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## ***Interactive comment on “Temporal variations in CO<sub>2</sub> and CO at Ahmedabad in western India” by N. Chandra et al.***

**N. Chandra et al.**

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Anonymous referee #2

General: We are very much grateful to the referee for appreciating our work and giving very helpful suggestions and comments, which have significantly improved the manuscript. We have revised the manuscript by carefully taking into account all the comments point by point. Text in red and blue colour show the questions and answers, respectively.

General comments: This paper addresses temporal variations of atmospheric CO<sub>2</sub> and CO in an urban site in western India. There are not so many studies on greenhouse gases in urban environments. Furthermore, such study is rare in countries in

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development. This work is interesting to be published and is fully within the scope of ACP.

The authors address the seasonal and the diurnal scales, as well as CO/CO<sub>2</sub> ratios from which they infer information on the anthropogenic vs natural sources of CO and CO<sub>2</sub>. They also propose a calculation of CO emissions for the studied city using such ratio and CO<sub>2</sub> emissions from inventory, as well as a short model/observations comparison. I acknowledge the large amount of work provided by the authors and interesting information issued from this study.

Response: We are very much grateful to the referee for the positive comments and appreciation of our work. We have tried our best to give all the answers point by point.

However, there are also some major issues to be addressed and reviewed before publication in ACP. These issues concern:

1. The form: The text is quite difficult to read and needs to be synthetized, especially the introduction, the seasonal study and the diurnal study. Some sentences are even repeated twice.

Response: According to the suggestion, we have synthetized all mentioned sections very carefully after removing, adding and rearranging sentences. Please note that, rather than adding, removing and rearranging sentences in between the written text, we have added modified text of whole section in new paragraph. The modified sections are given below.

Introduction: Carbon dioxide (CO<sub>2</sub>) is the most important anthropogenically emitted greenhouse gas (GHG) and has increased substantially from 278 to 390 parts per million (ppm) in the atmosphere since the beginning of the industrial era (circa 1750). It has contributed to more than 65% of the radiative forcing increase since 1750 and hence has significant impact on the climate system (Ciais et al., 2013). Major causes of CO<sub>2</sub> increase are anthropogenic emissions, especially fossil fuel combustion, cement

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production and land use change. Land and oceans are the two important sinks of atmospheric CO<sub>2</sub>, which remove about half of the anthropogenic emissions (Le Quéré et al., 2014). The prediction of future climate change and its feedback rely mostly on our ability to quantify fluxes of greenhouse gases, especially CO<sub>2</sub>, at regional (100-1000 km<sup>2</sup>) and global scales. Though the global fluxes of CO<sub>2</sub> can be estimated fairly well, the regional scale (fluxes are associated with quite high uncertainty, especially over the South Asian region and the estimation uncertainty being larger than the value itself (Patra et al., 2013; Peylin et al., 2013). Detailed scientific understanding of the flux distributions is also needed for formulating effective mitigation policies.

Along with the need for atmospheric measurements for predicting the future levels of CO<sub>2</sub>, quantifying the components of anthropogenic emissions of CO<sub>2</sub> is similarly important for providing independent verification of mitigation strategies as well as understanding the biospheric component of CO<sub>2</sub>. Only CO<sub>2</sub> measurements cannot be helpful in making such study due to the larger role of biospheric fluxes in its atmospheric distributions. The proposed strategy for quantification of the anthropogenic component of CO<sub>2</sub> emissions is to measure simultaneously anthropogenic tracers,. CO can be used as a surrogate tracer for detecting and quantifying anthropogenic emissions from burning processes, since it is a major product of incomplete combustion (Turnbull et al., 2006; Wang et al., 2010). The vehicular as well as industrial emissions contribute large fluxes of CO<sub>2</sub> and CO to the atmosphere in urban regions. Several grounds and aircraft based simultaneous studies of CO and CO<sub>2</sub> have been made in the past from different parts of the world (Turnbull et al., 2006; Wunch et al., 2009; Wang et al., 2010; Newman et al., 2013) but such a study is lacking in India except recently reported results from weekly samples for three Indian sites by Lin et al. (2015).

Measurements at different regions (eg. rural, remote, urban) and at different frequency (eg., weekly, daily, hourly etc) have their own advantage and limitations. For example, the measurements at remote locations at weekly interval can be useful for studying seasonal cycle, growth rate, and estimating the regional carbon sources and sinks af-

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ter combining their concentrations with inverse modelling and atmospheric tracer transport models. However, some important studies, like diurnal variations, temporal covariance...etc are not possible from these measurements. Analysis of temporal covariance of atmospheric mixing processes and variation of flux on shorter time scales, e.g., sub-daily, is essential for understanding local to urban scale CO<sub>2</sub> flux variations (Ahmadov et al., 2007; Pérez-Landa et al., 2007; Briber et al., 2013; Lopez et al., 2013; Ammoura et al., 2014; Ballav et al., 2015). Urban regions contribute about 70% of global CO<sub>2</sub> emission from anthropogenic sources and further projected to increase over the coming decades. Therefore, measurements from these regions are very helpful for understanding emissions, growth as well as verifying the mitigation policies. The first observations of CO<sub>2</sub>, CO and other greenhouse gases started in February 1993 from Cape Rama (CRI: a coastal site) on the south-west coast of India using flask samples (Bhattacharya et al., 2009). After that, several other groups have initiated the measurements of surface level greenhouse gases (Mahesh et al., 2014; Sharma et al., 2014; Tiwari et al., 2014; Lin et al., 2015). Most of these measurements are made at weekly or fortnightly time intervals. Two aircraft based measurement programs, namely, Civil Aircraft for the regular Investigation of the atmosphere Based on an Instrument Container (CARIBIC) (Brenninkmeijer et al., 2007) and Comprehensive Observation Network for TRace gases by AirLiner (CONTRAIL) (Machida et al., 2008) have provided important first look on the South Asian CO<sub>2</sub> budget, but these data have their own limitations (Patra et al., 2011; Schuck et al., 2010, 2012). It is pertinent to mention here that till now, there are no reports of CO<sub>2</sub> measurements over the urban locations of the Indian subcontinent, which could be an important player in the global carbon budget as well as mitigation purpose due to strong growing anthropogenic activities specifically fast growing traffic sector and sinks (large areas of forests and croplands). Hence, the present study is an attempt to reduce this gap by understanding the CO<sub>2</sub> mixing ratios in light of its sources and sinks at an urban region in India.

In the view from above, simultaneous continuous measurements of CO<sub>2</sub> and CO have been made since November 2013 from an urban site Ahmedabad located in the west-

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ern India using very highly sensitive laser based technique. The preliminary results of these measurements for one month period have been reported in (Lal et al., 2015). These detailed measurements are utilized for studying the temporal variations (diurnal and seasonal) of both gases, their emission characteristics on diurnal and seasonal scale using their mutual correlations, estimating the contribution of vehicular and biospheric emission components in the diurnal cycle of CO<sub>2</sub> using the ratios of CO to CO<sub>2</sub> and rough estimate of the annual CO emissions from study region. Finally, the measurements of CO<sub>2</sub> have been compared with simulations using an atmospheric chemistry-transport model to discuss roles of various processes contributing to CO<sub>2</sub> concentration variations.

**Seasonal cycle of CO<sub>2</sub> and CO:** The seasonal cycles of CO<sub>2</sub> and CO are mostly governed by the strength of emission sources, sinks and transport patterns. Although they follow almost identical seasonal patterns, but the factors responsible for their seasonal behaviours are distinct as for the diurnal variations. We calculate the seasonal cycle of CO<sub>2</sub> and CO using two different approaches. In the first approach, we use the monthly mean of all measurements and in the second approach we use monthly mean of the afternoon period (1200-1600 hrs) measurements only. The seasonal cycle from first approach will present the overall variability in both gases. On the other hand, second approach removes the auto covariance by excluding CO<sub>2</sub> and CO data mainly affected by local emission sources and represent seasonal cycles at the well mixed volume of the atmosphere. The CO<sub>2</sub> time series is detrended by subtracting a mean growth rate of CO<sub>2</sub> observed at Mauna Loa (MLO), Hawaii, i.e., 2.13 ppm yr<sup>-1</sup> or 0.177 ppm/month ([www.esrl.noaa.gov/gmd/ccgg/trends/](http://www.esrl.noaa.gov/gmd/ccgg/trends/)) for clearly depicting the seasonal cycle amplitude.

In general, total mean values of CO<sub>2</sub> and CO are observed lower in July having concentration  $398.78 \pm 2.8$  ppm and  $0.15 \pm 0.05$  ppm respectively. During summer monsoon months predominance of south-westerly winds which bring cleaner air from the Arabian Sea and the Indian Ocean over to Ahmedabad and high VC (Figure 1) are

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mostly responsible for the lower concentration of total mean of both the gases. CO<sub>2</sub> and CO concentrations are also at their seasonal low in the northern hemisphere due to net biospheric uptake and seasonally high chemical loss by reaction with OH, respectively. In addition, deep convections in the summer monsoon season efficiently transport the Indian emission (for CO, hydrocarbons) or uptake (for CO<sub>2</sub>) signals at the surface to the upper troposphere, resulting lower concentrations at the surface in the as compared to the winter months (Kar et al., 2004; Randel and Park, 2006; Park et al., 2009; Patra et al., 2011; Baker et al., 2012). During autumn and early winter (December), lower VC values cause trapping of anthropogenically emitted CO<sub>2</sub> and CO and is the major cause for high concentrations of both gases during this period. In addition to this, wind changes from cleaner marine region to polluted continental region, especially from IGP region and hence could be additional factor for higher levels of CO<sub>2</sub> and CO during these seasons (autumn and winter). Elevated levels during these seasons are also examined in several other pollutants over Ahmedabad as discussed in previous studies (Sahu et al., 2006, Mallik et al., 2016). Maximum concentrations of CO<sub>2</sub> and CO are observed to be  $424.85 \pm 17$  ppm and  $0.83 \pm 0.53$  ppm, respectively, during November. From January to May the total mean concentration of CO<sub>2</sub> decreases from  $415.34 \pm 13.6$  to  $406.14 \pm 5.0$  ppm and total mean concentration of CO decreases from  $0.71 \pm 0.22$  to  $0.22 \pm 0.10$  ppm. Higher VC and predominance of comparatively less polluted mixed air masses from oceanic and continental region results in lower total mean concentrations of both gases during this period.

There are some clear differences which are observed in the afternoon mean concentrations of CO<sub>2</sub> as compared to daily mean. The first distinct feature is that significant difference of about 5 ppm is observed in the afternoon mean of CO<sub>2</sub> concentration from July to August as compared to the difference in total mean concentration about  $\sim 0.38$  ppm for the same period. Significant differences in the afternoon concentrations of CO<sub>2</sub> from July to August is mainly due to the increasing sink by net biospheric productivity after the Indian summer monsoonal rainfall. Another distinct feature is that the daily mean concentration of CO<sub>2</sub> is found higher in November while the afternoon

mean concentration of CO<sub>2</sub> attain maximum value ( $406 \pm 0.4$  ppm) in April. Prolonged dry season combined with high daytime temperature (about 41°C) during April-May make the tendency of ecosystem to become a moderate source of carbon exchange (Patra et al., 2011) and this could be responsible for the elevated mean noon time concentrations of CO<sub>2</sub>. Unlike CO<sub>2</sub>, seasonal patterns of CO from total and afternoon mean concentrations are identical, although levels are different. It shows that the concentrations of CO is mostly governed by identical sources during day and night time throughout the year.

The average amplitude (max - min) of the annual cycle of CO<sub>2</sub> is observed around 13.6 and 26.07 ppm from the afternoon mean and total mean respectively. Different annual cycles and amplitudes have been observed from other studies conducted over different Indian stations. Similar to our observations of the afternoon mean concentrations of CO<sub>2</sub>, maximum values are also observed in April at Pondicherry (PON) and Port Blair (PBL) with amplitude of mean seasonal cycles about  $7.6 \pm 1.4$  and  $11.1 \pm 1.3$  ppm respectively (Lin et al., 2015). Cape Rama (CRI), a coastal site on the south-west coast of India show the seasonal maxima one month before than our observations in March annual amplitude about 9 ppm (Bhattacharya et al., 2009). The Sinhgad (SNG) site located over the Western Ghats Mountains, show very larger seasonal cycle with annual amplitude about 20 ppm (Tiwari et al., 2014). The amplitude of mean annual cycle at the free tropospheric site Hanle at altitude of 4500 m is observed to be  $8.2 \pm 0.4$  ppm, with maxima in early May and the minima in mid-September (Lin et al., 2015). Distinct seasonal amplitudes and patterns are due to differences in regional controlling factors for the seasonal cycle of CO<sub>2</sub> over these locations, e.g., the Hanle is remotely located from all continental sources, Port Blair site sampled predominantly marine air, Cape Rama observes marine air in the summer monsoon season and Indian flux signals in the winter, and Sinhgad represents a forested ecosystem. These comparisons show the need for CO<sub>2</sub> measurements over different ecosystems for constraining its budget.

The annual amplitude in afternoon and daily mean CO concentration is observed to

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be about 0.27 and 0.68 ppm, respectively. The mean annual cycles of CO over PON and PBL show the maxima in the winter months and minima in summer months same as our observations with annual amplitudes of  $0.078 \pm 0.01$  and  $0.144 \pm 0.016$  ppm, respectively. So the seasonal levels of CO are affected by large scale dynamics which changes air masses from marine to continental and vice versa and by photochemistry. The amplitudes of annual cycle at these locations differ due to their climatic conditions and sources/sinks strengths.

Diurnal variation of CO<sub>2</sub> : Figure 5a shows the mean diurnal cycles of atmospheric CO<sub>2</sub> and associated 1- $\sigma$  standard deviation (shaded region) during all the four seasons. All times are in Indian Standard Time (IST), which is 5.5 hrs ahead of Greenwich Mean Time (GMT). Noticeable differences are observed in the diurnal cycle of CO<sub>2</sub> from season to season. In general, maximum concentration has been observed during morning (0700-0800 hrs) and evening (1800-2000 hrs) hours, when ABL is shallow, traffic is dense and vegetation respiration dominate due to absence of photosynthetic activity. The minimum of the cycles occurred in the afternoon hours (1400-1600 hrs), when PBL is deepest and well mixed as well as when the vegetation photosynthesis is active. There are many interesting features in the period of 0000-0800 hrs. CO<sub>2</sub> concentrations start decreasing from 0000 to 0300 hrs and increases slightly afterwards till 0600-0700 hrs during summer and autumn. Respiration of CO<sub>2</sub> from the vegetation is mostly responsible for this night time increase. During winter and spring seasons CO<sub>2</sub> levels are observed constant during night hours and small increase is observed only from 0600 to 0800 hrs during the winter season. While in contrary to this, subsequent section shows a continuous decline in the night time concentrations of main anthropogenic tracer CO, which indicates that there is enough vertical mixing of low CO air from above that once the CO source is turned off, its concentration drops. Hence, constant levels of CO<sub>2</sub> at night hours during these seasons gives the evidence of a continued but weak sources (such as respiration) in order to offset dilution of mixing of low CO<sub>2</sub> air from aloft. Dry soil conditions could be one of the possible caused for weak respirations. Further, distinct timings have been observed in the morning peak of CO<sub>2</sub>

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during different seasons. It is mostly related to the sunrise time, which will decide the evolutions time of PBL height and beginning of vegetation photosynthesis. The sunrise occurs at 0555-0620 hrs, 0620-0700 hrs, 0700-0723 hrs and 0720-0554 hrs during summer, autumn, winter and spring, respectively. During spring and summer, rush hour starts after sunrise, so the vehicular emissions occur when the PBL has been already high and photosynthetic activity has begun. The CO<sub>2</sub> concentration is observed lowest in the morning during the summer season as compared to other seasons. This is because CO<sub>2</sub> uptake by active vegetation deplete entire mixed layer during day time and when the residual layer mixes to the surface in the morning, low-CO<sub>2</sub> air is mixed down. In winter and autumn, rush hour starts parallel with the sunrise, so the emissions occur when the PBL is low and concentration builds up is much stronger in these seasons than in spring and summer seasons.

The diurnal amplitude is defined as the difference between the maximum and minimum concentrations of CO<sub>2</sub> in the diurnal cycle. The amplitudes of monthly averaged diurnal cycle of CO<sub>2</sub> from July, 2014 to May 2015 are shown in Figure 5b. The diurnal amplitude shows large month to month variation with increasing trend from July to October and decreasing trend from October onwards. The lowest diurnal amplitude of about 6 ppm is observed in July while highest amplitude about 51 ppm is observed in October. The amplitude does not change largely from December to March and is observed in the range of 25-30 ppm. Similarly, from April to May the amplitude also varies in a narrow range from 12 to 15 ppm. The jump in the amplitude of the CO<sub>2</sub> diurnal cycle is observed highest (around 208%) from July to August. This is mainly due to significant increase of biospheric productivity from July to August after the rains in Ahmedabad. It is observed that during July the noon time CO<sub>2</sub> levels are found in the range of 394-397 ppm while in August the noon time levels are observed in the range of 382-393 ppm. The lower levels could be due to the higher PBL height during afternoon and cleaner air, but in case of CO (will be discussed in next section), average day time levels in August are observed higher than in July. It rules out that the lower levels during August are due to the higher PBL height and presence of cleaner marine

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air, and confirms the higher biospheric productivity during August. Near surface diurnal amplitude of CO<sub>2</sub> has been also documented in humid subtropical Indian station Dehradun and a dry tropical Indian station Gadanki (Sharma et al., 2014). In comparison to Ahmedabad, both these stations show distinct seasonal change in the diurnal amplitude of CO<sub>2</sub>. The maximum CO<sub>2</sub> diurnal amplitude of about 69 ppm is observed during the summer season at Dehradun (30.3oN, 78.0oE, 435m), whereas maximum of about 50 ppm during autumn at Gadanki 400 (13.5oN, 79.2oE, 360 m).

Diurnal variation of CO: Figure 7a shows seasonally averaged diurnal variation of CO. In general, the mean diurnal cycle of CO show lower concentration during noon (1200-1700 hrs) and two peaks in the morning (0800 to 1000 hrs) and in the evening (1800 to 2200 hrs) hours. This cycle exhibits the same pattern as the mean diurnal cycle of traffic flow, with maxima in the morning and at the end of the afternoon, which suggests the influence of traffic emissions on CO measurements. Along with the traffic flow, PBL dynamics also play a critical role in governing the diurnal cycle of CO. The amplitudes of the evening peak in diurnal cycles of CO are always greater than the morning peaks. It is because the PBL height evolves side by side with the morning rush hours traffic and hence increased dilution, while on the other hand, during evening hours, PBL height decrease along with evening dense traffic and favours accumulation of pollutants until the late evening under the stable PBL conditions. The noon time minimum of the cycle is mostly associated with the deepest and well mixed PBL.

In general, the average diurnal cycle patterns of both gases (CO<sub>2</sub> and CO) are similar, but having few noticeable differences. The first difference is observed in the timing of the occurrence of morning peaks: CO<sub>2</sub> peaks occur slightly before than CO peak due to the triggering photosynthesis process by the sunrise. On the other hand, the morning peaks of CO mostly depend on the rush hour traffic and are consistent at 0800-1000 hrs in all seasons. The second difference is that the afternoon concentrations of CO show little seasonal spread as compared to the afternoon concentrations of CO<sub>2</sub>. Again, this is due to the biospheric control on the levels of CO<sub>2</sub> during the afternoon hours

of different seasons while CO levels are mainly controlled by the dilution during these hours. The third noticeable difference is that the levels of CO decrease very fast after evening rush hours in all seasons while this feature is not observed in the case of CO<sub>2</sub> since respiration during night hours contributes to the levels of CO<sub>2</sub>. The continuous drops of night time concentrations of CO indicates that there is enough vertical mixing of low CO air from above that once the CO source is turned off. The average morning (0800-0900 hrs) peak values of CO is observed minimum ( $0.18 \pm 0.1$  ppm) in summer and maximum ( $0.72 \pm 0.16$  ppm) in winter while evening peak shows minimum value ( $0.34 \pm 0.14$  ppm) in summer and maximum ( $1.6 \pm 0.74$  ppm) in autumn. The changes in CO concentrations show large fluctuations from morning peak to afternoon minima and from afternoon minima to evening peak. From early morning maxima to noon minima, the changes in CO concentrations are found in the range of 20 -200%, while from noon minima to late evening maxima the changes in CO concentrations are found in the range of 85% to 680%. Similar diurnal variations with two peaks have also been observed in earlier measurements of CO as well as NO<sub>x</sub> at this site (Lal et al., 2000).

The evening peak contributes significantly to the diurnal amplitude of CO. The largest amplitude in CO cycle is observed in autumn (1.36 ppm) while the smallest amplitude is observed in summer (0.24 ppm). The diurnal amplitudes of CO are observed to be about 1.01 and 0.62 ppm, respectively during winter and spring. Like CO<sub>2</sub>, the diurnal cycle of CO (Figure 7b) shows the minimum (0.156 ppm) amplitude in July and maximum (1.85 ppm) in October. After October the diurnal amplitude keeps on decreasing till summer. The evening peak contributes significantly to the diurnal amplitude of CO. The largest amplitude in CO cycle is observed in autumn (1.36 ppm) while the smallest amplitude is observed in summer (0.24 ppm). The diurnal amplitudes of CO are observed to be about 1.01 and 0.62 ppm, respectively during winter and spring. The monthly diurnal cycle of CO (Figure 7b) shows the minimum (0.156 ppm) amplitude in July and maximum (1.85 ppm) in October. After October the diurnal amplitude keeps on decreasing till summer.

2. The content given on the emissions and the conclusions on the traffic sector vs the cooking and industrial one: there is a lack of information on the studied region and on the relative role of the different emission sectors that should be given quantitatively, with proper references. Especially, the part of emissions due to residential and slum cooking is almost not discussed, while the available literature explains that this emission sector is responsible for a large amount of atmospheric CO (less in CO<sub>2</sub>).

Response: As per suggestion, we have included following more information about the relative contributions of different emission sectors in Section 2.

“An emission inventory for this city, which is developed for all known sources, shows the annual emissions (for year 2010) of CO<sub>2</sub> and CO about 22.4 million tons and 707,000 tons respectively (<http://www.indiaenvironmentportal.org.in/files/file/Air-Pollution-in-Six-Indian-Cities.pdf>). Out of these emissions, transport sector contribute about 36% in CO<sub>2</sub> emission and 25% in CO emissions, power plants contribute about 32% in CO<sub>2</sub> emissions and 30% in CO emission, industries contribute about 18% in CO<sub>2</sub> emissions and 12% in CO emissions and domestic sources contribute about 6% in CO<sub>2</sub> emissions and 22% in CO emissions.”

The conclusion on the strong influence of traffic given in sector 4.4 is not convincing according to Table1.

Response: Thank you very much for raising these points. This is also pointed by the Referee #1. Now we have modified our conclusions as per further suggestions. As per suggestions, we have removed old figure and added a new Fig. 1 (shown in the end of the comments) in the main text, in which the data are segregated seasonally in different time windows. After carefully analysing the CO: CO<sub>2</sub> ratios, now we have included the domestic and industrial emission sectors along with the transport sector. After this modification, the crux of this modified study is that, during evening hours, emission from transport and domestic sources mostly dominate while rest of periods transport and industrial emission sources mostly dominate mostly over the study region.

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But it is then used as acquired in the following sections (e.g. 4.5 and 4.6), that should also be reviewed in function, taking into account especially the cooking sector, which is another anthropogenic source (nor a natural nor a fossil one), as well as the industrial one (as the authors mentioned there are several industries in the studied area).

Response: As per your and the first referee suggestions, now Section 4.5 includes the validity only from the night-time and rush-hours periods. The estimated slopes for these periods will show the combined influence from all anthropogenic sources and hence will be helpful to validate the anthropogenic emissions of CO. Rest of the results corresponding for different period, we have removed from the section for presenting results more clearly. Now we have modified our conclusion accordingly. We have replaced the fossil fuel emission terms from the anthropogenic emissions. It includes all emissions such as vehicular emission, industrial emission as well as cooking sector emissions.

3/. The validation of the data: the quality of the CO data set is poorly explain, and the data treated with a single calibration standard while no test is given on the instrumental linearity for this species. The data treatment should thus be better reported and these specific points precisely addressed.

Response: Thank you very much for your suggestion. We have calibrated this instrument with one calibration mixture from Linde, UK. As per suggestion, we have checked the instrument linearity. We have diluted the calibration mixture of CO from 970 ppb to 100 ppb. The calibration mixture is diluted in a glass made dead volume using highly precise and accurate pure air (air free from water vapour, particles, carbon monoxide (CO), sulphur dioxide (SO<sub>2</sub>), oxides of nitrogen (NO<sub>x</sub>), ozone (O<sub>3</sub>), and hydrocarbons (HC)) from ECO Physics generator. The flows of calibration mixtures and pure air were regulated using two separate mass flow controllers from Aalborg. For increasing the interaction time both gases (zero air and calibration mixture) to insure a homogeneous mixing, the spring shaped dead volume is used. Each diluted mixture is passed for 30 minutes in the instrument and average of last 10 minute concentration is used for the

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test. The instrument shows excellent linearity for CO and slope is observed 0.98. The calibration graph (Fig.2: shown in the end of the comments) is given for the reference only. Accordingly, we have modified the text in the section.

4/. The question of entertainment of air on top of the PBLH is not addressed, and the PBLH seems to be considered as mixed at any time of the day.

Response: We have added the information regarding the entertainment of air on top of the PBLH in the diurnal variation of both CO<sub>2</sub> and CO sections.

“During winter and spring seasons CO<sub>2</sub> levels are observed constant during night hours and small increase is observed only from 0600 to 0800 hrs during the winter season. While in contrary to this, subsequent section shows a continuous decline in the night time concentrations of main anthropogenic tracer CO, which indicates that there is enough vertical mixing of low CO air from above that once the CO source is turned off, its concentration drops. Hence, constant levels of CO<sub>2</sub> at night hours during these seasons gives the evidence of a continued but weak sources (such as respiration) in order to offset dilution of mixing of low CO<sub>2</sub> air from aloft. Dry soil conditions could be one of the possible caused for weak respirations. Further, distinct timings have been observed in the morning peak of CO<sub>2</sub> during different seasons. It is mostly related to the sunrise time, which will decide the evolutions time of PBL height and beginning of vegetation photosynthesis. The sunrise occurs at 0555-0620 hrs, 0620-0700 hrs, 0700-0723 hrs and 0720-0554 hrs during summer, autumn, winter and spring, respectively. During spring and summer, rush hour starts after sunrise, so the vehicular emissions occur when the PBL has been already high and photosynthetic activity has begun. The CO<sub>2</sub> concentration is observed lowest in the morning during the summer season as compared to other seasons. This is because CO<sub>2</sub> uptake by active vegetation deplete entire mixed layer during day time and when the residual layer mixes to the surface in the morning, low-CO<sub>2</sub> air is mixed down. In winter and autumn, rush hour starts parallel with the sunrise, so the emissions occur when the PBL is low and concentration builds up is much stronger in these seasons than in spring and summer

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seasons”.

The CO/CO<sub>2</sub> ratio diurnal variability should take this into account, a point that carefully needs to be studied at different time windows.

Response: As per the suggestions in further comments, we have modified this section, which discuss about the diurnal variability of CO/CO<sub>2</sub> ratios. Instead of using whole measurements of both gases for studying the emission characteristics, now the modified section discuss the emission characteristics of dominant emission sources at different time windows during all the four seasons. Most of the details about this section are given in the following comments.

After these major revisions, I am convinced that this work will be of very good quality for publication in ACP.

Specific comments:

Abstract: A sentence on your objectives should be given after the first sentence. What is the reliability of the CO<sub>2</sub> emissions inventory? Response: We thank you very much for the suggestions. We have added and rearranged following lines in the abstract section.

“In order to draw effective emission mitigation policies for combating future climate change as well as independently validate the emission inventories based on statistical approaches with large range of uncertainty, there is an urgent need for greenhouse gases measurements over representative urban regions. India is a fast developing country, where fossil fuel emissions have increased dramatically in the last three decades and predicted further to continue to grow by at least 6% per year through 2025. In the absence of systematic CO<sub>2</sub> measurements over the Indian urban locations, CO<sub>2</sub> along with an anthropogenic emission tracer carbon monoxide (CO) are being measured at Ahmedabad, a major urban site in western India, using a state-of-the-art laser based cavity ring down spectroscopy technique from November 2013 to

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May 2015 with a break during March to June 2014”.

Introduction Much too long. Remove detailed information. Response: Referee #1 also pointed out this issue. So now we have very carefully modified, rearranged and synthesized the introductory section.

p. 32187 : Remove lines 8-11 (too long) ——Removed Line 14: a country can be very small or very large so give rather km (100-1000 km<sup>2</sup> for the regional scale generally) ——Modified the text accordingly

Line 21: different. . . = this sentence is very unprecise —— Removed

Line 28: not only traffic but also industry etc. —— Changed accordingly

p. 32188 : Lines 2-8: too long ——

Response: According to comment #1 we have synthesized the introductory section. So as a part of this change, we have removed some information from this para and modified text by adding the following lines.

“Along with the need for atmospheric measurements for predicting the future levels of CO<sub>2</sub>, quantifying the components of anthropogenic emissions of CO<sub>2</sub> is similarly important for providing independent verification of mitigation strategies as well as understanding the biospheric component of CO<sub>2</sub>. Only CO<sub>2</sub> measurements can not be helpful for making such study due to the large role of biospheric fluxes in its atmospheric distributions. The proposed strategy for quantification of the anthropogenic component of CO<sub>2</sub> emissions is to measure simultaneously the anthropogenic tracers. CO can be used as a surrogate tracer for detecting and quantifying anthropogenic emissions from burning processes, since it is a major product of incomplete combustion (Turnbull et al., 2006; Wang et al., 2010). The vehicular as well as industrial emissions contribute large fluxes of CO<sub>2</sub> and CO to the atmosphere in urban regions. Several grounds based and aircraft based simultaneous studies of CO and CO<sub>2</sub> have been done in the past from different parts of the world (Turnbull et al., 2006; Wunch et al., 2009; Wang et al., 2010;

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Newman et al., 2013) but such study is lacking in India except recently reported results from weekly samples for three Indian sites by Lin et al. (2015)".

Line 9-29: too detailed info. Remove most of these lines, and focus more on urban studies. \_\_\_\_\_

Response: As per general comments, we have synthesized the introductory section. We have removed some detailed information and modified text by adding following lines.

"Measurements at different regions (e.g., rural, remote, urban) and at different frequency (e.g., weekly, daily, hourly etc) have their own advantage and limitations. For example, measurements at remote locations at weekly interval can be useful for studying seasonal cycle, growth rate, and estimating the regional carbon sources and sinks after combining their concentrations with inverse modelling and atmospheric tracer transport models. However, some important studies like their diurnal variations, temporal covariance. etc are not possible from these measurements due to their limitations. Analysis of temporal covariance of atmospheric mixing processes and variation of flux on shorter timescales, e.g., sub-daily, is essential for understanding local to urban scale CO<sub>2</sub> flux variations (Ahmadov et al., 2007; Pérez-Landa et al., 2007; Briber et al., 2013; Lopez et al., 2013; Ammoura et al., 2014; Ballav et al., 2015). Urban regions contribute about 70% of global CO<sub>2</sub> emission from anthropogenic sources and further projected to increase further over the coming decades. Therefore, measurements over these regions are very helpful for understanding emissions growth as well as verifying the mitigation policies. "

p.32190 :Objectives not clear, reformulate please. Response: We have added following lines before the paragraph for highlighting the importance of the study.

"It is pertinent to mention here that till now there are no reports of CO<sub>2</sub> measurements over the urban locations of the Indian subcontinent, which could be an important player in the global carbon budget as well as mitigation purpose due to strong growing an-

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thropogenic activities specifically fast growing traffic sector and sinks (large areas of forests and croplands). Hence, the present study is an attempt to reduce this gap by understanding the CO<sub>2</sub> mixing ratios in light of its sources and sinks at an urban region in India”.

Section 2:

Lines 15-27 What is the height of the sampling height above ground level?

Response: The sampling height is about 25 meter above the ground level. This information is already given in the line #6 of p. 31293.

The information given on the emission sectors should be improved. It is a key point of your argumentation next. Please quantify here and give numbers on the relative role of the different CO<sub>2</sub> and CO emission sectors in Ahmedabad (there are several sources to compare, here is one: <http://www.indiaenvironmentportal.org.in/files/file/Air-Pollution-in-Six-IndianCities.pdf>).

Response: We thank you very much for bringing out these points. We have added following information in Section 2.

“An emission inventory for this city, which is developed for all known sources, shows the annual emissions (for year 2010) of CO<sub>2</sub> and CO about 22.4 million tons and 707,000 tons respectively (<http://www.indiaenvironmentportal.org.in/files/file/Air-Pollution-in-Six-Indian-Cities.pdf>). Out of these emissions, transport sector contributes about 36% in CO<sub>2</sub> emission and 25% in CO emissions, power plants contribute about 32% in CO<sub>2</sub> emissions and 30% in CO emission, industries contribute about 18% in CO<sub>2</sub> emissions and 12% in CO emissions and domestic sources contribute about 6% in CO<sub>2</sub> emissions and 22% in CO emissions”.

p.32191 :Line 17 and line 21: check months consistency. ....Checked

Section 3 This section generally lacks of precision on the procedures.

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p.32193 Lines 1-2: do you mean the CRDS instruments in general, or yours? Your instruments should be discussed here, each CRDS instrumental is specific and needs to be validated (although this is right that they are usually within WMO recommendations).

Response: In this section we have discussed mostly about our instrument. We have modified respective sentences accordingly.

p.32194: Lines 1-12: this part is critical. The CO Dataset is calibrated with one single tank from Linde UK. Is it linked to the WMO scale? Despite this single cal tank, no linearity tests are reported for CO. How can you make sure your CO data set is not biased by an instrumental drift? Also, you need to report the accuracy of your measurements (both CO<sub>2</sub> and CO).

Response: The calibration tank of CO does not follow the WMO scale. Its traceability is based on NIST by weight. The accuracy of both gases is included in the respective section. As per suggestion and discussed previously, we have checked instrument linearity for CO and found that the instrument is highly linear for CO. The graph is already given in the previous discussion for the reference only. The modified text is given below.

“The measurement system is equipped with three high pressure aluminium cylinders containing gas mixtures of CO<sub>2</sub> ( $350.67 \pm 0.02$ ,  $399.68 \pm 0.002$  and  $426.20 \pm 0.006$  ppm) in dry air from NOAA, Bolder USA, and one cylinder of CO (970 ppb) from Linde UK. These tanks were used to calibrate the instrument for CO<sub>2</sub> and CO. An additional gas standard tank (CO<sub>2</sub>: 338 ppm, CO: 700 ppb), known as the “target”, is used to monitor the instrumental drift and to assess the dataset accuracy and repeatability. The target tank values are calibrated against the CO<sub>2</sub> and CO calibration mixtures. The target tank and calibration gases were measured mostly in the mid of every month (Each calibration gas is passed for 30 minute and target tank for 60 minutes). The target gas is introduced into the instrument for a period of 24 hours also once in a six month, for checking the diurnal variability of instrument drift. Maximum drift for 24 hours has been

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calculated by subtracting the maximum and minimum value of 5 minute average, which were found to be 0.2 ppm and 0.015 ppm respectively for CO<sub>2</sub> and CO. For all calibration mixtures, the measured concentration is calculated as the average of the last 10 minutes. The linearity of the instrument for CO<sub>2</sub> measurements has been checked by applying the linear fit equation of the CO<sub>2</sub> concentration of the calibration standards (350.67 ppm, 399.68 ppm and 426.20 ppm) measured by the analyzer. The slope is found in the range of 0.99 - 1.007 ppm with a correlation coefficient (r) of about 0.999. Further, linearity of the instrument for CO is also checked by diluting the calibration mixture from 970 ppb to 100 ppb. The calibration mixture is diluted in a glass made dead volume using highly precise and accurate pure air (air free from water vapour, particles, carbon monoxide (CO), sulphur dioxide (SO<sub>2</sub>), oxides of nitrogen (NO<sub>x</sub>), ozone (O<sub>3</sub>), and hydrocarbons (HC)) from ECO Physics generator and two mass flow controller. For increasing the interaction time of both the gases (zero air and calibration mixture) and to insure a homogeneous mixing, the spring shaped dead volume is used. Each diluted mixture is passed for 30 minutes in the instrument and average of last 10 minute data are used for the test. The instrument shows excellent linearity for CO and slope is observed to be 0.98. The accuracy of the measurements is calculated by subtracting the mean difference of measured CO<sub>2</sub> and CO concentration from the actual concentration of both gases in target gas. The accuracies of CO<sub>2</sub> and CO are found in the range of 0.05-0.2 ppm and 0.01-0.025 ppm respectively. The repeatability of both gases are calculated by the standard deviation of the mean concentration of target gas measured by the analyser over the period of observations and found 0.3 ppm and 0.04 ppm for CO<sub>2</sub> and CO respectively.”

Section 4 p.32195 :This part are interesting but too long.

Response: We have shortened this part after removing some sentences and rearranging them. The modified text is given following.

“Figures 3a and 3c show the time series of 30 minute average CO<sub>2</sub> and CO concentrations for the period of November, 2013 - February, 2014 and July, 2014 to May, 2015.

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Large and periodic variations indicate the stronger diurnal dependence of both gases. Overall, the concentrations and variability of both gases are observed lowest in the month of July and August, while maximum scatter in the concentrations and several plumes of very high levels both gases have been observed from October, 2014 until mid-March 2015. Almost all plumes of CO<sub>2</sub> and CO are one to one correlated and are found mostly during evening rush hours and late nights. Figures 3e and 3f show the variations of CO<sub>2</sub> and CO concentrations with wind speed and direction for the study period except July, August and September due to non-availability of wind data. Most of the high and low concentrations of both gases are found to be associated with low and high wind speeds. There is no specific direction for high levels of these gases. This probably indicates the transport sector is an important contributor to the local emissions since the measurement site is surrounded by city roads”.

Lines 23-26: please reformulate

Response: We have reformulated the sentences, which are given below. “Figures 3b and 3d show the probability distributions or frequency distributions of CO<sub>2</sub> and CO concentration during the study period. Both gases show different distributions from each other. This difference could be attributed due to the additional role of biospheric cycle (photosynthesis and respiration) on the levels of CO<sub>2</sub>, apart from the common controlling factors (local sources, regional transport, PBL dynamics etc) responsible for distributions of both gases. The control of the boundary layer is common for the diurnal variations of these species because of their chemical lifetimes are longer (> months) than the timescale of PBL height variations (ˆLij hrs)”.

p.32196 :Lines 22-23: what is the demonstration for this argument?

Response: We have modified the sentence. The modified part is given below. “The seasonal cycle from first approach will present the overall variability in both gases. On the other hand, second approach removes the auto-covariance by excluding CO<sub>2</sub> and CO data mainly affected by local emission sources and represent seasonal cycles at

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the well mixed volume of the atmosphere”.

p.32197 Remove lines 1-2 ———— Removed.

Lines 9-11: not clear — As mentioned in general comments, this section is synthesise and hence these lines are removed.

Lines 11-18: synthesise — As mentioned in previous comments also, after synthesising the respective section, some of mentioned lines are rearranged, while some are removed.

Lines 17-20: how much of the data coverage does this step represent? ——— Mean of every month  $\sim 24 \text{ hrs} \times 30 = 720 \text{ hrs}$  data.

p.32200 : Lines 1-4: reformulate ———— As per previous suggestion for synthesizing this section, these sentences are already modified. The modified text is given below.

“CO<sub>2</sub> concentrations start decreasing from 0000 to 0300 hrs and increases slightly afterwards till 0600-0700 hrs during summer and autumn. Respiration of CO<sub>2</sub> from the vegetation is mostly responsible for this night time increase. During winter and spring seasons CO<sub>2</sub> levels are observed constant during night hours and small increase is observed only from 0600 to 0800 hrs during the winter season. In contrary to this, subsequent section shows a continuous decline in the night time concentrations of the main anthropogenic tracer CO, which indicates that there is enough vertical mixing of low CO air from above that once the CO source is turned off, its concentration drops. Hence, constant level of CO<sub>2</sub> at night hours during these seasons give the evidence of a continued but weak sources (such as respiration) in order to offset dilution of mixing of low CO<sub>2</sub> air from aloft. Dry soil conditions could be one of the possible cause for weak respirations.”

p.32202 : Lines: 15-20: reformulate

Response: We have reformulated the text by the following lines.

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“The first difference is observed in the timing of the occurrence of morning peaks: CO<sub>2</sub> peaks occur slightly before the CO peak due to the triggering photosynthesis process by the sunrise. On the other hand, the morning peaks of CO mostly depend on the rush hour traffic and are consistent at 0800-1000 hrs in all seasons. The second difference is that the afternoon concentrations of CO show little seasonal spread as compared to the afternoon concentrations of CO<sub>2</sub>. Again, this is due to the biospheric control on the levels of CO<sub>2</sub> during the afternoon hours of different seasons while CO levels are mainly controlled by the dilution during these hours”.

p.32203: Define the baseline and background terms.

Response: Both words are used for same purpose: “the least affected levels of CO<sub>2</sub> from local sources”. But since there may be some misunderstanding from different terminology, we have removed baseline word from the text. We have included the following lines for defining the background.

“The measurements are generally affected by the dilution due to the boundary layer dynamics, but considering their ratios will cancel this effect. Further, the interpretation of correlation ratios in terms of their dominant emission sources needs to isolate first the local urban signal. For this, the measurements have to be corrected from their background influence. The background concentrations are generally those levels which have almost negligible influence from the local emission sources”.

How sensitive is your 5th percentile method? This was for example assessed against MACC fields in Ammoura et al ACPD 2015 (a new method. . .). Give clues.

Response: We have added following lines for clarity about using 5th percentile method for calculating the background. “It is observed that the mixing ratios of both gases at low wind speed, which show the influence of local urban signal, are significantly higher than background levels and hence confirm that the definition of background will not significantly affect the derived ratios (Ammoura et al., 2015). This technique of measuring the background is extensively studied by Ammoura et al (2015) and found

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suitable for both the gases CO and CO<sub>2</sub>, even having the role of summer uptake on the levels of CO<sub>2</sub>".

p.32204: The role of cooking (poor combustion => large co/co<sub>2</sub>), other FF sources etc should be considered here.

Response: We have modified whole section according to previous comments and Referee #1 comments.

p.32205: Be careful here at hours when the PBLH evolves (see general) Lines 17-20: this is critical. I do not agree with your argumentation. Table 1 does not show that the observed ratios (30-50 ppb/ppm) are much lower than the domestic sources (52.99 ppb/ppm). You cannot conclude that this is driven by gasoline emissions. And several solutions exist. You could have a mix of emissions from traffic and domestic sources for example. At what time do people have dinner in Ahmedabad? Same time than rush hours or not? Etc. This section needs to be thought more and the different options argued to drive to a solid conclusion.

Response: The average dinner time is 1900-2100 hrs. So these ratios could be influenced by the mix emission sources such as vehicular emissions, domestic sources. Hence rather than fossil fuel emission now we have included anthropogenic emission. We have added following explanations in the modified text.

"Except monsoon, the  $\Delta\text{CO}$  (exc) /  $\Delta\text{CO}_2$  (exc) ratios and their correlations are fairly comparable in other seasons in the evening rush hours, which indicate stronger influence of common emission sources. Ratios during this time can be considered as fresh emissions since dilution and chemical loss of CO can be considered negligible for this time. Most of these data fall in the domestic and transport sector emission ratio lines, which indicate that during this time intervals these sources mostly dominate (Table 1 in the main text). On the other hand, during other time intervals most of the data are scattered between industrial and transport sectors emission ratio lines. Hence, from this we can conclude that during evening hours, transport and domestic sources mostly

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dominate while during other periods transport and industrial emission sources mostly dominate”.

p.32206 : Remind the question of the entrainment pb in the morning for example (check my general comments).

Response: We have already discussed the modified sentence previously according the general comments.

p.32207 :It would be interesting to try to give an explanation about this. What emissions is EDGAR missing then? Is it a sector or is it underestimated on all sectors? What about emissions from slum /residential cooking for example? You might found this paper interesting on the CO emissions from New Delhi: [http://aaqr.org/VOL15\\_No3\\_June2015/36\\_AAQR-14-07-TN0132\\_1137-1144.pdf](http://aaqr.org/VOL15_No3_June2015/36_AAQR-14-07-TN0132_1137-1144.pdf)

Response: Thank you very much for the reference and wonderful suggestion. Accordingly, we have added following explanation in the text.

“The EDGAR inventory estimate the relative contributions of CO from industrial, transport and slum/residential sectors to be about 42%, 42% and 10%, respectively. The possible cause for underestimation of CO by the EDGAR inventory could be the underestimation of residential emission, since other inventory particularly for major urban Indian cities (<http://www.indiaenvironmentportal.org.in/files/file/Air-Pollution-in-Six-Indian-Cities.pdf>) show large relative contributions from residential sector. The uncertainty associated with the emission factors for different sectors could be another cause for the underestimation of CO emissions, since these are important parameters for developing the inventory (Sahu et al., 2015)”.

Lines 14-15: following my remarks above, I do not agree with your argument on the large role you attribute to CO emissions from fossil fuels incomplete combustion only. Other sectors are still on the race as long as you did not demonstrate the contrary.

Response: We have modified the sentence. We replace the fossil fuel term from the

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anthropogenic emission. Accordingly, text is modified.

“CO has virtually no natural source in an urban environments except oxidation of hydrocarbons and hence can help to disentangle the relative contributions of anthropogenic (from transport, power plant, industrial etc) and the biospheric sources (mainly from respiration) of CO<sub>2</sub>, by serving as a tracer of combustion activity on shorter timescale (Duren and Miller, 2012).”

Lines 27-28: this was not clearly demonstrated as well.

Response: We have modified/removed the text by following lines.

“Figure 9a shows the excess diurnal variations of CO<sub>2</sub> above the background levels during different seasons. The observed concentrations of both gases can also be directly used for calculating the emission ratio, provided the measured levels are not highly affected from natural sources and share the same origin. We have used the evening time (1900-2100 hrs) data of CO<sub>2</sub>exc and COexc for whole study period to calculate the emission ratio of CO/CO<sub>2</sub> from predominant anthropogenic emission sources, since the correlation ( $r = 0.95$ ) for this period is very high and can hence, be considered that the levels of both gases for this period are mostly affected by same types of anthropogenic sources. Also, there can be considered negligible contribution of biospheric sources in CO<sub>2</sub>”.

p.32208 : Lines 4-6: very surprising, aren't people cooking at this time? 47 ppb/ppm is more than gasoline and in between gasoline and biofuels/coal.

Response: We have modified the sentence. As discussed previously also, this ratio is mostly dominating by vehicular and domestic fuel emissions. So we have included anthropogenic emission source in place of gasoline and biofuels.

Line 13: same, no solid argumentation given for this

Response: We have removed the sentence from the text.

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Lines 26-27: I do not agree here as well. I do not think this is true to say that the other sources do not emit CO<sub>2</sub>. What about wood burning, cooking etc. again. These are not natural but anthropogenic.

Response: This sentence is already modified as per previous suggestions.

p. 32209 : Lines 1-13: this part should be fully rewritten according to the comments above.

Response: We have modified this section by adding following lines in the text.

“The average diurnal cycles of CO<sub>2</sub> above its background for each season are shown in (Figure 9a). In Section 4.3.1, we have discussed qualitatively the role of different sources in the diurnal cycle of CO<sub>2</sub>. With the help of the above method, now the contributions of anthropogenic (CO<sub>2</sub> (ant)) and biospheric sources (CO<sub>2</sub> (bio)) are discussed quantitatively. Due to unviability of PBL measurements, we cannot disentangle the contributions of boundary layer dynamics. The diurnal pattern of CO<sub>2</sub> (ant) (Figure 9c) reflects the pattern like CO, because we are using constant RCO/CO<sub>2</sub> (ant) for all seasons. Overall, this analysis suggests that the anthropogenic emissions of CO<sub>2</sub>, mostly from transport and industrial sectors during early morning during 0600-1000 hrs varied from 15 to 60% (4-15 ppm). During afternoon hours (1100-1700 hrs), the anthropogenic originated (transport and industrial sources, mainly) CO<sub>2</sub> varied between 20 and 70% (1-11 ppm). During evening rush hours (1800-2200 hrs), highest contributions of combined emissions of anthropogenic sources (mainly transport and domestic) are observed. During this period the contributions vary from 50 to 95% (2-44 ppm). During night/early morning hours (0000-0700 hrs) non-anthropogenic sources (mostly biospheric respiration) contribute from 8 to 41 ppm of CO<sub>2</sub> (Figure 9d). The highest contributions from 18 to 41 ppm are observed in the autumn from the respiration sources during night hours, since there is more biomass during this season after the South Asian summer monsoon. During the afternoon hours, lower biospheric component of CO<sub>2</sub> could be due to a combination of the effects of afternoon anthropogenic

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emissions, biospheric uptake of CO<sub>2</sub> and higher PBL height”.

Line 19: rewrite with 1 time  $\hat{A}$  component  $\hat{Z}$ , not 3 times; remove  $\hat{A}$  diurnal amplitude $\hat{Z}$ ; add  $\hat{A}$  s  $\hat{Z}$  to observation. Response: Done as per suggestion.

Lines 24-26: give reference Response: We have included the following reference. Law, R. M., Peters, W., Rödenbeck, C., Aulagnier, C., Baker, I., Bergmann, D. J., Bousquet, P., Brandt, J., Bruhwiler, L., Cameron-Smith, P. J., Christensen, J. H., Delage, F., Denning, A. S., Fan, S., Geels, C., Houweling, S., Imasu, R., Karstens, U., Kawa, S. R., Kleist, J., Krol, M. C., Lin, S.-J., Lokupitiya, R., Maki, T., Maksyutov, S., Niwa, Y., Onishi, R., Parazoo, N., Patra, P. K., Pieterse, G., Rivier, L., Satoh, M., Serrar, S., Taguchi, S., Takigawa, M., Vautard, R., Vermeulen, A. T., and Zhu, Z.: TransCom model simulations of hourly atmospheric CO<sub>2</sub>: Experimental overview and diurnal cycle results for 2002, *Global Biogeochemical Cycles*, 22, doi:10.1029/2007GB003050, gB3009, 2008.

p.32211 Line 23: this part is vague about the tracers you used. Please give more clues to the reader.

Response: We have modified the explanations by adding informations in detail and following reference.

“Separate correlations of each CO<sub>2</sub> tracer with the observations are helpful to determine relative importance of each flux component in the CO<sub>2</sub> variation (Patra et al., 2008). Hence, we separately studied the correlation between biospheric component of CO<sub>2</sub> and anthropogenic component of CO<sub>2</sub>, estimated by model using CASA-3hr fluxes and EDGAR v4.2 inventory, and the measurements. The correlation coefficients give the hint about major controlling factors on the levels of CO<sub>2</sub>. Figure 11b shows the resulting correlations for separate flux component with respect to measurements”.

Patra, P. K., Law, R. M., Peters, W., Rödenbeck, C., Takigawa, M., Aulagnier, C., Baker, I., Bergmann, D. J., Bousquet, P., Brandt, J., Bruhwiler, L., Cameron-Smith, P. J., Christensen, J. H., Delage, F., Denning, A. S., Fan, S., Geels, C., Houweling, S., Imasu, R.,

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Karstens, U., Kawa, S. R., Kleist, J., Krol, M. C., Lin, S.-J., Lokupitiya, R., Maki, T., Maksyutov, S., Niwa, Y., Onishi, R., Parazoo, N., Pieterse, G., Rivier, L., Satoh, M., Serrar, S., Taguchi, S., Vautard, R., Vermeulen, A. T., and Zhu, Z.: TransCom model simulations of hourly atmospheric CO<sub>2</sub>: Analysis of synoptic-scale variations for the period 2002–2003, Global Biogeochemical Cycles, 22, doi:10.1029/2007GB003081, gB4013, 2008.

p.32212:

5 Conclusions Lines 2-4 Transition with CO should be improved. ....Improved

Lines 7-10: yes, this is an excellent remark! .....Modified

Lines 20: here again you mix seasons and climate features. Please modify. ———  
Done

p.32214: Line 2: remove (fossil fuel) unless you manage to demonstrate it correctly.....

Response: We have modified the text after including the following lines according to the change made in the respective sections previously.

“The availability of simultaneous and continuous measurements of CO<sub>2</sub> and CO have made it possible to study their correlations at different time windows (during morning (0600-1000 hrs), noon (1100-1700 hrs), evening (1800-2200) and night (0000-0500 hrs) hours) of distinct seasons. Using the correlation slopes and comparing them with the emission ratios of different sources, contributions of distinct sources is discussed qualitatively. It is observed that during the evening hours, measurements over the study region are mostly affected by transport and domestic sources, while during other periods the levels of both gases are mostly dominated by the emissions from transport and industrial sources”.

Lines 7-9: These lines should be changed (see remarks p.32208) .....Changed

Technical corrections: Define ppm as part per million .

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Response: Defined in the beginning of text Do not mix season and climate regime (ex. winter and monsoon => winter and summer)

Response: Replaced monsoon by the summer monsoon season.

CO should be expressed in ppb (to be defined) and not in ppm, for consistency with the tables. Do not mix noon (12h) and afternoon. .... Response: Changes

Add " the " before model or before CASA model. You can also replace model by modeling framework to avoid repetitions.

Response: .Done

Title: " In CO<sub>2</sub> and CO " should be replaced by " of atmospheric ". ....

Response: Replaced

The short title " CO<sub>2</sub> over urban region " requires as well the keywords CO and India. Abstract Line 25: replace variations by ones.....

Response: Replaced

Introduction P.32157 Line 18: remove (such as the Kyoto Protocol) .

Response: Removed

Section 2 P.32191 line 9/10: already said p.32190 line 17. ....

Response: This sentence is removed.

Section 3 Lines 12-26: check English style please (First... the second...).

Response: We have checked the style and suitably modified the text wherever needed. The modified text is the following.

"The ambient measurements of CO<sub>2</sub> and CO concentrations have been made using the wavelength scanned cavity ring down spectroscopic (CRDS) technique based analyser (Picarro-G2401) at 0.5 Hz. CRDS offers highly sensitive and précised measure-

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ments of trace gases in the ambient air, due to its three main characteristics (Bitter et al., 2005; Chen et al., 2010; Karion et al., 2013). (1) Longer interaction path length (around 20 km). (2) Low operating pressure (~140 Torr) of cell to avoid pressure broadening which ensures that the peak height or area is linearly proportional to the concentration. (3) Decay time of light intensity is measured instead of the absorption of light intensity, making it independent of the fluctuation in the incident light intensity. “

P.32194 Line 23/24: remove for the model simulations .....Done Section 4 P.32196 Line 19: In the first approach .....Done (Fig.4a). . . 5 Conclusions .....Done p.32212 Line 12: remove “The unique flow of  $\dot{V}$ . . . . .” Removed Line 20: is  $\dot{V}$  transported  $\dot{V}$  the right term? Response: Thank you very much for pointing this. The appropriate term is emitted. We have removed the sentence “representing CO<sub>2</sub> and CO transported from anthropogenic sources” for more clear explanation.

Lines 23-24: remove “seasonal. . . season)  $\dot{V}$  and replace by activity (do not explain such process in your conclusions) .....We have modified the sentence accordingly p.32213 Line 12: replace effects of by undergo .....Replaced p.32215 Line 3: validity is a bit strong... You could say we assessed independently, which is more neutral. ....Done

Tables Table 4: the legend does not seem appropriate to the table.

Response: The legend has been changed by the following text. “Seasonal mean concentrations and diurnal amplitudes (max-min) of CO<sub>2</sub> and CO over Ahmedabad.”

Figures Figure 1: “from the EDGAR. . .  $\dot{V}$  . . . . .” Corrected

Figure 2: does the pump belong to the CRDS analyzer? Then make a box of both items together. Replace analysis system by “experimental set-up  $\dot{V}$ . Replace “in  $\dot{V}$  by “upstream of  $\dot{V}$ . Remove the form “from the ambient air  $\dot{V}$ . Replace “the calibration mixtures (three)  $\dot{V}$  by “Three calibration mixtures  $\dot{V}$ . . . . .

Response: We have made all the mentioned corrections. Further, a box around the

analyser and pump and two boxes around the two stage water removing systems (designed at PRL) have been made.

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 32185, 2015.

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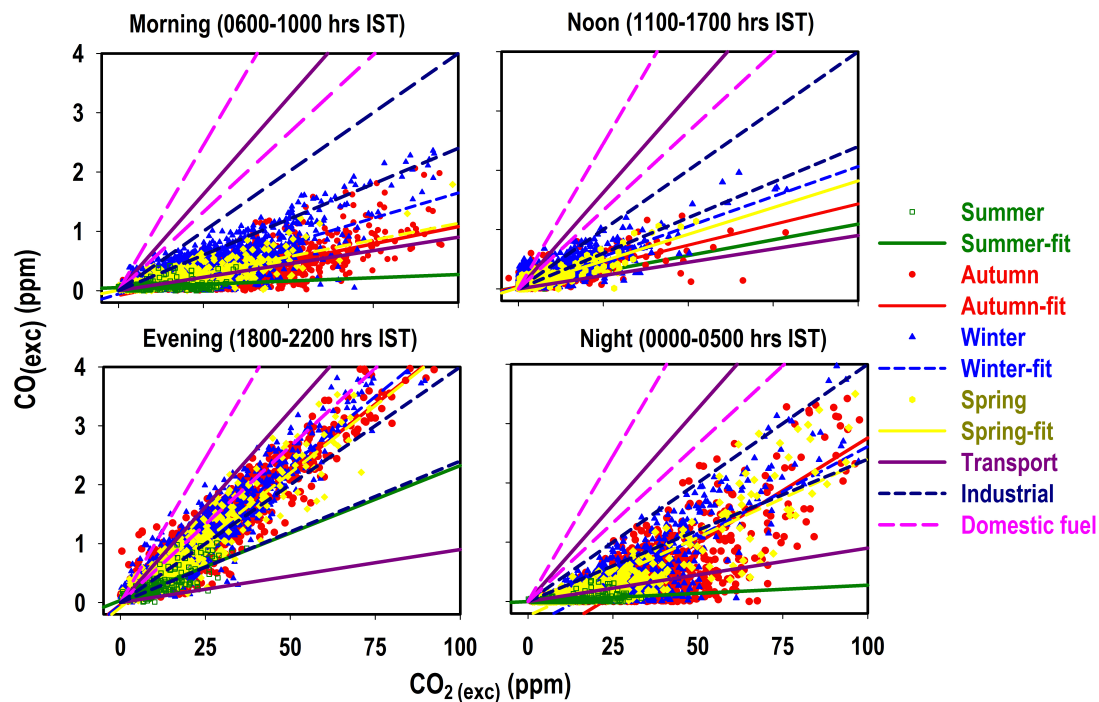
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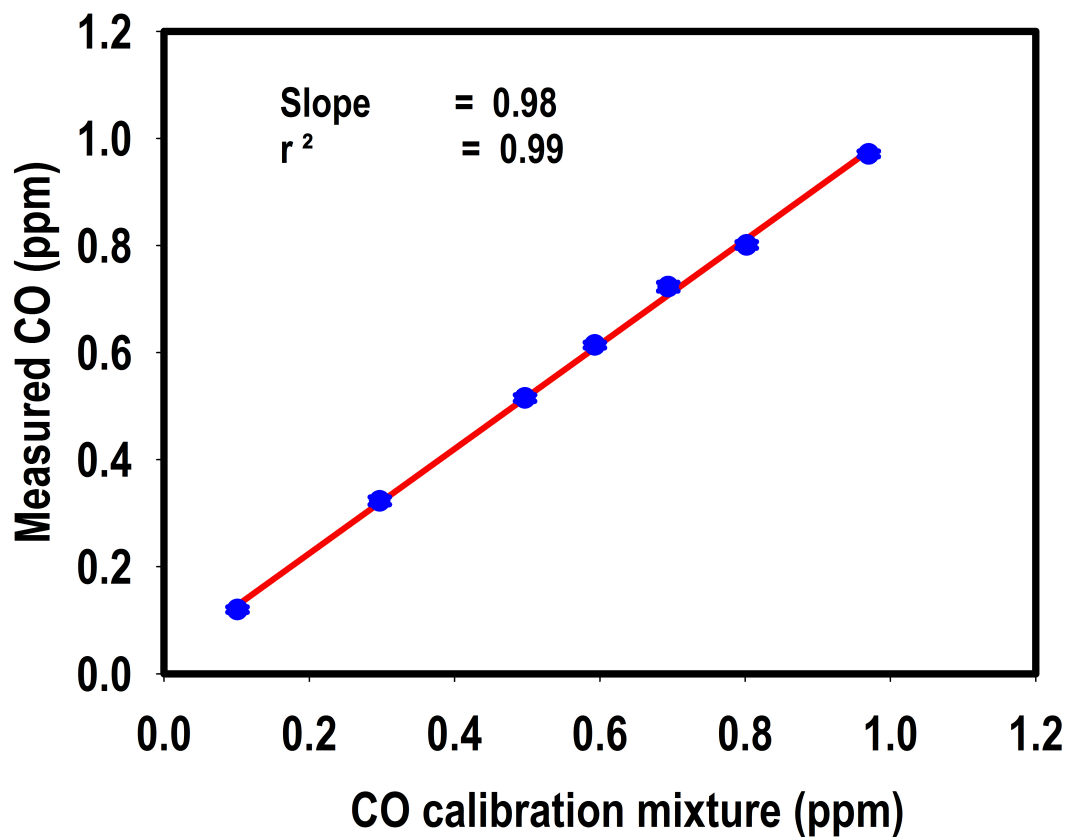






**Fig. 1.** Scatter plots and regression fits of excess CO ( $\text{CO}(\text{exc})$ ) vs. excess  $\text{CO}_2$  ( $\text{CO}_2(\text{exc})$ ) during morning (0600-1000 hrs), noon (1100-1700 hrs), evening (1800-2200) and night (0000-0500 hrs) hours.

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**Fig. 2.** The calibration graph for CO. The x-axis and y-axis show the calibration values and measured values by the analyzer, respectively.

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