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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Carbon monoxide (CO) and ethane (C_2H_6) trends from ground-based solar FTIR measurements at six European stations, comparison and sensitivity analysis with the EMEP model" *by* J. Angelbratt et al.

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Interactive comment on "Carbon monoxide (CO) and ethane (C2H6) trends from ground-based solar FTIR measurements at six European stations, comparison and sensitivity analysis with the EMEP model" by J. Angelbratt et al.

Anonymous Referee #3

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General Comments

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Interactive Discussion

Discussion Paper

This paper presents measurements of the tropospheric partial columns of CO and C2H6 made by FTIR spectrometers at six European stations. Trends from 1996 to 2006 are presented for four of the stations, and are negative for both gases, at about -1% per year. Simulations by the EMEP chemical transport model are compared with the measurements, revealing generally reasonable agreement in the average amplitudes, the seasonal cycle, and the differences between stations. However, there are some discrepancies for CO during periods of large-scale biomass burning in North America and Russia. EMEP tends to overestimate the seasonal cycle and underestimate the columns for CO, while overestimating both the seasonal cycle and columns for C2H6. The model was also used to perform a sensitivity analysis to examine possible causes of the observed CO trends. Reductions in European CO emissions were found to largely explain the trends, while the decrease and increase in North American and East Asian CO emissions, respectively, also had an impact on the measured columns. This paper is a useful contribution to the field, providing a ten-year time series of tropospheric measurements in Europe, and using a chemical transport model to interpret the results. I recommend publication in ACP after the comments below are addressed.

Specific Comments

1. Page 13725, line 2: The title mentions six FTIR stations, while this first sentence refers to four. This causes some confusion until the reader realizes that data from six stations are presented and compared with the model, but that measurements from only four of the stations are used to derive trends. Nowhere is it explained why trends are not derived for Bremen and Ny Alesund. This should be explained somewhere near the start of the paper.

Reply: A sentence is added to clarify/explain this in the trends and tracers section.

2. Page 13725, line 2: Define FTIR. Acronyms are poorly defined throughout the paper. Define all acronyms once on first use in the Abstract, and again once on first use in the

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main body of the paper. Also EMEP MSC-W on line 8.

Reply: The acronym of FTIR is added and is now defined both in the abstract and in the paper, this according the reviewers comment. The EMEP MSC-W acronym is not added to the abstract because a full definition would cover almost two lines. The paper is also published in an EMEP special issue and the readers are assumed have basic knowledge about the EMEP model. The full acronym is instead defined in the introduction.

3. Page 13725, line 5: State what the +-% terms in the trends represent.

Reply: The $\pm$ is the confidence intervals (2-σ). An explanation is added to the abstract.

4. Page 13730, lines 4-6: This appears to be the only place in the paper where errors on the FTIR measurements are discussed. No error bars are included in the plots of the FTIR data. This is unsatisfactory. Ideally, a full error budget would be calculated for each of the six sites, following the formalism of Rodgers (2000). Failing that, perhaps a representative error budget could be presented for one or two of the sites. As NDACC stations, this information should be available. If this task is too difficult, then at least some discussion of the applicability of the Zhao et al. (2002) errors to this work should be added. e.g., what terms were included in the error budget calculated by Zhao et al.? How appropriate are those low-altitude, mid-latitude errors to the variety of sites used in this work, which include high altitudes and high latitudes?

Reply: The Zhao error budget includes both systematic and random errors and is also relevant for the European FTIR sites. The used micro-windows with interfering species and retrieval algorithm (SFIT2) are the same for the two papers. Although, there are site specific errors due to the Instrument Line Shape (ILS), the a priori profile, the spectral quality and pressure and temperature profiles. The site specific errors account in the Zhao case for roughly 50% of the total error for the two gases. Of those 50% the a priori and pT profiles are the dominant error sources while the ILS only contributes to a relatively small error. The error budget presented by Zhao can be compared with

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Rinsland et al. (2000 and 2007). Here the CO and C2H6 errors are quantified for Jungfraujoch and Kitt Peak. For CO the total error is between 5.2-6.7% and for C2H6 the total error is between 8.4 and 8.9%. An expanded discussion based on these three papers is therefore added to the paper.

5. Page 13733, line 24: Add a brief discussion about the differences between the meteorological drivers, and the implications of these differences for the results shown in this study.

Reply: We have added the description of the different drivers in section 4 (the EMEP model). We also refer to Tarrason et al. (2008) who discussed the differences between the drivers for European air quality simulations.

6. Page 13734, line 9: Line 1 of the Abstract mentions partial columns of 0-15 km. Here, it appears that 100 hPa is used as the upper boundary for the partial columns for compatibility with the EMEP lid. Explain why the FTIR tropospheric partial columns are calculated by subtracting the stratospheric component derived from just two sites from the total columns at each site, rather than just integrating the retrieved VMR profiles up to 100 hPa. How do the stratospheric partial columns compare for the other sites?

Reply: First, profiles were only available for two of the stations when the data analysis for the paper was done. Secondly, finding the average above 100 hPa for these two stations is considered as a good approximation since the FTIR has a low vertical resolution and the partial column at a certain altitude range contains information from both above and below its own range. This means that also when summing up the partial columns from the surface to the 100 hPa some information from the layers above will be included in the final partial column and introduce an error.

7. Page 13734, Section 5.1: Have the model data been smoothed by the FTIR averaging kernels and a priori profiles? It appears not. Ideally this should be done when comparing measurement and model data.

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Reply: The model data have not been smoothed with the average kernels. Since the model extends only to ∼15 km and the measurements up to 100 km a smoothing of the model with the measurement kernel would introduce errors instead of improving the result. If the EMEP model was working to the same altitude as the measurements a smoothing with the kernel might be a good idea.

8. Page 13738: It is not entirely clear why Sections 5.3 and 5.4 are separate. Clarify the difference between the sensitivity analysis and the uncertainty analysis. Table 3 lists all of the runs as sensitivity scenarios.

Reply: Section 5.3 is the sensitivity analysis where the global scale EMEP model with convection is used to find possible reasons for the CO trends. Here the model with convection is modified in 9 scenarios (this is marked as sensitivity scenarios in Table 3). In section 5.4 other model versions are compared with each other and the FTIR data, this to find out how a model with forest fire module and without convection scheme would perform. Table 3 does not only list all sensitivity versions. The upper part of the table is denoted EMEP model versions and the lower sensitivity scenarios.

9. Page 13738, line 14-17: The two tests Gc-high and Gc-low are not listed in Table 3. Neither are the FTIR-high and FTIR-low tests. Why not? Results are shown in Figure 5 and discussed briefly in Section 6.2.3.

Reply: Gc high and low represent the upper and lower model layer used in the interpolation to obtain the partial column at the station altitude. They are therefore not a model version and neither a sensitivity analysis related to the trends. A clarification of this is added in section 6.2.3 and in the caption to Figure 5. FTIR high and low is the uncertainties related to the calculations of the partial columns from the FTIR data. Also here a clarification is added both in the text and in the caption.

10. Page 13739, line 16: Clarify whether this refers to the horizontal or vertical model-boundary.

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Reply: Here it should be the horizontal boundary. This is added in the paper.

11. Page 13741, line 12-13: It is not clear what is meant by the sentence starting with "Except the trends given in Rinsland : : :". Clarify. Can the trends from this paper be compared with those in Rinsland et al. and in Mahieu et al.? If not, why not? If so,include them.

Reply: The trends in this paper can be compared with Rinsland and Mahieu and they agree quite well. The sentence is reformulated and the trends with confidence intervals from the two papers are added.

12. Page 13742, line 12-16: Clarify what was done here, e.g., 1996-2006 versus 2006, definition of the baseline scenario.

Reply: In order to obtain this an average value is calculated for all sensitivity and reference runs. In this paper Gc is used as reference for each station. The January and February data is removed due to the high influence from the start conditions. The differences between the reference run and the sensitivity scenarios is then related to the reference run, this for a certain station, and then scaled with the 10 year period (1996-2006). The explanation in the paper is clarified.

13. Page 13745, Section 7: The Conclusions section is rather short. It could be expanded with some discussion of quantitative results and their significance.

Reply: The conclusion is expanded and a more detailed discussion regarding the trends is added.

Technical Corrections There are many distracting typographical errors. The manuscript should be carefully proof-read to correct these. Some are identified below. There are also numerous places where a comma would help clarify the flow.

Reply: This is done and corrections are made at several locations.

Page 13725, line 5: estimated to BE Page 13725, line 14: modelS have been Page

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13725, line 17-18: is bias more appropriate than deviation? Page 13725, line 22: year is unnecessary, just 2006 (similarly, elsewhere in the paper time period is redundant as in “time period 1996-2006”) Page 13725, line 23: 2006 ARE shown : : : 37-50% OF THE MEASUREMENTS for k Page 13725, line 25: assumptions MADE in this paper Page 13726, line 25-26: Define FTIR and EMEP MSC-W Page 13727, line 1: datasetS Page 13727, line 4: mid-infrared : : : high-resolution (also change throughout the paper) Page 13727, line 11: especially FOR the spring maximum Page 13727, line 10: delete earlier, or replace with previously Page 13727, line 24: 2/3 of THOSE FROM biomass burning Page 13728, line 8: photodissociation Page 13728, line 9: and REACTION WITH water VAPOUR (or H2O) Page 13728, line 11: intra-annual (also change throughout the paper) Page 13728, line 16-17: remove italics from O(1D), O2, and O3 Page 13728, line 19: biofuel Page 13728, line 20: also estimate the Page 13728, line 21: To BE 2.1 Page 13728, line 22: located IN the Northern Hemisphere Page 13729, line 4-5: partial column data for CO Page 13729, line 17: ill-posed Page 13729, line 18: is a weightED COMBINATION of an Page 13729, line 19: delete method – M in OEM stands for method Page 13729, line 22: sumMING Page 13729, line 24: which useS PROFFIT Page 13729, line 25: microwindows or micro-windows Page 13729, line 26: ARE in the region Page 13729, line 27: For CO, the species Page 13730, line 1: and H2O interfere in the C2H6 microwindow Page 13730, line 2: define UFTIR Page 13730, line 11: EMEP should be defined on first use, on Page 13726, line 26, not here Page 13730, line 19: Chemical Transport Model or chemical transport model Page 13730, line 22: define EQSAM Page 13731, line 3: delete “of EMEP model” Page 13731, line 24: concentrationS Page 13731, line 27: change period after Eq. (2) to comma Page 13732, line 1: where h : : : height, with Page 13732, line 4: no indent Page 13732, line 12: delete hyphen: global scales Page 13732, line 13: define HTAP Page 13733, line 1: databases Page 13733, line 5: compoundS Page 13733, line 5: define SNAP Page 13735, line 9: above-mentioned Page 13735, line 27: model; the Page 13736, line 1: baseline Page 13736, line 5: the mass balance IS Page 13736, line 6: representS Page 13736, line 9 and 14: inconsistent formatting of subscripts (biomass burning,

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CH4 oxidation) Page 13736, line 17: 11-year period Page 13736, line 19: have BEEN shown Page 13736, line 20: have BEEN shown to Page 13737, line 10: reduced by 20% Page 13737, line 17-18: Clarify whether the 0.2 degree C increase is per year. Page 13737, line 20: "last sensitivity scenario" – Table 3 lists another scenario after this one, Gcnobvoc, discussed in Section 5.4. Page 13737, line 22: have BEEN shown to Page 13737, line 25: 2009; Angelbratt et al., 2011) Page 13737, line 27: and calculate the Page 13738, line 2: with the global model, which explore the Page 13738, line 10: baseLINE case Page 13738, line 11: Sect. 5.1 Page 13739, line 6: delete further Page 13739, line 7: change ; to comma Page 13739, line 13: BICs has already been defined Page 13739, line 27: and so WERE omitted Page 13740, line 1: FurtherMORE, Page 13740, line 6: add period after column Page 13740, line 8: and deviates FROM THE FTIR DATA BY as much as Page 13740, line 9: inter-station Page 13740, line 21: the reasons FOR the strong Page 13741, line 26: delete year; Figs. 3 and 4 Page 13742, line 4: overestimate Page 13742, line 9: for some BY as much as : : : The European model also underestimates Page 13742, line 14: baseline scenario Page 13742, line 17: reduction by 20% Page 13742, line 22: emission by 20% Page 13742, line 23: add semicolon after yr-1 Page 13742, line 25: Is 0.4%/year correct? The text and Table 3 say 1.2%/year for CH4. Page 13742, line 26: 11-year period Page 13742, line 27: theSE last two Page 13743, line 3: delete will Page 13743, line 4: deviateS BY a factor Page 13743, line 10: delete layer Page 13743, line 14: explanation FOR the Page 13743, line 18: on the other HAND Page 13744, line 7: contribute Page 13744, line 16: reproduceS Page 13744, line 18: illustrateS : : : decreaseS Page 13744, line 20: It is also SEEN that Page 13744, line 25: emissions Page 13745, line 3: ground-based Page 13745, line 12: was only working for 2006, the analysis Page 13745, line 13: the effect of Page 137456, line 3: Is J. Geophys. Res.-Ocean Atmos. correct? Page 13754, Table 1 caption: Global sources of CO and C2H6 (Tg yr-1 and % of total). Page 13756, Table 3: For GcEAAll20, shouldn't it be a 20% increase rather than a 20% reduction? See page 13737, line 14. Make clear the time periods for each reduction (1996-2006 or 2006) and that they are per year. Add Gc-high, Gc-low, FTIR-high, and

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FTIR-low? Page 13760, Table 7 caption: Swap the order in the caption to match that in the table (FTIR, Gc, E). ": : : because of the strong influence from the initial conditions : : :" Page 13761, Table 8: Use the same order for the sensitivity cases by column as they are listed in Table 3. Here or in the text, explain why N/A for Bremen and Ny Alesund.

Reply: all technical corrections above are corrected according to the reviewer's suggestions.

Page 13763, Figure 1 caption: "The difference between measurements and the model IS marked: : :" Why show the differences as lines rather than points?

Reply: The difference is clearer, easier to see, as lines. We have considered to plot the difference as dots but this did not improve the figure.

Page 13765, Figure 3: Difficult to distinguish the blue and black points. Caption: (red triangles) (blue? squares) (black? diamonds) : : : data are also shown. Delete "in the figures". : : : the model initial conditions. What decreases (line 3)? The amount or the influence? Delete "given in the literature" – the citation is sufficient.

Reply: the caption is reformulated and some of the sentences are removed/changed according to the reviewer's suggestions. The two figures are changed. FTIR is presented as dots without connecting lines and the European model is presented as dotted lines.

Page 13766, Figure 4 caption: As in Figure 3: : :

Reply: this is changed.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13723, 2011.