

***Interactive comment on “Trends, interannual and seasonal variations of tropospheric CO, C<sub>2</sub>H<sub>6</sub> and HCN columns measured from ground-based FTIR at Lauder and Arrival Heights” by G. Zeng et al.***

**Anonymous Referee #2**

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General Comments:

This paper presents a long-term time series of CO, C<sub>2</sub>H<sub>6</sub> and HCN columns in the middle and high Southern Hemisphere (New Zealand and Antarctica), and a tropospheric chemistry-climate model is used to simulate the observations. The key finding is negative long-term trends in all three species in the SH. Seasonal and interannual biomass burning variability is also observed.

My assessment of this paper is mixed. The observations of negative long-term trends

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of these species in the SH is a new and important contribution to the literature. However several aspects of the data analysis and referencing need major revisions. The introduction needs to be updated and revised. Many of the references are older and do not accurately reflect the current state of knowledge of CO, C<sub>2</sub>H<sub>6</sub> and HCN. As a result the paper has omitted some important findings from the literature that are directly relevant. For example, the paper has not adequately recognized the importance of interhemispheric transport of ethane from the Northern Hemisphere to the SH, even though this is a larger ethane source to the SH than biomass burning within the SH. It also isn't clear why the model needs high ethane emissions and low isoprene emissions to correctly simulate the observations, and whether this limits the usefulness of the model results. Other parts of the discussion seem speculative, for example the possibility that tropical biomass burning has decreased or that OH levels have changed substantially within the SH. Such statements need to be backed up quantitatively in order to meaningfully contribute to the discussion. My specific comments are given below.

Specific Comments:

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”.

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

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.

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

P6186 L24-25: Why would seasonal cycles of CO and C<sub>2</sub>H<sub>6</sub> be impacted by biomass

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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?

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

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

P6187 L18: These references are also 20+ years old. For example in the case of C<sub>2</sub>H<sub>6</sub>, newer references show the importance of interhemispheric transport from the northern hemisphere as a source of C<sub>2</sub>H<sub>6</sub> to the southern hemisphere, in addition to biomass burning within the southern hemisphere. Please update and revise.

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).

P6187 L22-24: In the case of C<sub>2</sub>H<sub>6</sub>, how does the significant amount of interhemispheric transport affect this statement?

P6188 L1: If this data set extends back to 1993 for CO and C<sub>2</sub>H<sub>6</sub>, why does this paper only report data since 1997?

P6189 L28: Provide a reference for 1760 ppb of CH<sub>4</sub> in 2000.

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?

P6189 L27-29: Methane has a strong interhemispheric gradient; how is this handled by the model? In other words is using a global average of 1760 ppb compatible with

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the SH focus of your study?

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.

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.

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?

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 C<sub>2</sub>H<sub>6</sub> observations? Please be quantitative in your response.

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

P6190 L24: Regarding "in the absence of industrial sources in the Southern Hemisphere mid- to high latitudes", see comments above about import of

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ethane from the NH (e.g., P6190 L1).

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

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.

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?

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.

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

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

P6193 L10: Change "considerably large" to "reasonable" or "some".

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.

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

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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.

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

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?

Technical Corrections:

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".

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

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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”.

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”.

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

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