

Interactive comment on “A new method to detect long term trends of methane (CH₄) and nitrous oxide (N₂O) total columns measured within the NDACC ground-based high resolution solar FTIR network” by J. Angelbratt et al.

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Received and published: 10 June 2011

Repy to Referee # 1

This paper analyses long term trends of total column CH₄ and N₂O at four European sites in the Network for Detection of Atmospheric Composition Change (NDACC): Kiruna, Harestua, Zugspitze and Jungfraujoch. The novelty of the analysis lies in including terms proportional to anomalies in “atmospheric parameters” such as surface pressure, tropopause height, CO column and HF column as well as linear trend and

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annual cycle in the least squares fit to the data. These anomalies are themselves determined as the residuals of a polynomial plus seasonal cycle fit to the atmospheric parameter time series. The anomaly terms are expected to reduce the residuals of the trendline fit if there is any systematic dependence of the CH₄ and N₂O columns on the atmospheric parameters. This is indeed observed when the fits are compared to the usual methods of fitting of trend and seasonal cycle only, or the bootstrap method of Gardiner et al. The trends determined by this multiple regression method are little different from those of the other methods – however I find this unsurprising since essentially the same trend function is fitted in all cases, and the additional regression terms only serve to reduce the residuals.

Reply: The reviewer claims that the difference in trends between the multiple and the other approaches is small. This is not true. For example: Zugspitze CH₄ show a trend of 0.13%/year with the multiple and 0.09%/year with the Bootstrap approach, this is a 30% difference. Harestua N₂O shows 0.40%/year vs. 0.45%/year, this is a ~ 13% difference.

If the additional atmospheric parameter anomalies themselves are roughly randomly distributed over time, as they most likely are, then we can expect to retrieve a similar trend, but with reduced uncertainty. Thus the impact of the new approach is limited.

Reply: We think that the anomalies are not randomly distribution over time since:

a) The measurements, for many FTIR sites, are not equally distributed over time. Some sites have more measurements in the winter than in the summer and this will give an uneven effect on the residuals when fitting a trend.

b) Certain processes exists only certain years a random number of times. For instance the amount of stratospheric subsidence, indicated by HF, varies depending on if it is a cold stratospheric year and if the polar vortex happens to be above a given site. The same applies for CO, since fires occur in summer, but only certain dry years.

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The only parameter that may be randomly distributed over time is pressure.

General comments

The atmospheric parameters considered include surface pressure, tropopause height, and total columns of HF, CO and ethane. Firstly I would like to see all these parameters tabulated, with their impacts, for a clearer overview – Fig 2 covers only those found to be significant, but the reader must take the authors' word for it.

Reply: This was considered in the first draft of the manuscript and a table like this is added to the article.

It is also not clear exactly how the impacts of the anomalies are defined and calculated (p 8220, line 1 et seq. and Figure 2). For example it is not clear to me what the difference is between the linear trend anomaly in Fig 2 and the trends determined for the eventual fits (Table 5), but they are quite different. In fact I do not understand what the linear trend anomaly in Fig 2 is at all – this should be clarified.

Reply: An updated figure2 is added with a more detailed description of how the impacts are calculated. The impact from the linear trend is removed to avoid misunderstandings with the trends presented in table 5.

I do not think the surface pressure should be considered as an anomaly at all – it is clear that to first order the total column should scale linearly with surface pressure, and we can expect that surface pressure is randomly distributed around the station mean. It would be better to correct all columns to the same surface pressure, such as the station mean, to remove this well-understood dependence before the fit. If there is still a significant dependence on surface pressure, it must then have another physical basis.

Reply: The authors do not agree. First, the developed trend method is supposed to be general and work for most of the gases measured with the G-B solar FTIR technique. For species where the volume mixing ratio profile not is constant with

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height the pressure dependence is no longer linear as in the methane and nitrous oxide case. Secondly, we have actually tested the reviewer suggestion on the data from the Harestua site. The difference is very small when comparing the two methods $0.249 \pm 0.0151\%/year$ and $0.252 \pm 0.0153\%/year$ for the normalization-method and anomaly-method, respectively. Thirdly, it is an advantage to include pressure as anomaly since this makes it possible to quantify its contribution to the reduction in overall variability at a given site.

The authors discuss correlation between the atmospheric parameters, but I find this discussion underdone. There are grounds to expect significant correlations, and I would like to see a table of correlation coefficients for all atmospheric parameters anomalies so this can be better assessed. For example the authors recognize that CO and ethane may be correlated (both sourced from biomass burning), but I also expect a strong correlation between tropopause height and HF column, since both are dependent on the depth of the stratosphere. The selection of parameters to include in the trend fits is currently made on the basis of the improvement in the reduced Rsquared, but this hides the overview of what is a significant and independent. Reply: A table that shows the linear correlation coefficients between all the anomalies has been added in the paper. Noteworthy is the fact that there is little correlation between the trop height and HF. The reason for this is that most of the HF variability at the mid and northely sties are caused by atmospheric down-welling in the winter months, which is extremely strong at the northern and mid latitude site due to influence of polar air.

Technical comments and corrections

P 8210 Introduction. Here the existing knowledge of trends is described, though this is itself the objective of this paper. Please specify that the trends described here are from in situ data, while the paper is concerned with total column data.

Reply: This is corrected in the paper

L 11 1.1 not 1,1.

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Reply: This is corrected in the paper

L 17. Most N₂O in soils is produced by denitrification, which happens under anaerobic (not aerobic) conditions. But since N₂O is produced by both nitrification and denitrification, I suggest leaving the phrase “under aerobic conditions” out altogether.

Reply: We agree and this is corrected

L22. Prasad 1997 is a poor choice of reference here, the stratospheric sink of N₂O was recognised in the 1970s or earlier and the credit should go to those to whom it is due.

Reply: This reference is changed to Bates and Hays (1967) who were the first to discover the stratospheric N₂O sink

L 23. The methane trend is also monotonic in that it does not reverse and become negative – “continuous” might be a better word to describe the N₂O trend.

Reply: We agree and this is corrected

P8211 L2: “: : as accurately as possible” L13 and L20: “: : unevenly sampled: : :” L14: “Furthermore there are: : :” P8212 L1: “reliable” rather than “trustful” trends L8: FTIR measurements have been performed: : :.

Reply: all above is corrected

L10-12: This is ambiguous, which periods apply to which stations?

Reply: Sentence is reformulated to clarify

L20: remove “under operation”. L25: “calculating” not “calculate” P8213 L5: “: : :according to the principles described by Rodgers (2000)” L7: “The two codes have been shown: : :”

Reply: All above is corrected

L10: The cell measurements are not made continuously, they are made regularly or
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periodically.

Reply: We agree and this is corrected

L16: “biases” between stations would be better than “errors”. The meaning of the last sentence of this paragraph is unclear.

Reply: The last sentence is removed and errors is changed to biases

L18: The retrievals were carried out: : : L26: minimises, not minimise L27: reduces, not reduce P8214 L6: . uses not use

Reply: All above is corrected

L13: “every second day ” is less ambiguous usage than “every other day”

Reply: We agree and this is corrected

P 8215 L3: replace “useless” with “of limited use”.

Reply: This is changed to “of limited use” although the authors could not see any value in a trend without an error estimation.

L5: the residuals are: : : L6: remove the reference to Eq 1 or include a full reference to it – it has not been introduced yet, and does not describe the residuals anyway. L15: constant time spacing. P8216 L8 remove “used” before “obtained” P8217 L11: use “did not” rather than “didn’t”

Reply: All above is corrected

L16: I found this paragraph’s reference to “anomalies” confusing at first. Perhaps by stating at the start “To find the anomalies in the atmospheric parameters that affect: : :.” would help make it clearer.

Reply: We agree, this improves the sentence

L21: : : : as a parameter for which the confidence interval: : :.

Reply: This is corrected

P8218 L11: see general comments, here I think the anomalies should be defined algebraically and tabulated or listed more clearly.

Reply: As stated earlier a table with the anomalies is added in the paper

P8219 L16: point out that Fig 1 is just an example of an anomaly plot.

Reply: This is corrected

P8220 L7-13: How does the difference in sensitivity to HF column for CH₄ and N₂O relate to the fraction of CH₄ and N₂O in the stratosphere in each case.

Reply: We assume that fraction in this case means the stratospheric part of the total column at each station for methane and nitrous oxide. For the Alp station this fraction is larger due to the station altitude but here less dynamics is present (lower latitudes). At the northern stations the fraction is smaller (lower altitude) but more dynamics is present due to e.g. the polar vortex. It is therefore not a straight forward task to do this comparison and we think it is outside the scope of this paper which is a trend paper.

L21: : : and 1.0% respectively.

Reply: This is corrected

L27: here possibility of co-linearity of CO and C₂H₆ is mentioned, but HF and tropopause height is ignored. See general comment.

Reply: The issue of co-linearity in the model is discussed under section 5.4, model stability, where the VIF factor for the trend models is presented. It is stated that "In our case the calculated VIF factors are well below 10 for all FTIR stations and both of the species under investigation".

P8221, and Fig 3 and 4. I would like to see the residuals plotted with the fits in each case here, From Fig 3 it is not at all obvious that the piecewise linear regression over

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three periods would be any better than the simple linear fit. Is it significantly better in a statistical sense?

Reply: A figure like Figure 3 is added for methane with three piecewise trends. In this way the readers can by themselves conclude if the piecewise approach is better or not. It is always difficult to find change points in time series and quite often these are detected with the human eye (in this article both with the eye and by previous articles regarding methane time series in the Northern Hemisphere). In this article we therefore present the trend results both with and without the change points for methane.

P8222 L11, 12, 26: specify when you mean trends in in situ data, and when in total columns

Reply: This is clarified throughout the article

P8223 L8: : : : a function consisting of : : : L24: authors' (add apostrophe) P8224 L2: do not, not "don't"

Reply: All above is corrected

L9: In figures 1, 6 and 7, it would be better to use symbols, not lines in the plots of residuals. These are uneven and discontinuous series, and the connecting lines between points are distracting.

Reply: We agree, this is corrected. In addition a normal distribution for the given standard deviation and mean will be added to the histograms.

P8225 L14: criterion (singular), not criteria (plural) P8227 L6: two possible reasons: : : L18: : : less influenced by this transport: : : L24: : : is most likely not responsible

Reply: All above is corrected

Figure 2. See general comment, I do not follow what the Linear trend is in these charts, it is not an "atmospheric parameter", and not the same as the values in Table 3. Also the calculation of the plotted values is not explicitly explained.

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Reply: See earlier reply under General comments

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 8207, 2011.

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