Atmos. Chem. Phys. Discuss., 12, C3228–C3240, 2012
www.atmos-chem-phys-discuss.net/12/C3228/2012/
© Author(s) 2012. This work is distributed under
the Creative Commons Attribute 3.0 License.

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trends, interannual and seasonal variations of tropospheric CO, C2H₆ and HCN columns measured from ground-based FTIR at Lauder and Arrival Heights" *by* G. Zeng et al.

G. Zeng et al.

guang.zeng@niwa.co.nz

Received and published: 5 June 2012

Reply to Reviewer#2

We are grateful for the reviewer's very detailed and constructive comments. We totally agree with the reviewer's insightful comments on the shortcoming of the data analysis and the model exploitations. We have revised the paper substantially as a result. The main revisions are: 1) we have added HCN time series at Arrival Heights and calculate trends of CO, C2H6, and HCN partial columns at Lauder and Arrival Heights using daily

mean partial columns (rather than instantaneous data as in the submitted manuscript) for the period 1997-2009 in a way that is more consistent with previous studies (e.g. Rinsland et al. 2002). As a result, the calculated trends are changed slightly. 2) We have added model simulations of HCN, and have applied averaging kernels to all the model data for comparison with the instantaneously observed partial columns of CO, C2H6, and HCN for the period of 1997-2009, as requested by reviewer#1. In this case we compare model data with observed data in a more robust way. 3) We have performed sensitivity simulations to quantify the contribution of non-biomass burning C2H6 sources to the SH ethane columns through inter-hemispheric transport and the results indicate an substantial impact on the C2H6 columns at Lauder and Arrival Heights, as pointed out by the referee. This is consistent with the literature. 4) We have further examined the hypothesis of declining C2H6 emissions from the non-biomass burning sources (i.e. industrial sources) during the past decade and the impact on C2H6 trends at Lauder and Arrival Heights; we need to assume at least $\sim$25% decline of C2H6 industrial sources in our model to reproduce the trends of the observed C2H6 columns at both locations. We also apply the 25% decline of CO industrial sources in the model over the period of 1997 to 2009 and, as a result, the model almost completely catches the observed CO trends at both locations. We have revised various statements accordingly in the paper. Detailed revisions are listed below.

Re: P6185 Title: I think the title would have a broader appeal if "Lauder and Arrival Heights" was replaced by "New Zealand and Antarctica" or "the mid-and high Southern Hemisphere".

We adopt the reviewer's suggestion to change the title to "Trends and interannual and seasonal variations of tropospheric CO, C2H6 and HCN columns measured from ground based FTIR over New Zealand and Antarctica".

Re: P6186 L6: Why isn't the HCN trend at Arrival Heights cited? Its omission contradicts "all species at both locations".

ACPD
12, C3228–C3240, 2012

CC BY

We have added in HCN data at Arrival Heights and have calculated corresponding trends (∼-1.41±0.71%/yr). We have also added model simulations of HCN data at both locations. Note that we did not include the Arrival Heights HCN data in the first instance as we see the observed data as preliminary. However, the model results and the data from Lupu et al (2002) both confirm the reasonable HCN values and phase of the cycle.

Re: P6186 L11-14: This result has already been reported for Lauder in the abstract of Rinsland et al. (2002). Please consider whether it is more appropriate for the abstract or the introduction.

We have moved the statement to the introduction.

Re: P6186 L18: Also tell the reader whether or not the model reproduced the observed HCN trends.

We have carried out simulations of HCN in the model (assuming main emission sources are from biomass burning), and calculate the residuals between convolved model data (by applying a priori and averaging kernels) and the observed data at both locations. We then perform linear fits to the residuals and it shows no obvious trends of the residual which indicates that the model does catch the observed HCN trends. This is shown in a new figure.

Re: P6186 L24-25: Why would seasonal cycles of CO and C2H6 be impacted by biomass burning from South America and Asia, but interannual cycles be impacted by Australian emissions? Why wouldn't Australian emissions also impact seasonal cycles, and vice versa?

Seasonal cycles of CO and C2H6 are influenced by biomass burning from all regions mentioned but was more pronouncedly impacted by biomass burning (BB) from Southern Africa (shown in black line instead of blue line in the figure – this has now been corrected) and South America (red line) as shown in Figure 5. Largest interannual

ACPD

12, C3228–C3240, 2012

anomalies are characterised by Asian BB (blue line) related to 1997-1998 ENSO, Australian bush fire (green line) and burning from South America. These are noticeable events that contribute to the anomalies of CO and C2H6 columns at Lauder and Arrival heights. We have modified the text to state this more clearly.

Re: P6187 L14: This reference is 20+ years old; please find a more updated reference for the lifetime estimates.

We have added Holloway et al. (2000) and Rudolph (1995). Holloway, T., H. Levy II, and P. Kasibhatla (2000), Global distribution of carbon monoxide, J. Geophys. Res., 105(D10), 12,123–12,147, doi:10.1029/1999JD901173. Rudolph, J. (1995), The tropospheric distribution and budget of ethane, J. Geophys. Res., 100(D6), 11,369-11.381.

Re: P6187 L14: Change “hence” to “and” because short-lived species are also influenced by vertical mixing and long-range transport.

Done

Re: P6187 L18: These references are also 20+ years old. For example in the case of C2H6, newer references show the importance of interhemispheric transport from the northern hemisphere as a source of C2H6 to the southern hemisphere, in addition to biomass burning within the southern hemisphere. Please update and revise.

We have added Matsueda et al. (1998, 1999) and Rinsland et al. (1998). We now have cited Xiao et al. 2008 and have added text to state that inter-hemispheric transport of NH C2H6 is an important source of C2H6 in the SH.

Re: P6187 L19-20: Same comment about old HCN references. You could also cite the budget studies of Li et al. (2003) and Singh et al. (2003).

We have added Li et al. (2000, 2003) and Singh et al. (2003).

Re: P6187 L22-24: In the case of C2H6, how does the significant amount of interhemispheric transport affect this statement?

ACPD

12, C3228–C3240, 2012

CC BY

We have now cited Xiao et al. 2008 and modified the text accordingly: "and also the impact of NH industrial sources through inter-hemispheric transport in the case of C2H6 (Xiao et al. 2008)".

Re: P6188 L1: If this data set extends back to 1993 for CO and C2H6, why does this paper only report data since 1997?

The data before 1997 were reported by several previous studies. Another reason that we only show data from 1997 is because the biomass burning emission inventory (GFEDv3) which we use for the model simulations is only available for the period of 1997-2009. Hence we report the data for the same period for a more consistent comparison with the model results.

Re: P6189 L28: Provide a reference for 1760 ppb of CH4 in 2000.

We use 1760 ppbv of methane based on averaging methane levels between 1995 and 2005 (1745-1775ppbv) in IPCC AR4 that is representative of present-day global averaged methane levels. The reference has been added in.

Re: P6189 L27-29: The global concentration of methane has shown interannual variations as well as long-term trends from 1997-2009. For example its global increase over this period is on the order of 1.7%. What sensitivity tests have you performed to determine whether or not methane's year-to-year variations will affect your results, and what impact keeping methane constant will have?

We have performed a sensitivity simulation by increasing the global methane concentration by 100 ppbv (5.7%); this results in an approximately 3% increase in the CO columns at Lauder and Arrival Heights. Hence a 1.7% increase of global methane during the period of 1997-2009 is estimated to lead to a 0.9% increase of CO columns (0.068%/yr) which is substantially smaller than the trends of CO column at both locations.

Re: P6189 L27-29: Methane has a strong interhemispheric gradient; how is this han-

ACPD

12, C3228–C3240, 2012

dled by the model? In other words is using a global average of 1760 ppb compatible with the SH focus of your study?

We did not take into account of the interhemispheric gradient in methane. The typical ratio of NH/SH methane is around 1.06. From the sensitivity simulation mentioned above, we don't expect that using a global constant methane of 1760 will have visible bias on the SH focus of the study here.

Re: P6190 L1: Again if you look into the more recent ethane literature, interhemispheric transport is a greater source of ethane to the SH than is biomass burning within the SH (e.g., Rudolph et al., 1995; Xiao et al., 2008). Was this accounted for in the model, and how does this affect your results? For example Aydin et al. (2010) show a 10% decline in ethane levels at high southern latitudes from 1980-2000, apparently in response to a declining global fossil fuel source of ethane. This suggests that the stated assumption of no year-to-year variations of non-biomass burning sources of ethane is incorrect. I think the discussion needs to be reworked with careful consideration of the complexity of the ethane signal in the SH, including interhemispheric transport and long-term trends of non-biomass burning sources.

The annual total C2H6 emissions that are stated in the paper (P6190 L9-10) are NOT the actual numbers that we use in the simulations. The actually ethane emissions in the model are 6.1 Tg/yr from the anthropogenic sources and 1.5 Tg/yr to 3.0 Tg/yr from the biomass burning (interannually varying). Therefore the ethane emission in our model is lower than that used by Xiao et al. 2008, but is more in line with the EDGAR recommendation (Olivier et al., 1996). We also performed a simulation using global annual total non-biomass burning C2H6 emissions equivalent to Xiao et al. (10.2 Tg/yr) but then the model visibly overestimates the observed time series. We have now summarised all emissions used in the model in a table.

We have also applied averaging kernels to the modelled C2H6 data and calculated the residuals (modelled-observed). We find that the linear fits to the residuals for C2H6 at

ACPD

12, C3228–C3240, 2012

both locations have substantial trends; this indicates that by assuming constant emissions from non-biomass burning (i.e. industrial sources) for C2H6 (large proportion of C2H6 sources) the model does not capture the trends in the observed C2H6 columns (shown in a new figure). We have carried out a few sensitivity tests assuming declining C2H6 anthropogenic emissions over the period of 1997-2009. The results show that with an approximately 25% decline of the global non-biomass burning C2H6 sources during 1997-2009 the model catches the observed trends at both locations.

We have applied the same procedure to modelled CO data and find moderate trends in the linear fits to the residuals (modelled-observed) at both locations. This is shown in a new figure. It may indicate some decline in other sources (e.g. fossil fuel) that are not accounted for in the model (by including interannually varying biomass burning sources in the model, the model is able to catch the trends of observed HCN which has predominately biomass burning sources). We have performed a further test by assuming a 25% decline in global CO industrial sources and, as a result, the model is able to catch the observed CO trends at both Lauder and Arrival Heights almost completely. We show these results in a new figure.

We have modified the text and figures substantially to show modelled and observed data and the residuals (modelled-observed) for CO, C2H6 and HCN. We apply a priori and averaging kernels to each model data set. These new figures would now replace figure 4a.

Re: P6190 L5-8: This sentence about partitioning doesn't make sense to me. Also are the 6 NMVOCs listed on P6189 the only ones included in the model? If so then "higher" is not appropriate.

The total emissions of NMVOCs (excluding isoprene) from the recent estimates (Lamarque et al. 2011) are partitioned among C2H6, C3H8, HCHO, CH3CHO and CH3COCH3 in the model to account for the effect of NMVOCs that the model does not include. However we do not lump any other species' emissions into the C2H6

ACPD

12, C3228–C3240, 2012

emissions so that the model produces realistic C2H6 concentrations. Other primary compounds not listed above are effectively “lumped” together with the listed ones. We have modified the text to clarify. We no longer call these "higher organics".

Re: P6190 L9-10: Where do the estimates of 11.5 and 5.2 Tg/yr that are being used come from? They total 16.7 Tg/yr, which is much higher than estimates of ethane’s global budget within the past 10 years (e.g., Xiao et al., 2008; Pozzer et al., 2010). Why would the model need such high ethane emissions in order to correctly simulate the observations?

These numbers have now been corrected as stated above (6.1 Tg/yr from industrial source and 1.5-3.0Tg/yr from biomass burning source). We apologize for the confusion.

Re: P6190 L12: Same comment for isoprene. Why would the model need such low isoprene emissions in order to correctly simulate the observations? What limits does this place on the certainty of the model output and the extent to which it can be used to interpret the CO and C2H6 observations? Please be quantitative in your response.

Firstly, global isoprene emissions are very uncertain (see refs. in the text) and there is a large range of total annual emissions used in individual models. It is a difficult task to constrain global isoprene emissions. Moreover, CO columns in the SH are more sensitive to isoprene emissions than in the NH as isoprene is a more source of CO in the SH compared to the NH. We performed a simulation based on a global total annual emission of 560 Tg/yr (Guenther et al., 1995). In this case the model overestimates CO partial columns at Lauder by $\sim$20% in average and Arrival Heights by $\sim$25% in average. We change the global total annual emissions of isoprene but do leave the seasonal cycle intact. The magnitude of isoprene emissions has a minimal influence on C2H6. We plan to carry out a further study to assess the impact of isoprene emissions on CO columns in the SH and explore the feasibility to constrain isoprene emissions using SH CO columns measurements.

ACPD

12, C3228–C3240, 2012

CC BY

Re: P6190 L14-16: Are the uncertainties as large as 170 Tg/yr?

Other sources of CO are relatively well-defined but the global isoprene emissions are poorly constrained.

Re: P6190 L24: Regarding "in the absence of in the absence of industrial sources in the Southern Hemisphere mid- to high latitudes", see comments above about import of ethane from the NH (e.g., P6190 L1).

We have now corrected this statement.

Re: P6192 L1: Again I do not believe the statement about lack of industrial ethane sources in the SH is correct.

Agree. It has been corrected and the paper has been revised substantially in this respect.

Re: P6192 L2: The difficulty with the OH argument is it would impact all OH-controlled species equally. The statement that changes in OH could play a critical role seems too speculative if there isn't any kind of concrete evidence to back it up.

Agree. We have now modified the statement.

Re: P6192 L19: I agree with the difficulty of the 1997-98 El Nino at the start of the time series forcing negative trends. What does the HCN trend look like if you begin the time series in 1999?

The HCN trends change from -0.93% for 1997-2009 to -0.62% for 1999-2009.

Re: P6192 L24: This statement is also speculative. What evidence is there in the literature for a downward trend in tropical biomass burning? For example several papers on global biomass burning emissions have shown either upwards or steady trends in the past few decades. The larger ethane decline (compared to CO) could be explained by declining fossil fuel contributions in the ethane component that is imported from the NH.

ACPD

12, C3228–C3240, 2012

We agree and have modified the text. Indeed, the large C2H6 decline at Lauder and Arrival heights is the result of declining fossil fuel sources of C2H6 in the NH that is transported to the SH, due to the large fraction of the fossil fuel component in C2H6 emissions. This has now been addressed in the revised paper.

Re: P6192 L25: Omit "thought to" because HCN is well known to be produced from biomass burning.

Changed.

Re: P6192 L28: What happens if you begin the trend in Feb 1998 or Feb 1999?

The trend is reduced from -0.2*e15 molecules cm-2 yr-1 beginning in Feb 1997 to -0.12*e15 molecules cm-2 yr-1 beginning Feb 1998.

Re: P6193 L10: Change “considerably large” to “reasonable” or “some”.

Done

Re: P6193 L18: “The focus of the simulation is on characterizing the seasonal and interannual variations rather than improving the comparison between the modelled data and the observed data.” It seems that if the model can reproduce the data then it has a better chance of correctly characterizing the variations.

We agree with the reviewer’s point. We have convolved the model data with averaging kernels and have modified the text accordingly.

Re: P6194 L11: “although they might affect SH through inter-hemispheric transport”. This is already established in the existing literature, for example in the case of ethane. The paper needs to be reworked with a better understanding of how interhemispheric transport will affect the observations, and this needs to be correctly incorporated into the model.

We have updated model simulations of C2h6 and examine the impact of inter-hemispheric transport of NH C2H6 source to the SH. We have modified the text ac-

ACPD

12, C3228–C3240, 2012

CC BY

cordingly. In detail, we calculate an average of 77% contribution of global fossil fuel sources to Lauder C2H6 partial columns and an average of 72% to Arrival Heights C2h6 partial columns. (89% of fossil fuel emissions are from NH). We also calculate separate contributions from NH C2H6 industrial sources to C2H6 partial columns at both locations; an average of 60% contribution is calculated for Lauder and an average of 50% for Arrival heights. These calculations are now added in the paper.

Re: P6194 L8-17: In my opinion this paper would make a more substantial contribution if these effects could be resolved and addressed more quantitatively.

We have quantified the impact of declining NH industrial sources on SH C2H6, which is substantial. We have modified the text accordingly. We cannot quantify other potential impacts at this stage.

Re: P6196 L8: Elsewhere in the paper it talks about the absence of industrial sources in the SH; what non-biomass burning sources are you referring to here?

The industrial sources in the SH account for 11% of the global total. We have modified the text.

Re: Technical Corrections:

We have made all corrections that are specified below and wherein still applies as we have modified the paper substantially.

P6185 Title: Correct from “Trends,” to “Trends and”.

P6186 L4-10: This is a small thing but the order of Lauder and Arrival Heights keeps changing from sentence to sentence. It will be easier for the reader to follow if Lauder is always presented first and Arrival Heights second.

P6186 L12-13: Another small thing but if you write “from August to November” then you could also write “from March to June” to keep it parallel.

P6186 L13: Clarify by changing “this season” to “this latter season”.

ACPD

12, C3228–C3240, 2012

P6186 L13: No need to hyphenate “southern hemisphere”. Also it is capitalized elsewhere in the paper.

P6186 L16: Hyphenate “model-simulated” so “simulated” doesn’t seem like a verb.

P6186 L18: Change “re-produce” to “reproduce”.

P6186 L19: For better transition you could change “Weak” to “Instead, weak”.

P6186 L20: Change “from model” to “from the model”.

P6186 L24: “Nino” needs a tilde.

P6187 L2: “FTS” would need to stand for “Fourier transform spectrometer” or “Fourier transform spectroscopy” but not “Fourier transform spectroscopic”.

P6187 L22: Change “SH” to “SH at”.

P6187 L27-28: The verb tense changes in this sentence; it should stay in the past tense.

P6189 L2: Change “mid- infra-red” to “mid-infrared”.

P6189 L6: All acronyms need to be defined the first time they’re used. Define UM-CAM.

P6189 L11: Define AMIP II.

P6189 L13: Change “compounds” to “compound”.

P6189 L18: Define IPCC AR5.

P6189 L22: Define ENSO.

P6189 L24: All compounds being mentioned for the first time need to be defined.

P6190 L24: Change “tropical” to “as tropical”.

P6192 L1: Change “an critical” to “a critical”.

ACPD

12, C3228–C3240, 2012

CC BY

P6192 L6: Change “different at” to “different at a”.

P6194 L6: Change “Model” to “the model”.

P6194 L11: Change “affect SH” to “affect the SH”.

P6194 L12: Do not capitalize “There”.

P6194 L14: Change “its”’ to “its”.

P6195 L16: Remove “, respectively”.

P6198 L22: Typo in “Crutzen”.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 6185, 2012.