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> Interactive Comment

Interactive comment on "A new method to detect long term trends of methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) total columns measured within the NDACC ground-based high resolution solar FTIR network" by J. Angelbratt et al.

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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 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

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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. 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.

The work appears to have been thoroughly carried out, and the paper well produced (notwithstanding corrections requested below). Although the analysis does not radically improve trend analysis from such datasets, it is of sufficient value to the NDACC community and suitable for publication in ACP after general and technical revisions and corrections listed below are addressed.

#### 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. 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.

I do not think the surface pressure should be considered as an anomaly at all - it is

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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. 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 recognise 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 R-squared, but this hides the overview of what is a significant and independent.

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.

L 11 1.1 not 1,1.

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.

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.

L 23. The methane trend is also monotonic in that it does not reverse and become

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negative – "continuous" might be a better word to describe the N2O trend.

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....

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

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..."

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

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

L18: The retrievals were carried out...

L26: minimises, not minimise

L27: reduces, not reduce

P8214

L6: . uses not use

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L13: "every second day " is less ambiguos usage than "every other day"

P 8215

L3: replace "useless" with "of limited use".

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"

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.

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

P8218

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

P8219

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

P8220

L7-13: How does the difference in sensitivity to HF column for CH4 and N2O relate to

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the fraction of CH4 and N2O in the stratosphere in each case.

L21: ...and 1.0% respectively.

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

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

#### P8222

L11, 12, 26: specify when you mean trends in in situ data, and when in total columns P8223

- L8: ... a function consisting of ...
- L24: authors' (add apostrophe)

P8224

L2: do not, not "don't"

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.

P8225

L14: criterion (singular), not criteria (plural)

P8227

L6: two possible reasons...

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L18: ...less influenced by this transport...

L24: ... is most likely not responsible

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.

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

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