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Interactive Comment

# *Interactive comment on* "Toward consistency between bottom-up $CO_2$ emissions trends and top-down atmospheric measurements in the Los Angeles megacity" *by* S. Newman et al.

**S. Newman et al.**

sally@gps.caltech.edu

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Abstract, p. 29593 l.23 The absolute agreement between the bottom up Hestia and top- down approach is not assessed. Therefore, the word "consistent" may be misleading. At some latter point in the manuscript it may be worthwhile to clearly state that emission inventories are not validated absolutely, but only trends (and relative contributions) of bottom-up and top-down approaches are compared. We have made a minor modification in the title to clarify that this study compares trends in emissions between top-down atmospheric data and bottom-up inventories and data products, not the absolute emissions. The title is now: Toward consistency between trends in bottom-up CO2

emissions and top-down atmospheric measurements in the Los Angeles megacity

In the abstract, only variations, seasonality, and trends are discussed. Hopefully, the change in the title will clarify the focus.

p. 29597 l.12 The PDB scale has long been replaced by the VPDB scale and it is recommended to use the VPDB scale. Thank you for finding this error. It has been corrected, both in the text and in the references.

p. 29597 l.14-17 In this study, an integrated 14C(CO2) sample is obtained by combining 3-7 CO2 samples (afternoon) into one sample. Thus, 14C(CO2) from this integrated sample provides the average fossil fuel CO2 offset. In this manuscript, the monthly-integrated source signature $\delta$ss is used to obtain a value for $\delta$ff. However, averaging the source signature $\delta$ff over time is only valid if $\delta$ff and $\delta$bio do not change over time as otherwise correlation between cff and $\delta$ff and cbio and $\delta$bio can lead to biases (Vardag et al., 2015). As the CO2 samples are always taken during the same (short) time of the day and the integration period is not long, it may be that the effect of the integration is small. However, it might be worthwhile to check and mention this in the manuscript. This is an interesting point. The Miller-Tans plots suggest that monthly average compositions for $\delta$xs are constant, within uncertainty, as indicated by the very high correlation coefficients (Fig. A1). Although this does not guarantee that $\delta$ff and $\delta$bio do not change over time, it does suggest that the system is stable over this time scale. A comment has been added to this effect in Section 2.3.3.

p. 29599 l.20 Please give information on the background $\Delta$14C values such as sampling resolution, precision etc. The background $\Delta$14C record at Pt. Barrow, AK is obtained through the UCI/NOAA ESRL (Earth System Research Laboratory) flask network program that collects whole air samples using 6-L, 1-valve stainless steel canisters (Silco Can, Restek Co.) that have been pre-evacuated at UCI. The canisters are pressurized to $\sim$2 atm using an oil-free pump. Two biweekly samples were collected before 2008, and one weekly afterwards. For the period from 17 June 2005 to 17 March



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2006, some duplicate samples were collected using 32-L, 1-valve stainless steel canisters. Subsamples were then taken from these samples for 14C analysis. CO2 is extracted cryogenically at UCI then converted to graphite by the sealed tube zinc reduction method (Xu et al. 2007). Each sample is ∼2.7 mg C in size. Analysis of Δ14C is performed at the W M Keck AMS facility at UCI with total measurement uncertainty of ±1.3–2.4‰Ṁass dependent fractionation is corrected for using "on-line" δ13C measurements during AMS analysis, which accounts for fractionation that occurred during graphitization and inside the AMS.

Comparison was made over 22 common sample dates spanning 5 yr, differences in measured Δ14C from Barrow between the UCI and the Scripps Institution of Oceanography's CO2 Program. It shows differences in measured Δ14C are consistent with the reported uncertainties and there is no significant bias between the programs (Graven et al., 2013).

The other is the inter-comparison of AMS-based atmospheric 14CO2 measurements organized by the NOAA Earth System Research Laboratory, Boulder, Colorado. UCI lab is one the three groups having interlaboratory comparability within 1‰ for ambient level 14CO2 (Miller et al. 2013).

This information has been added to Section 2.3.2.

p. 29601 Section 2.3.3. The samples used were all taken during the afternoon hours. However, Miller and Tans (2003) have pointed out that the determination of source signature does not work when CO2 sinks with a different signature than the sources occur. If this is the case in your setting, it may lead to potential biases of the source signature, which should be discussed here. There are minimal, if any, sinks for CO2 in this region. The biosphere, as indicated by our data, is not an important player, in general.

p. 29602 l.11 The authors use δbio of -26.6, δng of -40.2 and δpet of -25.5 without stating an uncertainty or typical variation within one year. It should be elaborated how



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these uncertainties (especially on a seasonal scale) influence the results for cff, cbio, cpet and cng. This should also be included in Fig. 3. We do not have a measure of the uncertainty for $\delta$bio, although the sensitivity studies of Bakwin et al. (1998) suggest little variation. However, we have analyzed $\delta$ng and $\delta$pet over many years and at different times of year and the standard deviations are $\pm$ 0.5 and 1.0 ‰ respectively. We did, in fact, expect a seasonal variation in $\delta$pet, because there are winter and summer blends of gasoline. However, we have observed no significant differences.

p.29606 l.16-22 This section is a bit confusing as the assumption of having no biospheric influence is not correct and also not used in this study. It might be more straightforward to leave this passage out as it seems to be of no use for the reader at this point. Thank you for pointing this out. This was mentioned above and therefore is redundant here. It has been deleted.

p. 29607 l.5 Throughout the manuscript emission and concentration are used synonymously. Without a model, only the contribution of fossil fuel CO2 can be derived but not the fossil fuel CO2 emission. Please correct this in the entire manuscript. Here and in many other places throughout the manuscript fossil fuel “emissions” has been replaced by Cff.

p. 29608 l.9 Jiang et al. (2012) concluded that the semi-annual oscillation is a consequence of a combination of gross primary production (GPP) and respiration (resp), not net primary production (NPP=GPP+resp) and respiration. CO2 semi-annual oscillation is a consequence of a combination of gross primary production and respiration. We have revised this in the main text.

Fig. 11 It might be worthwhile to insert error bars on cpet and cng.âĂĺ Cpet and Cng are now shown as annual averages with standard error bars.

p. 29612 l.2 Could the lag be an artifact of not including seasonal variations of the source signatures (e.g. $\delta$bio) into the consideration? We conclude that the major cause of the seasonal variations is the seasonal shift in wind direction, resulting in a different



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source region for winter versus summer. None of our information suggests seasonal variations in $\delta$bio.

Fig. 12 The uncertainty of cpet should be included in this Figure. Why do EIA and ARB statistics differ by a factor 10ËĘ3 ? Please elaborate what “mo” stands for in the unit of [kg CO2/mo] ? We have the uncertainties plotted in Figure 6 and feel that Figure 12 will be too cluttered if they are included here again. Thanks very much for noticing the error of 10ˆ3 in the units for the EIA data. This has been corrected. [kg CO2/mo] refers to kg CO2 per month. This has been added to the caption.

p.29612 l.20 They agree in the direction of the sub-annual variation, but not in their absolute values. A phrase has been added to say that they agree in their seasonality.

Fig. 14 Same emission sectors should have the same y-scale so that differences between emission inventories become obvious. This is the case for the green and black axes in Fig 14. The blue axis is for the LA basin Hestia product, covering a much smaller domain, as does the grey axis, which is for LA and Orange county power plant emissions. The major point being portrayed in this figure is the seasonality. The absolute numbers do not really matter, since the temporal variations are being emphasized.

p.29613 l.6-7 It is interesting that the increase in natural gas consumption is seen earlier in the data than in the emission inventories. If other sectors have not changed significantly (as one might expect?), this might point towards a false emission inventory and might be worth to point out here. This is a very good point, and we have added a sentence mentioning possible explanations for the different timing. We feel that we cannot suggest a false emission inventory because of the large uncertainties and the lack of atmospheric modeling here.

p. 29613 l.27 What about long-term changes in source signature of natural gas or petrol? Only if the endmembers of the signature do not change over time it is possible to validate emission inventories as presented here. We have analyzed natural gas combustion over 30 years and gasoline combustion over 9 years and do not see any



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significant changes. A comment to this effect has been added in Section 2.3.3.

p. 29615 l.9-14 What are typical uncertainties of the emission inventories used? What are the uncertainties of the top-down approach? Uncertainties are not given for the bottom-up inventories. We do not produce an emission inventory with our top-down approach. We conclude that we can see a significant 10 % change in Cff using our measurements.

p. 29638 Fig. A3 Please explain why the long-term trend (last row) changes after removing the average (repeated seasonal cycle). The major reason for the change in the long-term trend between the analysis of the raw values for Cff and those after removing the seasonal cycle is that the uncertainties are overwhelming for the raw data analysis and are much smaller after removing seasonality, which is a common artifact of EEMD known as mode mixing between the dominant mode (i.e. annual cycle in our case) with other modes (e.g. noise). In our case, we can minimize the mode-mixing problem by removing the annual cycle and perform EEMD again. The variations observed are not significant for the raw data analysis, whereas they are for the modified data set. In the revised text: "Note that there are severe mode mixing problems in IMF3 (e.g. during 2011–2013) between the dominant annual cycle and subseasonal variations, which also affects the nonlinear decompositions in the higher modes. To minimize the effects of mode mixing on the extractions of inter-annual trends, we perform the EEMD again after removing the average annual cycle (minus the mean of the raw data), defined as monthly averages over the entire time period (2006-2013; resulting time series shown in Fig. 10e). The revised inter-annual trend is shown in Fig. 10f."

Technical corrections: p. 29594 l.13 This is only true in very large cities (Megacities). We have added the phrase "especially in megacities."

p. 29595 l.27 It is not clear what "all three" refers to here. We have added a parenthetical remark to explicitly list "all three sources" for clarification.



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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C13047/2016/acpd-15-C13047-2016-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 29591, 2015.



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