Interactive comment on “Measurement of fossil fuel derived carbon dioxide and other anthropogenic trace gases above Sacramento, California in Spring 2009” by J. C. Turnbull et al.

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Received and published: 28 December 2010

We appreciate the thoughtful comments from the reviewers. Both reviewers mentioned as their main comment that our small (two afternoon flights in February/March) observational dataset makes the comparison with annual emission inventories difficult. We agree. In this manuscript, we hope to convey the potential of this type of observational measurement to constrain emissions, using our dataset to demonstrate the feasibility. For several of the species that we examine, other independent observational datasets have been measured, and our comparisons with inventories are consistent with those other studies. We have revised the text to emphasize this caveat.

Anonymous Referee #1 Received and published: 17 October 2010 General Comments: This well-written paper presents an interesting study in which multiple trace gas measurements from a campaign-based flask and in-situ data set are analyzed. The analysis is soundly and well structured also giving all details on sample retrieval and treatment. Using 14CO2 as tracer for fossil fuel CO2 is a well established technique, yet the use of fossil fuel CO2 as a means to improve knowledge about the emission of other anthropogenic tracers (such as VOC, . . .) in an urban environment is not discussed thoroughly in other publications. This study conclusively shows how a combined interpretation of 14CO2, CO2, CO, VOC and other trace gases can be very beneficial. C8802 One issue which should be further discussed in this study is a potential bias of the flux (and x:CO2ff ratio) estimates due to the limited amount of data. Both flights presented were conducted within 8 days and always at the same time of day 2 pm – 5 pm. Hence any seasonal and diurnal variation of the fossil fuel CO2 fluxes and the tracer:fossil fuel CO2 ratio are not accounted for. The question arising is: Are the fluxes derived during these 3 hours at 2 days comparable to mean annual emission estimates? Although this is mentioned, it should be further discussed as the results indicate a strong influence from road traffic emissions (p21580 l.24/25) which is known to have a substantial diurnal variation of emission (Easily a factor of 2-3 during rush-hours compared to other times of day). Hence the derived flux estimates could be biased towards higher values. Furthermore, the emissions from domestic heating (usually) show a distinct correlation with the ambient temperature thus this might also lead to a bias in the estimate of the CO2 ff fluxes and the mean annual ratio of e.g. CO:CO2ff as this should vary within in the year as the fossil fuel CO2 source mix changes (Rather constant emissions from the road traffic sector throughout the year, while domestic heating emissions vary from winter to summer). A typical mean diurnal cycle of road traffic emissions and the seasonal variability of other sources should/could be used to adjust the estimated fluxes to derive a more "representative" estimate to compare with the mean annual fluxes from bottom-up inventories. Besides this single issue, that in my opinion needs further discussion, only minor changes are
suggested. As beside the high quality the overall topic of this study is completely in
the scope of ACP and I recommend this paper for publication, after re- visiting the
mentioned point. Specific and technical comments:

p.21571 l.22 At this point I think an influence from biomass burning on the local CO2
cannot be generally ruled out and should be mentioned. Authors: Done.

p.21573 l.20 As the water correction for the CO2 data of SAC306 is quite large (5ppm)
the uncertainty of this correction would be of interest and if this is accounted for in the
latter calculations? Authors: The uncertainty due to the water correction is factored into
the overall in situ CO2 uncertainty (and it is smaller than other sources of uncertainty
in the measurement). The CO2 uncertainty is much smaller than the uncertainty in
\( \Delta^{13} \text{CO}_2 \), and doesn't have much effect on the calculations of CO2ff and CO2bio.

P21577 l.11 Besides the respiration (from the soil) do you also “principally” consider
an influence from BMB to CO2_other? Authors: Added to text.

P21578 l.18/19 The comparison to the tower measurements seems to also have some
potential to further investigate the diurnal changes of at least the CO:CO2 ratio using
the Walnut Grove in-situ measurements and might be worthwhile including. Authors:
We considered including the tower measurements and a discussion of them in this pa-
per, but since they cover a longer time period (and are continuing), and many different
wind regimes, we believe that this discussion would confuse this current paper, and
that the interpretations of these measurements deserve their own (forthcoming) paper.

P21579 l.4 " removal of CO" is negligible. . . because the samples were collected
within a day of emissions from the source region” Your assumption seems to rule out
any influ- ence from sources farther away. Without a conclusive modeling result I would
hesitate to believe that all of the excess CO2, CO2ff, and so on, originate from Sacra-
mento alone. Usually I would expect the footprint to be larger maybe also including
surround- ing counties/states. Authors: Indeed. We have added a clarifying sentence.

P21579 l.20ff For the comparison of the derived CO:CO2ff ratios from the flasks with
the vehicle tail pipe emissions the representative issue arises again (see general com-
ments) Authors: See general comments.

P21580 l.24/25 the found “strong influence from road-traffic” may imply that the diurnal
variability of fluxes (and emission ratios) from this emitter group has to be more deeply
discussed. (see general comments) Authors: See response to general comments.

P21582 l.23/24 I would assume that the given 10-20% uncertainty derived from the
comparison of two bottom-up inventories should be regarded as lower bound for the
uncertainty, as both bottom-up techniques surely share a significant amount of statis-
tical information and might both have similar methodological biases (missing sources,
emission displacements). Authors: Agreed. Text revised to clarify.

P21583 l.28 Given the (seasonally) changing contributions of different emission sectors
to the CO2ff and CO overall emission was there a specific reason assume “flat emission
throughout the year” for Sacramento County? Authors: Higher temporal resolution
data is not available for CO, nor for CEPAM CO2ff. Vulcan CO2ff does have higher
temporal resolution, but is for 2002 only (our data is extrapolated from the 2002 values),
and without temporally varying CO inventory information, varying the CO2ff inventory
would not be very useful.

P21584 l.4 What is the rational for an exponential extrapolation of the emission de-
crease? Authors: Added clarifying sentence in the text.

P21485 l.14 At this point the potential influence of the sampling interval and the com-
parison of campaign data to annual mean fluxes is mentioned, I guess here the needed
further clarification on that issue would be very beneficial. Section 3.6. Using a mass-
balance approach is a generally valid approach, yet many assumptions have to be
made (single source etc.). Nevertheless even with the limited accuracy of this approach
nice results are found, still to fully exploit the available an accompanying modeling
study would have been insightful. Authors: We agree that a modeling study would
be interesting, however, we believe it is beyond the scope of this paper for a number of reasons. As noted in the paper, and by both reviewers, it is difficult to compare observations from two flights with the annual inventory data that is available. This would not be solved by a modeling study. Running a regional model such as WRF is not a trivial undertaking, and we believe that for a difficult-to-model region such as the California Central Valley, such a modeling study would require enough work that it deserves a separate paper. Given amount of information available from our data-based study, we believe we are justified in leaving the modeling work for another paper.

P21591 l.5 Again comparing annual means with this campaign-based data maybe needs careful attention. Figure 4. A combined fit for SAC227 and SAC306 seems not reasonable for total CO2, as you pointed out they seem to display two different regimes. Authors: We believe this comment refers to figure 2, not figure 4. Changed.

Caption Fig.4. In the given color version: CO: CO2ff (black line) -> CO:CO2ff (green line), CO:CO2 (grey line) -> CO:CO2 (blue line) Authors: Caption revised. Thanks for picking up this error.

Figure 7. Changing dates to more human-friendly ones, (time of day in local time or UTC) could ease the interpretation of this plot. Authors: Done.

Anonymous Referee #2 Received and published: 18 November 2010 In this paper Turnbull et al. present a very interesting study on how to infer CO2 and other anthropogenic trace gas emissions from airborne in-situ measurements, using the carbon 14 content of CO2 to trace fossil fuel emissions and CO to partition the CO2 fossil fuel and biospheric contents. Using data recorded during a two-flights cam- paign other Sacramento area on 27 Feb. 2009 and 6 March 2009, they constrain the fossil fuel CO2 mole fraction in the Sacramento urban plume from DË˘G14Cmeasurements, analyze the urban signature of this plume and compare the results with bottom-up inventories for several species. An attempt to partition the biospheric from the anthropogenic signal is provided using the ratio of CO over CO2 as a tracer for fossil fuel CO2 emissions. A

mass balance approach is carried out to estimate the fossil C10027 fuel CO2 emission flux from the Sacramento urban area. General comments The paper is very well constructed and well documented. The introduction summa- rized the state-of-the-art on uncertainties in local to regional scale emission inventories of anthropogenic species. The instrumentation used is fully adapted to the study. A strong effort to infer fossil fuel emissions from airborne observations is remarkable, as well as the attempt to compare these results to existing inventories. However, there are a few points that need to be improved: a better description of the studied site (geography, size, population; eco- nomical activities in Sacramento...) ; meteorological information during the campaign (even if no meteorological data were recorded) ; data precision and accuracy in the instrumentation description ; precision on the definition of anthropogenic CO2 that is resumed to fossil fuel CO2 without explanation before page 21588 (we learn only on that page that there is no other anthropogenic CO2 sources than fossil fuel combus- tion). Another point I am very concerned with, and that needs revision before publi- cation, is the comparison undertaken between emis- sions inferred from some hours of airborne observations to annual inventories. The notion of variability or seasonal- ity in the emissions is not treated. I recommend that the authors be more moderate in their conclusions and discuss the potential variability of anthropogenic emissions in Sacramento urban area. They could retranslate it into un- certainty bars associated to their results and discuss the results plus their uncertainty in the comparison to annual inventories. My point of view is that this paper represents a very nice work that puts together several existing methods and makes of it a inovative approach to the problem of urban emis- sions quantification. It is in the scope of ACP and suitable for publication in this journal after revision of the above mentionned point and other mentionned minor revisions. Some other details to correct are listed below.

Detailed comments C10028Title: Since the paper focuses on emissions, and the cam- paign occured in Winter and not Spring, I suggest to change the title for the follow- ing: "Assessment of fossil fuel carbon dioxide and other anthropogenic trace gases emissions from airborne measurements above Sacramento California in Winter 2009":

C11702

C11703
Institutions should all be shortened or long but all given in the same format. Authors: Titles are always a challenge – thank you for this suggestion!

p.21569 Line 7 : please define what you mean by "recently" Authors: We have added clarifying statement in introduction section.

Line 15: "overestimate": please quantify Authors: Done here and elsewhere in paper.

Line 16: "substantial differences": give numbers Authors: Changed.

Line 20-22: give numbers Authors: Done.

Line 24: remove "however" Authors: Done.

p.21570 The introduction lacks a description of Sacramento's activities (main emission sectors, at least for CO2) and information on dominant winds, plus meteo information during the flights event if they are not be provided by in-situ measurements. Authors: Added this discussion.

p.21571 Line 15: same remark for "recently" Authors: Done.

Line 17: remove "because CO2...greenhouse gas" Authors: Done.

Line 19: After the point, add the last sentence of the section line 29 "Atmospheric ob-

servations of CO2ff ...described" (remove Finally at the start of the sentence). Authors: We did not change this, as we believe the paragraph is clearer as is.

Line 20: Define what is CO2ff. The definition here matches with anthropogenic emissions ie FF-ciment production + biomass burning. It is neces- sary to explain here that ciment production-biomass burning are negligeable and thus CO2antropogenic equals CO2ff. Authors: CO2ff is defined earlier in the introduction as the fossil fuel CO2 contribution, which includes CO2 produced from fossil fuels, principally coal, oil and natural gas. By definition, fossil fuel does not include biomass burning or cement production.

Line 28: "Not perfectly known": be more precise and give numbers Authors: This is described in detail in later sections, and we prefer not to clutter the introduction with a long explanation.

p.21572 Line 3: add after fraction "inferred from A Ì´lDË ˘GC14CO2" Authors: Yes.

Line 20: two-weeks campaign Authors: We believe that the grammar we used is appropriate.

p.21573 Line 11: give the precision on the standard concentration Authors: Added a reference for the standards.

Line 20: give the precision for CO2/CH4/H2O data measurements Authors: Done.

p.21574 Line 25: then give the distance that the plane has been travelling during the 2 mn flask filling. Authors: In fact, while the flasks are filled over 2 minutes, the bulk of the sample is taken during the last ∼20s of the fill. This has been corrected in the text.

p.21576 Lines 20-23: these lines are unclear to me Authors: Have rephrased for clarity.

p.21577 Line 16/17: explain what you mean by "reasonable" Authors: The further part of the sentence justifies this term.

Line 18: explain what you mean by "as expected" Authors: This is explained in the parenthetic statement at the end of the sentence.

p.21578 Lines 22-24: explain this before in your paper Authors: It is not clear what this comment is referring to.

p.21580 Line 3: Is there any reference on biofuel? To what extend it is developed in California / Sacramento? Authors: This is discussed later in the paper.

Line 18: any R2 calculation has to be associated with a statistic significance test. Please calculate and give the p-values associated to the R2 coefficients. Authors: True, but in this case, where the coefficients of determination (r2) are so high, the p-
values are <0.0001 in most cases, excepting, of course, those cases where r2 is low, which we discuss in the text.

p.21581 Line 7-9: How is the BL height measured or estimated? Authors: We removed this line from the text (see later in the paper for discussion of the boundary layer height estimate).

Line 14: OK but you rather want to precise this earlier in the text Authors: It is difficult to bring up this discussion of the lack of biomass burning signal earlier in the text. We believe that the flow of the text is best left as is.

p.21584 Line 22-26: how is this done? Is there any seasonality in emissions? This point is not treated, and comparing a campaign of a few hours to a whole year is quite challenging... please moderate your text and try to treat the seasonality factor. Authors: See general comments.

p.21585 Lines 6-8: Thus comparing to annual inventories does it make sense??? Authors: See general comments.

p.21588 Lines 1-5: this should be explained before, as already explained above Authors: Again, we have tried to make the paper flow as best as possible, and it is difficult to bring this up earlier in the paper.

p.21589 Line 23: How far is the hypothesis on a constant wind speed correct? You can look at wind reanalysis, make estimates and produce error bars. Authors: In the absence of high-resolution wind information or high resolution reanalysis, we obtained four estimates of wind speed (two from the WGC tower downwind of Sacramento, and two from the NCEP reanalysis). We used the maximum and minimum wind speed values, along with maximum and minimum boundary layer heights in our calculation. There is a large range in the wind speed values, and we do not have a ready method to ascertain which is most correct. We report the range of possible values from these calculations, and believe that this is a sensible way to show the range of possible values of the CO2ff flux in this case.

p.21590 Lines 22-23: Please give a reference Authors: There is no reference for this, we simply estimated the boundary layer height from our data.

p.21591 This section has to be moderated by taking into account a possible seasonal/ meteorological variability. Authors: See general comments.

p.21592 Line 10: permits Authors: We believe that the current grammar is correct. Line 25-27: again, what about seasonal variability? p.21593 This section has to be moderated by taking into account a possible seasonal/ meteorological variability. Authors: See general comments.

References p.21594 Line 25: add a coma between both authors. Authors: Done.

p.21597: same remark as before Table 1 is column 1 really needed? Authors: Yes, as the data reported here are from LLNL, and we are comparing with UCI to show that the LLNL data are consistent with our larger and longer UCI dataset.

Table 2 Add a reference for CEPAM Authors: This is referenced in the text, as are the other inventories.

What do you mean by "VULCAN... (high value)"? Authors: The inventory values are given as a range.

Why in the legend do you mention the abbreviation "bl" that does not appear in the table itself? Authors: Have revised the table to clarify.

Add a column with the p-values associated to the R2 significance test. Authors: See comment earlier.

Table 3 I think that the hypothesis is not correct. How could CO2 emissions calculated on a few hours be consistent with emissions from a whole year? Do you have access to seasonal inventories (winter data)? Add a column with the times of the flight segments.
Authors: See general comments.

Figure 1 Add the distance scale (axe on the top from zero to distance max), and draw the city borders. Authors: The distance can easily be determined from the lat/lon's. The city does not have an obvious border; hence we used the county borders instead.

Figure 2 Add (ppb) after CO on the y-axis legend. Authors: Fixed.

Figure 4 What means "short tons" on the y-axis? Authors: This is the unit of measure used for reporting CO emissions in the USA.

Figure 5a It seems we have the error on the x-axis only, not on both coordinated as indicated in the figure caption?? Authors: Y-errors are often smaller than the symbol size. Noted in caption.

Figure 6 The text on the figure seems too small. Authors: Perhaps this can be included in the final version as a larger figure? It is difficult to increase the font size without making it confusing.

Figure 7 It would be better to add a light grey color rectangle around the data in the BL rather than a line. The range of uncertainty is very large (0.2-2 ppm: how come this large range of uncertainty??) Authors: We believe adding the rectangle around the data would be more confusing. When CO2ff is calculated using RCO:CO2ff and CO values, the uncertainty is usually dominated by the uncertainty in RCO:CO2ff, and the uncertainty scales with the magnitude of CO2ff. For low values of CO2ff, however, the uncertainty is dominated by the uncertainty in the CO measurement itself, which results in the lower limit on the CO2ff uncertainty.

Figure 8 This sounds very strange, to get such negative and positive biospheric fluxes at the same moment...can you explain this?? Authors: This is presumably because net ecosystem exchange varies in sign across different landscape elements. This is discussed in the text.

Figure 9 It lacks a legend fot the insert (insert instead of inset). Authors: Fixed.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21567, 2010.