations, which is approximately 4*1 E 17 molec.cm-2, comes from the fact that Jungfraujoch, Zugspitze and Izana are high altitude sites. This has been clarified in the new manuscript.

10) Section 4.1: Careful with the numbers and descriptions.

The corrections have been made in the new manuscript.

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11) Differences between Zugspitze and Jungfraujoch? Why the DOFS (0.5 at Zugspitze) are so different? Are the authors saying that the Jungfraujoch did not see the low O3 columns in the 2003/2004 and 2004/2005 winters?

We have already answered the question of the smaller DOFS at Zugspitze in question 3). The referee is right that this smaller DOFS can not be responsible for the different trends since its impact would be in the opposite direction, i.e., giving a zero trend as in Jungfraujoch, and not a significant negative one.

We have indeed noticed that for about 25 days out of the 35 days when lowest values were seen at Zugspitze (in winters 2002/2003 and 2003/2004 only because the Zugspitze time-series ends in September 2004), there are no data at Jungfraujoch, probably due to bad weather conditions. The impact hereof on the trend is large because the Zugspitze time-series contains less data during the first years of measurements (especially in winters). The fact that there are fewer data at Zugspitze than at Jungfraujoch is reflected in the uncertainties on the trends in Table 6 (Table 7 in the ACP manuscript): they are almost twice as large at Zugspitze than at Jungfraujoch for the layer 27-42 km. To clarify the discussion, we have added a plot of the partial columns time-series of Zugspitze compared to the Jungfraujoch one, together with the fitted values from the bootstrap resampling method.

12) Kiruna and Harestua trends in the 18-27 km layer. The sondes results by Kivi et al., have a lower range of roughly 22 km and not 18 km. It is noted that in the lower stratosphere the trends from the FTIR data are lower which actually might make the matter worse.

It is indeed interesting to note the altitude differences in the compared trends (Kivi and present work). We did the exercise to calculate the FTIR trends at Kiruna for the 20.8-29.14 km layer (which corresponds at Kiruna to the 40-10 hPa layer, and gives us a DOFS of 1.04), and we obtain a trend of 0.54+/-0.55%. Thus the trends correspond better to the non significant trends given in Kivi et al. The reason is that the positive

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trend is situated in the 16-22 km layer (0.70+/-0.55%, DOFS=1.1). This is confirmed by the trend in the 10-22 km layer, of 0.49+/-0.58%, which is then increasing compared to the 10-18 km layer, and is closer to the one of Kivi et al. for these altitudes, even if it is still non significant in our case. The same behavior is observed at Harestua: the trend for the 22-29 km layer is smaller than for the 18-27 km layer, but in this case, it remains significant. One could argue that the partial columns limits could have been chosen to coincide with the one of Kivi et al., but we gave the priority to a homogeneous set of partial columns that gives a DOFS of about 1.0 at each station. Since the question of the referee leads to an interesting observation, we have added this result in the new manuscript.

Concerning the next question "Were the nearby sondes at Kiruna and Harestua used in the Kivi et al. study? It is possible to look at trends just from those sondes? (same question 13), for the troposphere at Harestua)"

The stations Sodankylä and Ny-Alesund are included in the study of Kivi et al., but not Gardermoen. They used seven stations together for their trends calculations to enhance the number of data, which is necessary for enabling seasonal studies for example. One could of course look at the trends just for individual sonde stations as we did for FTIR data. But we think this is beyond the scope of our paper to derive physical information ourselves from external databases. We would be happy to have the information from each group, as we got from A. Redondas at Izana.

13) What model input does the CTM2 model use in the simulations at Harestua?

The model uses pre-calculated meteorological IFS (Integrated Forecast System) data from ECMWF for selected years. Emission of source gases for different source categories are based on the RETRO and POET projects. The EDGAR data are used for anthropogenic emissions, the GEIA data for soil emissions, and the Müller (1992) data for other natural emissions (see references below), but taking into account the time variation between 1990 and 2001.

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EDGAR version 3.2: http://www.mnp.nl/edgar, based on: Olivier, J.G.J. and J. J. M. Berdowski, Global emission sources and sinks, In Berdowski, J., Guicherit, A. and P. J. Heij (eds.) "The climate system, pp. 33-78 A. A. Balkena Publishers/ Swets Zeihtlinger Publishers, Lisse, The Netherlands, ISBN 90 5809 2550, 2001.

GEIA90: http://www.geiacenter.org, based on: J.J. Yienger and H. Levy II, Empirical model of global soil-biogenic Nox emissions, J. Geophys. Res., 100, 11447-11464, 1995.

Müller, 1992: scaled to values agreed upon in the POET project. Müller, J.-F., Geographical distribution and seasonal variation of surface emissions and deposition velocities of atmospheric trace gases, J. Geophys. Res., 97 (D4), 3787-3804, 1992.

Within the time-frame of the UFTIR project, the model partners did not have time to make sensitivity tests to understand the causes of this negative trend in the troposphere.

B. Minor changes

We thank the referee for his/her careful reading of the paper. All corrections have been made in the revised manuscript.

2. Referee 2

2.1 Major comments

1) Errors: "What aspect of the measurement process can give an uncertainty of >15% at 23 km and <5% at 20 km (for example)? Is this resolution the best for representing the uncertainty variation with altitude?" ..."It would be an advantage to list [the partial columns errors] in a table"

The structure in the errors as function of altitude (Figs. 3 and 4 of ACPD) is correct, even if it looks unrealistic. It reflects the shape of the leading eigenvectors of the retrieval. In some altitude regions it tends to wiggle more strongly in response to a

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disturbance applied in spectral domain. In such an altitude region, the error bar tends to be larger. But we agree with the referee that giving only Fig. 3 and 4 is not very appropriate in our paper where only partial columns are discussed. Thus, we have kept Fig. 3 to illustrate the effect of the retrieval process on the errors profiles, but we have replaced Fig. 4 by a table where the smoothing error, the combined noise and model parameters error, the total random error and the natural variability are given for each partial column in the typical case of Jungfraujoch.

Furthermore, in the ACPD version of the paper, we had suggested that the random error budget could have been overestimated (Sect. 2.4.3 and 3.1.2). A new error calculation has been made that leads indeed to a reduced error budget. Thus, the values for total error and for the random error on the comparisons in Tables 4 and 5 (Tables 5 and 6 in the ACP manuscript) have been changed in the new manuscript.

2) Bootstrap resampling method

The objective of the bootstrap analysis approach is to determine the annual trend and associated uncertainties, based on a statistical model that makes minimal assumptions about uncertainty distributions associated with the raw data. The purpose of the Fourier series is to capture the regular intra-annual variations in a statistically robust manner without having to make detailed assumptions about the nature of the intra-annual behaviour. The year-to-year variability highlighted by the referee will not be captured by the intra-annual Fourier series, but is effectively a noise source in the trend determination, and therefore feeds into the uncertainties in the determined trends So, whilst the referee correctly notes that different sites will show different variability, with the Arctic sites typically showing greater year-to-year variation, the use of the same Fourier series order ensures statistical consistency in the ozone trends and uncertainties determined for the different sites. A 3rd order Fourier series was selected to match that used in the trend analyses carried out by Gardiner et al. (2007) to give both internal consistency in the ozone results, and external consistency with the results given in that paper. The 3rd order Fourier series was selected by looking how the trend uncertainty

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varied for different orders (from 1 to 5) and by choosing the order that provides the best overall result for all species. Generally, the variation we get in the trend and uncertainty values for different series is significantly smaller than the basic uncertainty in the trend. Thus we think that, since the variability effects the referee talks about are taken up in the trends uncertainty values given in Table 6 (Table 7 in the ACP manuscript), there is no real benefit in producing a separate table of the fit parameters. However, this is indeed an important point and it has been clarified in the introduction of Sect. 4 in the new manuscript.

Furthermore, we have added a figure in Sect. 4 to illustrate how the function fits to the data, as suggested by the referee.

Remark: We have modified the starting year of Ny-Alesund time series because it was the only station starting at 1994, and only one data was available. Thus, we simply start the series in 1995.

2.2 Minor comments

All suggested corrections have been taken into account in the revised version of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 5007, 2008.