

## ***Interactive comment on “Five-year flask measurements of long-lived trace gases in India” by X. Lin et al.***

### **Anonymous Referee #3**

Received and published: 9 June 2015

The authors present a multi-species time series of trace gas data from three flask stations in India. The data represent a very valuable contribution to this area of the world, which is currently poorly monitored, and the authors have analysed various aspects of the data (trends, gradients, etc) and covariance between species. However, the manuscript requires some revision to bolster some of the scientific conclusions that are made. If these comments can be addressed, the manuscript should be published.

General comments: 1. Introduction – There should be a comprehensive review of other measurement programs in South Asia – there is a description of CARIBIC and satellite-based studies, but there lacks a detailed description of other measurements (e.g., Bhattacharya et al., 2011, Ganesan et al, 2013 and Tiwari et al, 2014). At the moment it reads as though there are no other surface measurements (whether concur-

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rently or previously) and while the authors discuss some very brief comparisons in the Results section, this needs to be brought forward into the Introduction. On page 7175 Line 9: ‘Besides a lack of observation sites’ should be written more accurately, which is that there are few observation sites in addition to those presented here, but this is not enough to constrain a large country like India.

2. The authors compare their data to many other sites from NOAA and ICOS. While it is understandable that these measurements are directly linked to the authors and may overlap the time period of this study, there are measurements in India that should be compared to (see previous point), as these are very related to the conclusions made here (i.e. about regional sources, etc). Any comparisons made to other surface data are very minimal at present. The comparisons to CARIBIC, satellites, etc are important but to a lesser degree than other Indian surface observations.

3. The authors should be careful throughout the text to maintain that the mechanisms proposed for the various features in the data set are still speculative. This is a measurement-led study and without additional tools to quantitatively pinpoint the sources of air masses, these remain as hypotheses. An example of this would be on page 7187 line 21: “Moreover, the mean CH<sub>4</sub> seasonal cycle at HLE agrees well with the annual variation of convective precipitation over the Indian subcontinent (Fig. 5b), which is derived from ECMWF nudged Laboratoire de Météorologie Dynamique general circulation model (LMDz) (Hauglustaine et al., 2004). This agreement indicates that the summer maximum at HLE can be attributed to the enhanced biogenic CH<sub>4</sub> emissions from wetlands and rice paddies and deep convection that mixes surface emissions into the mid-to-upper troposphere.” There is not enough information to say conclusively that biogenic emissions are responsible for the summer maximum without additional data (i.e. though models or isotopic data). So while the mechanism is proposed, it is stated too definitively. There are several statements like this throughout the text, which should be toned down and the authors should rephrase or remove statements such as this one.

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4. Following up on the above statement, there are some sections, which are still quite speculative and not necessarily based on evidence and should be removed. These include: (a) Section 3.3 on elevated CH<sub>4</sub> and CO samples at PBL – There is not enough information to ascertain whether the samples at BKT are related to the samples at PBL. There would need to be a model simulation to show that the air mass at BKT on e.g., Sep 8 2009, arrived at PBL on Sep 16 2009. Otherwise it is too speculative and should be removed. (b) Discussion of bimodal H<sub>2</sub> on page 7196 line 17 – it is speculated the biomass burning from each hemisphere is the source of the double peaks. But there is no evidence to show that is the case.

5. Many conclusions are drawn about Indian fluxes using HLE. However, from the text and looking at the trajectories, HLE mainly samples air from Africa and the Middle East. There are only a few trajectories that sample Indian air masses. It seems that the conclusions to the HLE data (with regards to Indian sources) should be changed to reflect this. Can HLE be used to discuss Indian sources?

6. There appear to be some discrepancies in the text. The use of CARIBIC data and other remotely sensed data seems contradictory in places. In the discussion for SF<sub>6</sub>, it states that the CARIBIC samples are more representative of westerly jet transport rather than the SW monsoon. However, CARIBIC is used in the discussion for all other species in the context of Indian sources. It would also be useful to see trajectories for the comparison data to know whether they are sampling the same air masses.

7. PON is located in a large urban area. While sampling is done between 1200-1800, the site would still be affected by local emissions. The analysis using PON for gradients between other sites could potentially be complicated by the fact that the site is impacted by local emissions. Therefore, PON may not be the best site to use for trend analysis. Can the authors comment on this? Could CO be used as a tracer for local emissions?

Specific comments:

Page 7173 line 15: change 'dominant' to 'likely' source of emissions

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Page 7173 line 18-19: sentence needs restructuring. Suggest 'to better constrain the GHG budget at regional and continental scales'

Introduction first paragraph: Text should state that the emissions from EDGAR, etc are using bottom-up methods, which generally have large uncertainties, and therefore top-down studies are needed as well.

Page 7174 line 11: what percent are natural emissions?

Page 7174 lines 15 – 17: Monitoring is not required by the UNFCCC.

Page 7175 lines 1-23: Following general comment above, a review of other surface measurements in India is needed in this paragraph. 'Besides a lack of observation sites' I agree that sites are sparse but they are not discussed.

Page 7176 line 25: It is not possible to tell from the trajectories, what altitude these air masses originated from. HLE, for example, likely does not always sample surface emissions. It would be useful to see what altitude all of the sites are sampling. Also this would make comparison to aircraft observations easier to interpret.

Page 7177 line 17: manuscripts in preparation should not be cited

Page 7177 line 21: It looks like there are very few HLE trajectories coming from South Asia. Can the authors comment on the use of this site for regional work?

Page 7178 line 6: Do the sea breezes necessarily imply that they will be clean air masses? For example during the SW monsoon, the sea breeze will be a local effect on a dominant southwesterly flow. At PON, does this mean that air masses could still contain "local" emissions albeit the wind direction coming from the sea?

Page 7178 lines 5-7: Can CO be used as a tracer of local emissions for additional filtering for local emissions?

Page 7179 lines 19-23: Are flasks filled manually or automatically at a given time? Does an operator decide when conditions are correct for filling?

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Page 7180 line 19: Is there any impact of CO<sub>2</sub> on N<sub>2</sub>O concentrations through this method? It is known that CO<sub>2</sub> can “dope” the signal for N<sub>2</sub>O on an ECD.

Page 7180: No description of the ECD or RDG setup (temps, flow rates) or information about carrier gases or calibration scales. Perhaps a table could provide all of the measurement info for each detector concisely.

Page 7181 line 28: What are sampling uncertainties due to? local influence, human error?

Page 7183 lines 2-3: Were the biases corrected?

Page 7184 line 12: ‘additionally’ should be ‘additional’

Page 7185 line 12: HLE and CONTRAIL flights over New Delhi would likely be sampling different air masses, with HLE mostly seeming to sample air from the Middle East. Which altitude in the CONTRAIL profile represents the same air mass as HLE? Trajectories would be useful.

Page 7185 line 25: Again, it does not seem that HLE received many air masses from South Asia from the trajectories in Figure 1

Page 7185 line 28: KZM and WLG, if they are more affected by northern air, then they would show a greater amplitude of the seasonal cycle. Can the authors comment?

Page 7187 line 13: it seems that CARIBIC samples during the monsoon would not take one month to mix during this time of deep convection. Can the authors justify this statement? Also, why does vertical mixing lead to a larger seasonal cycle amplitude than HLE?

Page 7187 line 16: Remove ‘apparently’

Page 7187 lines 22-28: This discussion is too speculative and should be removed without further evidence (e.g., isotopic). There is not enough information to state that biogenic CH<sub>4</sub> emissions are responsible for the summer max at HLE. Furthermore, a

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model is needed to disentangle the meteorology from emissions.

Page 7187 line 29: be more specific - concentrations of trace gases would be enhanced at higher altitudes rather than the surface.

Page 7188 line 3: Earlier it is stated that KZM and WLG sample wetland emissions from the north. But here it is stated that their CH<sub>4</sub> increases are smaller because they are not influenced by deep convection. Does that necessarily imply that the increases will be smaller? There could be a large summer methane signal from wetlands.

Page 7188 line 10: Why does PON not sample surface emissions? The trajectories during July look like they pass over southern India.

Page 7189 line 18: Why would it be argued that N<sub>2</sub>O is ‘more noisy than CO<sub>2</sub> and CH<sub>4</sub> due to regional sources synoptic variability’? Also, N<sub>2</sub>O measurement has lower signal to noise (i.e. precision is lower than CO<sub>2</sub> and CH<sub>4</sub>).

Page 7190 line 7: CARIBIC enrichment only during monsoon – why April-December 2008?

Page 7191 line 24: Even if there were no SF<sub>6</sub> emissions (rather than weak SF<sub>6</sub> emissions), this would imply that sites should follow the background. This still doesn’t explain why there is a negative gradient.

Page 7192 lines 14-18: It is mentioned here that CARIBIC samples different air masses to HLE. It is unclear therefore why the CARIBIC comparison is made for CH<sub>4</sub> and N<sub>2</sub>O. This seems like a contradiction and so perhaps CARIBIC comparison should be removed for CH<sub>4</sub> and N<sub>2</sub>O as well for HLE.

Page 7193 line 23: It is difficult to see a one month lag in Fig 11.

Page 7193 line 16: Could the larger variability also be due to local sources?

Page 7196 lines 19-25: This discussion about bimodal H<sub>2</sub> seasonal cycle being due to biomass burning is very speculative and should be removed. There is not enough

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information or model runs to demonstrate that this is the case.

Page 7197 line 21: Describe why anthropogenic CO emissions are lower in summer than winter?

Page 7198 line 3: Why would uplift contribute to maximum CH<sub>4</sub>/CO ratio, as both species are uplifted together?

Section 3.3: This section is too speculative, as the appropriate model simulations have not been performed to assess whether the elevated events are related to elevations at BKT. With the model simulation, linking the time and position of the elevated event at BKT with the time and position of the elevated event at PBL, this section should be removed.

Conclusions page 7204 line 8 – The summertime maximum being attributed definitively to biogenic emissions is too speculative without other information. The authors should tone down the statement.

Figures 3,5,7 etc should show uncertainties, from measurement uncertainty, sampling uncertainty and if averaged into seasonal cycle, the variability in the seasonal cycle. This would provide an indication for the significance of the seasonal cycle. Some panels in figures contain uncertainties, some do not.

Supplement - Back trajectories for comparison data (KZM, WLG) should also include trajectories for CARIBIC, etc.

Supplement - A description of KZM and WLG is needed. Are they mountain sites, etc?

Short title - Change to 'Five years (plural) of flask measurements'

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 7171, 2015.