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## *Interactive comment on* "Diurnal tracking of anthropogenic CO<sub>2</sub> emissions in the Los Angeles basin megacity during spring, 2010" *by* S. Newman et al.

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Major comments:

1. The main result of the paper is that around 100% of the locally emitted CO2 in LA is of fossil fuel origin during the day, compared to around 50% at night. This result seems to agree with expectations and with previous finding. Therefore, I was left wondering what we have really learnt from this work? Is the main innovation in this paper that the technique is more "affordable and simple" than previous methods (P5773 L7), or are there wider implications? More detail should be given to convince the reader about the novelty of these results. Furthermore, it should be explained how we might expect

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these findings to be used in future work. The suggestion that the authors make in this regard is that satellites should be able to detect the midday fossil CO2 signal in a mega-city. Perhaps this could be elaborated upon. For example, we would hope that e.g. OCO-2 should be able to simply 'detect' the urban CO2 plume from a large city like LA, but is the size of the signal found by the authors large enough, relative to the expected measurement uncertainty, to be able to constrain emissions?

The motivation of the paper has been emphasized more clearly. The value of the analysis presented includes the "affordable and simple" technique applied to the megacity CO2 data to provide full 24-hour estimates of CO2 emissions. Kort et al. (2012) have shown clearly that the anthropogenic signal is visible from space, in this case using the Japanese Greenhouse Gas Observing Satellite (GOSAT). With time, accumulated data will demonstrate trends in this greenhouse gas over cities globally, and modeling will be able to provide information on the emissions giving rise to these trends. However, well-studied regions using the techniques demonstrated here will be needed validate the results from space-borne measurements and to extend the analysis to all hours of the day. Use of tracer ratios, such as COxs/CO2xs avoids some of the problems with assumptions that have to be made with modeling techniques.

2. The background mixing ratio measurements seem to me to be a key quantity in this work. However, the background sites are not mentioned in the main body of the text, but probably should be.

We have added brief descriptions of the sources of the background measurements to the main text.

3. In Appendix B1 it is stated that CO2 background levels were assumed constant throughout the time period. However, Figure A2 shows a clear diurnal cycle at the background site (Palos Verdes). Presumably this indicates a significant local influence at the background site? Therefore, two things occur to me: 1) can it really be assumed that it is a true background site? 2) By assuming a constant background when it actually

appears that the LA background does have a diurnal cycle, is it not likely that the results presented apply to an area much larger than the urban LA region? How are the results different if the diurnal cycle of the difference between these two sites is plotted (perhaps filtering for instances where the wind passed along a trajectory from one to the other)?

Yes, it is likely that there is some diurnal variation in CO2bg but it is likely often small compared to the local CO2ff and CO2bio signals (except perhaps in the case of Santa Anna winds, which did not occur during this campaign). We have calculated the time-varying background using STILT particle back trajectories intersecting the Pacific marine boundary layer background. The conclusion of this effort is that the constant background we had chosen, 393.1 ppm, agrees well with the average of the time-varying calculation,  $393.0 \pm 1$  ppm. The range of the time-varying values is 391 - 395 ppm, and diurnal variation is much less than 1 ppm. The variation we see at the Palos Verdes site does, indeed, show a diurnal pattern, but the data in Figure A2 have not been filtered to account for wind direction or local events. We are accounting for boundary layer variations by taking the average of the daily minimum values.

4. CO background levels were taken from GLOBALVIEW-CO. Can we be sure that these globally-averaged mixing ratios really apply to the area directly upwind of LA?

The CO background levels were not directly from GLOBALVIEW-CO. We have added a more complete description: For CO, suitable measurements of the background are not available, so we employed a method that has been used elsewhere for regional inverse modeling of CO2 (e.g., Gourdji et al., 2012; Schuh et al., in press) and N2O (Jeong et al., 2012b). Briefly, discrete (i.e., flask) measurements of CO from sites in the NOAA Cooperative Air Sampling network (Novelli and Masarie, 2012) are used to estimate time- and latitudinally varying CO values for the Pacific marine boundary layer. The methodology is identical to what is used to produce the GLOBALVIEW-CO (2012) product, but only Pacific sites operated by NOAA were included in the present study. Time- and latitude- dependent vertical gradients were estimated using CO data from Pacific and Gulf coast sites in the NOAA Earth System Laboratory's aircraft network

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(pending) The marine boundary layer and vertical profile information was combined into a time-, latitude- and altitude- dependent "curtain". The curtain is sampled at the western boundary of the modeling domain (130°W) for each of the 500 STILT trajectories corresponding to a particular observation and the values are averaged, producing results that agree quite well with NOAA P3 CO observations during the CalNex-LA campaign (Fig. A2b). The CO background mixing ratios varied from ~135 ppb in the beginning of the time period to ~112 ppb on 6 June, and ~110 ppb at the end of the campaign.

5. The "fossil CO2" diurnal cycle has a single peak around noon. Does this seem reasonable for LA? In many cities, emissions are expected to peak during the morning and evenings.

The single peak in CO2 emissions around noon is reasonable for this time of year and the low wind speeds we observe. Transport of the emissions from the heart of the basin during the morning takes some time to arrive at the receptor site in Pasadena. Indeed, downtown LA is 14 km away and therefore the peak should arrive about 4 hours after emissions there, given an average wind speed of 1 m/s, and the evening peak occurs  $\sim$ 3-4 hours after evening rush hour, at about 20:00. The lack of morning and evening local rush hour peaks probably reflects the fact that the boundary layer height is large during these times during summer. We do see morning local rush hour peaks during the winter, since rush hour begins before the boundary layer begins to expand, but it is frequently cut short by the expansion.

Minor comments:

1. If 50% of the nocturnal CO2 is of biogenic origin, but 100% of the midday is fossil origin, does this mean that all of the biogenic CO2 has been flushed out of the urban boundary layer between the morning and night? Do calculations of the ventilation rate substantiate this?

STILT calculations will give surface influence and background signals but not (trivially)

the average exchange rate for air in the basin. For that, we would use an Eulerian model. Therefore we do not have calculations of the ventilation rate of the basin. During mid-day the boundary layer expands and this is when the fossil fuel emissions are highest and when photosynthetic uptake of CO2 is most active. These effects combine to make the biogenic source component insignificant at this time.

2. The campaign covers a relatively short period (1 month) in 2010. How might the results vary with time of year or inter-annually?

The question of extending these results to annual or interannual scales is a goal of our on-going research, but it is beyond the scope of this paper, since it will require significantly more data and modeling. Preliminary results show that local morning rush hour is frequently observable during winter mornings in the in situ boundary layer measurements, but evening rush hour is rarely obvious. However, both morning and evening rush hours can be resolved during winter months when CO2 data are convolved with HYSPLIT-calculated boundary layer heights, although the morning peak much clearer.

3. In section B3 it is stated that 100 particles were traced in the particle dispersion model. Is this enough to remove noise from the calculate footprints? Previous studies have used tens of thousands of particles for each measurement.

For the STILT model, it is typical to run  $\sim$ 100 particles. The error associated with this number of particles is  $\sim$ 13% (Gerbig et al., 2003; JGR), more than small enough for footprint calculations such as that illustrated in Figure 1b. For the prediction calculations described in section B4 and plotted in Figures B2 and B3, 500 particles were used, following Jeong et al. (2012; GRL).

4. P5773, L16: "... variability OF emission sources..."?

P5773, L18: "... The large magnitude of emissions IS easily detected..."?

P5774, L6: "The latter..." instead of "This last..."?

P5774, L24: "... on the roof OF a trailer"

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The first two typographical errors actually are not errors and changing them as suggested will change the meaning. Therefore, they have been left as they were. The other two have been corrected – thank you for pointing these out!

5. P5778, L2: "... technique is not as successful as using radiocarbon...". This is rather subjective. Please explain in more how and why it is more successful.

We have added a comment comparing the two techniques, emphasizing that radiocarbon is an absolute method that requires no assumptions, whereas the CO/CO2 method requires assumption of the emission ratio, R, of CO/CO2, if this value is not known from measurement or experience.

6. P5781, L16: Daily calibration: can we be sure that there was no diurnal instrument drift (e.g. due to temperature fluctuations)?

The daily calibrations were not compromised by daily temperature variabilities that experienced frequently during field campaigns where instruments are housed in trailers, because the instruments involved, both the Aero-Laser AL5001 CO monitor and the Picarro Isotopic CO2 Analyzer, are very well temperature-controlled at 40 and 45°C, respectively, well above ambient.

7. P5785, L2: Extrapolation of La Jolla data. Was this extrapolation in space or time? If the latter, then when were the La Jolla data collected?

The La Jolla  $\triangle$ 14C data used as background for the  $\triangle$ 14C calculations were extrapolated from the data for La Jolla to 31 May, 2010, from the time series extending from 7/1/92 to 12/7/07 (Graven et al., 2012).

8. P5786: Which meteorological observations have been used to constrain the WRF winds, and at what resolution?

The meteorological observations used to create the WRF wind fields: a) for the footprint in Figure 1b were initialized/nudged with Global Forecasting System (GFS; NOAA) winds and compared with a number of observations, all delineated & explored in the manuscript: Angevine, W., L. Eddington, K. Durkee, C. Fairall, L. Bianco, and J. Brioude, 2012: Meteorological model evaluation for CalNex 2010. Mon. Wea. Rev. doi:10.1175/MWR-D-12-00042.1. These are also described in Kort, E.A., W.M. Angevine, R. Duren, and C.E. Miller, Surface observations for measuring urban fossil fuel CO2 emissions: minimum site location requirements for the Los Angeles megacity, J. Geophys., Res., doi: 10.1002/jgrd.50135, 2013. b) for the WRF-STILT predictions were the radar wind profiler at Los Angeles International Airport (LAX). We have added some clarification for meteorological model evaluation for winds as follows: Following methods described in Jeong et al. (2012), we compared WRF-simulated winds (modeled at a resolution of 4 km) with data from NOAA 915 MHz radar wind profiler (ftp://ftp1.esrl.noaa.gov/psd2/data/realtime/Radar915/) located at the Los Angeles Airport (LAX; lat = 33.94, lon = -118.44), the closest wind profiler to the GHG measurement site. Comparing WRF and profiler winds at a height of 182 magl (the lowest available profiler level) for observation hours (13 - 18 hours, local time), and removing (> 3 sd) outliers, the mean difference (1.1+/-0.7 m/s, 95% C.I.) is consistent with the expected (1 m/s) measurement accuracy of profiler (Coulter, 2005), the best-fit slope (1.1+/-0.2) from a reduced chi-square fit is consistent with unity (Press et al., 1992), and the RMS difference (2.1 m/s) is consistent with results for previous similar comparisons reported in Jeong et al. (2012).

9. Figure 1A: it would be helpful if both of these maps were on the same scale.

Figure 1a has been redone to be on the same scale as Figure 1b.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 5771, 2012.

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