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Interactive Comment

Interactive comment on "Trends of ozone total columns and vertical distribution from FTIR observations at 8 NDACC stations around the globe" *by* C. Vigouroux et al.

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Answers to Referee3

We thank the referee for his/her constructive remarks and suggestions. We answer below to the specific comments.

The phrase 'self-calibrated' is used in both the introduction and conclusions. Although the optical absorption due to ozone is measured with reference to the surrounding continuum there are a number of steps from this to derive a vertical profile of ozone, many of which are not 'self-calibrating'. The measurements

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made by the different sites are linked to a common spectroscopic database, but since different absorption lines are used, the absolute accuracy of the actual spectral parameters used could be different; the analysis requires PT profiles and any errors on these will affect the results; and changes/differences in instrumental performance (e.g. effective resolution, phase, etc.) could affect profile results. Significantly more justification is therefore needed before the data can be said to be 'self-calibrated'.

The referee is right. We used the term "self-calibrated" because the ozone columns measurements are repeatable and stable over time (if the same spectroscopic parameters is used, or model parameters in general), i.e., the technique of measurement itself would not change the ozone column that would be derived from two measurements with the same ozone amount in the atmosphere. However, for long-term series when the ILS can possibly change between the measurements, this indeed would not be true. Therefore, we have added in the Introduction and Conclusion Sections that a careful treatment of the ILS is required to obtain reliable ozone values and trends. This is especially true for the partial columns, the impact of the ILS being smaller on total columns.

Concerning the common spectroscopic database, but different absorption lines: the spectroscopic parameters have an impact on the systematic uncertainty of individual measurements. Therefore, if a bias can be obtained in the ozone amounts at one station by using slightly different or additional micro-windows, it would be systematic for the long-term series and therefore it would not imply different trend results. But, indeed, if there is a drift in the pT profiles, this could influence the trends differently in case of different micro-windows used. We have made the test of using at Kiruna the same 1000-1005 cm-1 as at other stations. The trends are very similar than with the settings used in the paper (Trop: -1.1 $\pm$2.5 %/dec. (instead of: -0.9 $\pm$ 2.5); LowS: -3.4 $\pm$2.5 (-3.9 $\pm$ 2.6); MidS: -0.0 $\pm$2.4 (+0.4 $\pm$ 2.6); UppS: +6.6 $\pm$3.1 (+7.4 $\pm$ 3.4); TotCol: -0.3 $\pm$1.5 (-0.3 $\pm$ 1.6). At Ny-Alesund, the approach of using only the 1000-1005 cm-1 window has also been tested, with again a little change on the trends well below the

uncertainty on the trends (1.4 %/dec. impact on Trop trend, and less than 0.7 %/dec. for the other partial/total columns). The conclusions on these tests have been added in the new manuscript in the Sect.2.2 where the impact on the different parameters on trends are more discussed (according to the Referee's next comment).
Concerning the possible errors on the pT profiles: the random uncertainties due to the temperature are taken into account in the uncertainty budget. If the NCEP temperature data contain a drift of 1%/decade, which seems not excluded from e.g. the analysis of D. Hubert on the impact of NCEP pT on SAGE-II v6.2 ozone drift (Hubert et al., in preparation for AMT), our uncertainties on single station trends of about 2%/decade would increase up to only 2.2%/decade if this systematic temperature component would be included (Daan Hubert, personal communication).

Section 2.2 describes the FTIR retrieval strategy, and one common theme is that there are very few aspect of the retrieval that are common to all groups. While the differences are acknowledged there is very little discussion of the reason for the differences or the potential influence these differences could have (either on the absolute values of the ozone data or on the trends derived). Without further discussion on this point it is difficult to know how much reliance can be placed on the differences in the results from difference sites being due to the atmosphere and how much to the differences in the analysis strategies. Some further analysis that actually assessed some of the implications of the different strategies would significantly enhance the robustness of the results and conclusions.

We have added more discussion in Sect.2.2 on the reason for the differences and on the influence of the differences on the trends. The two parameters that could have a significant impact on the trends is the micro-windows, if the pT NCEP profiles contain long-term drift; and the treatment of ILS. For the former, the test has been made at Kiruna and Ny-Alesund to use the 1000-1005 cm-1 window, and it has been found that this does not impact the trends significantly (see previous comment). Due to the small impact of this parameter, the time-series analyses have not been updated with a

unique choice of micro-window for the present paper, but this is a parameter which will be easily homogenized in future work.
For the ILS treatment, the individual choices were led by the type of spectrometer and the availability of cell measurements: Bruker 120M and Bomem are not stable enough to allow to use a fixed ideal ILS. Therefore, the LINEFIT results were used at stations where the HBr cell measurements were made (so not possible for the old Bomem spectra). At Jungfraujoch, the cell measurements started only in the early 2000's, so to stay homogeneous they used a fitted eap parameter for the whole time-series. We have tested the impact of fitting the eap instead of using an fixed ideal ILS at Ny-Alesund, and again little impact have been found on the trends (less than 0.6%/decade for all layers).

The determination of the ILS (pg 24630) is obviously important, particularly in the profile retrieval and long-term changes/drifts in the ILS could presumably map onto the trend results. There is a description of the ILS procedures followed but several times results are referred to as being 'close to' the ideal and therefore assumed to be ideal. It is important to know what is the definition of 'close to' is in each of these cases and how this criteria was selected. It would also be useful to know how often the ILS checks are done as this would cover the issue of potential long-term alignment drift.

We have highlighted more the importance of ILS in the new manuscript (Introduction, Sec. 2.2 and Conclusion). The discussion about the ILS treatment in Sect 2.2. has been updated to provide more information (also see the response to the Referee1 comment on Wollongong time-series). The ILS was assumed to be ideal if the loss of modulation efficiency at maximum OPD is below 2%. The frequency of the ILS checks is at least every 6 months for the Bruker 120/125 HR instruments used in the present study. But it can be more often for other stations (e.g. at Izaña, see García et al., 2012). We have clarified the reason for the different choices of ILS and we have made the test at Ny-Alesund to change the treatment of ILS (see previous comment).

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It would be useful to have further details on the sensitivity in Section 2.3. Although fig 1 shows the sensitivity profile for the Jungfraujoch station, it would be interesting to know something about the overall sensitivity for each of the four altitude layers for each station being analysed as this would indicate the potential influence of trends in the a priori data on the analysis.

We have added a plot for the Izaña station, since the layer limits are slightly different for this station, and since the calculated sensitivity is different because of the use of Tikhonov regularization instead of optimal estimation. We do not want to add plots for all stations to minimize number of pages / plots. But we have added a sentence in the manuscript: “Similar averaging kernels are obtained at each station (not shown).” For all stations (so even for Izaña and Kiruna that are using Tikhonov regularization), the DOFS above 49 km is small (from 0.006 for Jungfraujoch to 0.04 for Izaña).

Specific comments :

Page 24626 Line 14 : ‘stable data are needed’ – it is not the data that needs to be stable. Suggest replace with ‘reliable data from stable instruments are needed’.

Done.

Page 24626 Line 16. Do ozonesondes count as ‘ ground-based’ or are they ‘in-situ’ measurements ?

The idea was to oppose “ground-based” to satellite, but it is of course “technically” not correct. So we changed the text. “Ground-based (Dobson, Umkehr) and ozonesondes data are traditionally used for these studies…”

Page 24628 Line 19. Should it be ‘single scaling’ or ‘simple scaling’ ? ‘apriori’ is missing a space and should it be in italics (throughout document) ?

Indeed, we meant “simple”: changed. We prefer to let “a priori” in normal text, it is commonly used now.

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Page 24629 Line 17. This assumption implies there is no correlation from the measurement noise in the vertical information. This seems a large assumption to make without any further justification. Some more discussion on this point would be useful.

The measurement noise matrix S_ϵ (dimension: m x m, m being the number of points in the discretized measured spectrum y) is assumed diagonal meaning that we expect no correlation between the noise at different wavelengths (which is indeed an approximation but it is quite common, Rodgers 2000). Then, the measurement noise error matrix S_n is calculated with $S_n = GS_\epsilon G^t$, with G the gain matrix= $\delta x/\delta y$, x the retrieved state, y the measurement. So the measurement noise error S_n is not diagonal, meaning that there is indeed some correlation in the measurement noise error between altitude layers.

Page 24631 Line 12. If the sensitivity is the fraction from the measurement rather than the a priori, how can it be greater than 1 (see Fig 1) ?

The sensitivity is not "mathematically" the fraction between measurement and a priori. We have change the sentence to "the sensitivity . . . represents roughly the fraction of the retrieval that comes from the measurement rather than from the a priori information". The sensitivity is good (information is coming from the measurement) when it is close to 1 (Rodgers, 2000). It can happen (Rodgers, 2000) that it is greater than 1 (area of the averaging kernel at this altitude is greater than 1), showing that at this altitude the retrieved profile might be too sensitive to a change on the true state.

Page 24631 Line 21. The sensitivity shown in Fig 1 is >0 at 49 km, so what was the actual cut-off criteria ?

Again "goes to zero" was a language approximation, we meant "becomes negligible", not strictly zero. For the upper layer, we could use, as an upper limit, the last layer of the retrieval grid, which is 100 or 120 km, depending on the station. Having a layer 29-100 km, may be misleading: one could think that FTIR measurements are sensitive to ozone changes up to 100 km. We have decided to take a cut-off criteria above which the DOFS are very small. In the optimal estimation method of regularization, this coincide with a sensitivity close to zero. However, as shown in the new plot of the Izaña averaging kernels, the sensitivity when Tikhonov regularization is used does not decrease to zero with height, as the DOFS does. So in our case, the 49 km cut-off, coincide with a DOFS above this altitude that are between 0.006 (for Jungfraujoch) to 0.04 (for Izaña). The precise values (49 instead of 50 km; 29 instead of 30 km), are due to the retrieval grids of the stations. Each station has between 44 and 47 layers of varying widths (more layers in the troposphere than in the upper stratosphere), and we have chosen the limits of the 4 partial columns to coincide with some selected limits (based in DOFS) of the stations grids, to avoid interpolations.

Page 24632 Line 7. 'UV-VIS' rather than 'UV-VIs'.

Done.

Page 24632 Line 22. Suggest 'variable' rather than 'contrasted'.

Done.

Page 24633 Line 16. Parameter A0 is not defined.

Done.

Page 24633 Line 18 (and eq 2) should it be "(t) rather than just " ?

Done.

Page 24634 Line 22 Clarify which ones 'those proxies' refers to.

Done.

Page 24639 Line 17. As the total column results as also given in DU in section 4.2.5 it would be good to also do so here.

Done.

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Page 24645 Line 28. Does 8 sites constitute 'many' for a global network ? Suggest replace with the actual number.

Done. Note that only 8 stations have contributed to the present paper. However, more stations could contribute after re-analysis of their time-series, using the described retrieval strategies (Arrival Heights, Rikubetsu, Eureka, Bremen). Some were suffering from gaps in the measurements but could be used in the future when/if more years of data will become available (Mauna Loa, Rikubetsu). We have added this information in the manuscript.

Page 24646 Line 4. Suggest replace 'proposed' with 'demonstrated'.

Done.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24623, 2014.