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

Anonymous Referee #2

General comments:

The paper presents a new record of flask sample observations of long-lived trace gases from two sites in mainland India and one site on the Andaman Islands. Data from the two mainland sites are presented from 2007 to 2011 and from the island site from 2009 to 2011. The target sampling frequency is weekly, though there are significant gaps in the record, sometimes weeks/months, due to bad weather and technical problems (see figure S2 in the supplement). The target compounds are CO_2, CH_4, N_2O, SF_6, CO and H_2. The paper describes in detail the sampling and analysis stages and the data processing techniques used to generate annual means, seasonal cycles and gradients between stations. The paper compares these new flask data with observations from stations in other countries such as Kazakhstan and China. An attempt is made to relate the seasonal cycles and gradients between stations to variations in natural GHG fluxes, anthropogenic emissions and monsoon circulations.

The Indian continent is currently experiencing rapid industrial development and as such is an important region globally for emissions of these long-lived trace gases. There appears to be a paucity of ground-based GHG measurements from this region and these observations go someway to filling the data gap. The authors should be encouraged to continue their observations and where possible improve future measurement frequency/reliability to avoid large data gaps. Have the authors considered analysing the flask samples for stable carbon isotope ratio which would be a big help in determining source apportionment especially for CH_4 and CO_2?

I recommend that this paper be published after consideration to the following minor specific comments and technical corrections.

An interactive comment indicated that a site in India at Sinhagad has been collecting samples for CO_2 and CH_4 analysis since 2009 (Y.K. Tiwari et al, 2014). The authors do refer to this paper in the conclusions (p 7204, line23) but could also refer to it earlier in the CO_2/CH_4 discussion section.

Specific comments (individual scientific questions/issues):

1 Introduction, page 7174, line 1: it would be useful to compare the estimated increase in GHG emissions from India (1.4 to 2.8 GtCO2 eq) with the estimate from Europe or the USA.

1 Introduction, page 7174, line 17: suggest to add a sentence defining what is meant by the top-down and bottom-up approaches.

1 Introduction, page 7174, line 24: suggest to extend the sentence starting ‘Notably, these ...’ to include a comparison of Indian bottom-up uncertainties with those from say Western Europe where inventories are more accurate.

1 Introduction, page 7175, line 17: suggest to add here an extra sentence describing key meteorological features of the NE winter monsoon.

2.1 Sampling stations, page 7176, line 15: I would like to see a map, possibly as an extra panel in Fig.1 centred and zoomed on India showing the three sampling stations and terrain.

2.1 Sampling stations, page 7177, line 8: suggest to add sentence somewhere here describing the inlet location, height above the ground, type of inlet tubing used etc.

2.1 Sampling stations, page 7178, line 19: suggest to add an extra sentence detailing the inlet location, inlet height above ground, type of tubing sed etc.

2.2.1 Flask sampling, page 7179, lines 10-14: is the loss correction the same for both valve types used? If so please state in text.

2.2.1 Flask sampling, page 7179, lines 15-17: has the magnesium perchlorate drier been tested for loss of the target compounds?

2.2.2 Flask analyses, page 7180, lines 2-4: is the pressure inside each flask on arrival at LSCE the same as it was after filling in India? Some concern about loss/leakage during air-freight.

2.2.2 Flask analyses, page 7180, line 6: is this the Agilent micro-cell ECD? If so please state.

2.2.2 Flask analyses, page 7180, line 14: are samples flushed through the sample loop under the pressure inside the flask or is a pump used? If so, is the pump upstream or downstream of the sample loops?

2.2.2 Flask analyses, page 7180, lines 17-22: please also state column flow rates, oven temperature, isothermal or oven program etc.

2.2.2 Flask analyses, page 7180, line 25: please include more detail on the ECD detector temperature, make-up gas flow etc.

2.2.2 Flask analyses, page 7181, line 1: please include RGD detector temperature.

2.2.2 Flask analyses, page 7181, lines 2-7: are the working calibration cylinders filled with synthetic air or ambient air? Please state if they are filled by LSCE or purchased (then include supplier details). Also please state make and model of gas regulator used on the calibration cylinders.

2.2.2 Flask analyses, page 7181, line 10: please name the international calibration scale used.

2.3.2 Ratio of Species, page 7184, lines 2-7: please expand this section by adding a couple of sentences giving more detail of the procedure used.

3.1.2 CH_4, page 7187, line 1: the low observations of CH_4 at PBL in summer 2009 and 2011 are striking and should be mentioned here. Presumably the air arriving at PBL at this time of year has southern hemisphere origin, arriving on the SW monsoon flow.

3.1.3 N_2O, page 7188, lines 16-18: suggest to compare the observed N_2O growth rate at HLE to say AGAGE northern hemisphere average growth rate.

3.1.4 SF_6, page 7192, lines 1-2: there is also a strong possibility that some of the episodic SF_6 pollution events originate in China.

3.1.5 CO, page 7194, lines 26-28: China should also be considered as an influence on CO enriched air-masses arriving at PBL during NE monsoon.

4 Conclusions, page 7203, lines 7-9: This sentence needs some qualification as it implies a five year history of observations at PBL when in fact only 2.5 years are available at that site with a long data break during 2010. The site at PBL is somewhat under-sampled in relation to HLE and PON.

4 Conclusions, page 7204, lines 17-26: In this paragraph the authors could mention any plans to continue flask sampling and if possible extend the measurement suite. Although in-situ continuous measurement techniques are hard to deploy reliably in these remote tropical locations this would add considerably to the value of the sites. The authors could also consider adding stable carbon isotope ratio analysis to the flask measurement process which help with source apportionment, especially for CO_2 and CH_4.

Figure 14, page 7235: I suggest to also give in each panel the number of samples used to create each fitting line, there are noticeably fewer points available for PBL. Same also for Fig.15 and Fig. 16.

Technical corrections (typing errors, etc.):

1 Introduction, page 7173, line 24: '... during ~~the~~ recent decades ...'

1 Introduction, page 7174, line 5 to 7: remove brackets from '... (in 2010, the per capita...)' and make new sentence starting: 'For comparison, in 2010, the per capita GHG emission rates ...'

1 Introduction, page 7174, line 8: '... ~~agriculture-related~~ ...' replace with '... agricultural ...'

1 Introduction, page 7174, line 12: try to improve this sentence, e.g. 'Reducing emissions of these two non-CO_2 greenhouse gases may offer a more cost-effective way to mitigate future climate change than by attempting to directly reduce CO_2 emissions (Montzka et al., 2011)'

1 Introduction, page 7176, line 8: suggest to re-arrange this sentence to remove brackets, for e.g. 'We examine synoptic variations of CO_2, CH_4 and CO by analysing co-variances between species, using deviations from their smoothed fitting curves (Sect. 3.2).'

2.1 Sampling stations, page 7177, line 4: suggest to give HLE lat/lon co-ordinates to three decimal places. Same also for PON and PBL, with only two decimal places these two sites appear to be offshore.

2.1 Sampling stations, page 7177, line 15: suggest to re-arrange sentence for ease of reading, e.g ' ... background free tropospheric air masses in the northern mid-latitudes.'

2.1 Sampling stations, page 7177, line 26: suggest to modify sentence, e.g. 'The flask sampling inlet, ~~was~~ initially located on a 10m mast fixed on the roof of the University Guest House, was later moved to a 30 m high tower in June 2011.' Also give the type of inlet tubing used.

2.1 Sampling stations, page 7178, line 7: suggest to combine the sentences starting 'Flask sampling ...' and 'Over the period ...', how about: 'Flask sampling began in September 2006 and over the period 2007–2011, a total of 185 flask sample pairs were collected at the site.'

2.2.2 Flask analyses, page 7180, line 4: '... HP~~8~~6890 ...'

2.2.2 Flask analyses, page 7180, line 18: '... 3/16" ...', is this the internal or external column diameter? Please state. Same on line 19 and 20. Use either " or inches, both are used in text.

3.1.2 CH_4, page 7188, line 7: '~~This~~ These not only ...'

3.1.3 N_2O, page 7188, line 25: 'We also ~~analyze~~ analyzed ...'

3.1.3 N_2O, page 7190, line 11-13: Improve sentence starting 'One reason may be ...', for example: 'One reason may be that air arriving at the site during the SW monsoon period is relatively enriched in N_2O compared to CH_4, reflecting differences in their relative emissions along the air mass history.'

3.1.4 SF_6, page 7192, line 22: delete 'southwesterly' which would otherwise imply winds from the SW. Also consider China as well as Southeast Asia to explain some of the polluted air masses.

3.1.6 H_2, page 7195, line 27: 'The mean H_2 seasonal ~~cycle~~ cycles ...'

3.2.1 $\Delta CH_4/\Delta CO$, page 7198, line 10: add 'that', so: '... estimates are 1.5 to 4 times that of the ...'

Table 1, page 7220: define RSD abbreviation in the caption, same for D_{max} and D_{min}. Consider also adding additional row after the trend row for each compound, giving northern hemisphere average trends say from the AGAGE network.

Figure 2, page 7223: use "open circles" and "crosses" in the caption description rather than "o" and "x". Same throughout the text, figure captions and in supplement.

Figure 3, page 7224: all three panels are too small.

Figure 4, page 7225: Needs full caption, not just 'same as Fig. 2 ...'

Figure 8, page 7229: Needs full caption, not just 'same as Fig. 6 ...'

Figure 9, page 7230: Needs full caption, not just 'same as Fig. 7 ...'. Also can the uncertainty shaded areas be made clearer?

Figure 10, page 7231: Needs full caption, not just 'same as Fig. 2 ...'

Figure 12, page 7233: Needs full caption, not just 'same as Fig. 2 ...'

Figure 13, page 7233: Needs full caption, not just 'same as Fig. 11 ...'

Figure 15, page 7236: Needs full caption, not just 'same as Fig. 14 ...'

Figure 16, page 7237: Needs full caption, not just 'same as Fig. 14 ...'

Supplement, Table S5: what is the reason for the grey shaded columns? Different instrument network? Same for Table S7 and Table S9.

Supplement, Figure S3: can the shaded uncertainty areas be made clearer?

Supplement, Figure S7 caption: ‘ MHD, BGU, FIK~~, FIK~~ and LPO ...’
