

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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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.

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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 sound and well structured also giving all details on sample retrieval and treatment. Using $^{14}\text{CO}_2$ as tracer for fossil fuel CO_2 is a well established technique, yet the use of fossil fuel CO_2 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 $^{14}\text{CO}_2$, CO_2 , 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: CO_2 ff 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 CO_2 fluxes and the tracer:fossil fuel CO_2 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 di- urnal 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 CO_2 ff fluxes and the mean annual ratio of e.g. CO: CO_2 ff as this should vary within in the year as the fossil fuel CO_2 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

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

p.21573 I.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 $\Delta^{14}\text{CO}_2$, and doesn't have much effect on the calculations of CO₂ff and CO₂bio.

P.21577 I.11 Besides the respiration (from the soil) do you also "principally" consider an influence from BMB to CO₂_other? Authors: Added to text.

P21578 I.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 paper, 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 I.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 influence 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 Sacramento alone. Usually I would expect the footprint to be larger maybe also including surrounding counties/states. Authors: Indeed. We have added a clarifying sentence.

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P21579 I.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 comments) Authors: See general comments.

P21580 I.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 I.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 statistical information and might both have similar methodological biases (missing sources, emission displacements). Authors: Agreed. Text revised to clarify.

P21583 I.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 I.4 What is the rationale for an exponential extrapolation of the emission decrease? Authors: Added clarifying sentence in the text.

P21485 I.14 At this point the potential influence of the sampling interval and the comparison 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

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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 I.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 campaign 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 DE¹⁴C measurements, 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

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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 summarized 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; economical 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 combustion). Another point I am very concerned with, and that needs revision before publication, is the comparison undertaken between emissions inferred from some hours of airborne observations to annual inventories. The notion of variability or seasonality 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 uncertainty 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 innovative approach to the problem of urban emissions quantification. It is in the scope of ACP and suitable for publication in this journal after revision of the above mentioned point and other mentioned minor revisions. Some other details to correct are listed below.

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

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Institutions should all be shortened or long but all given in the same format. Authors: Titles are always a challenge – thank for you 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 observations 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 necessary to explain here that ciment production+biomass burning are negligible and thus CO₂anthropogenic 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.

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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 $\Delta^{14}\text{CO}_2$ " 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 $\text{CO}_2/\text{CH}_4/\text{H}_2\text{O}$ 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 ~ 20 s 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 extent it is developed in California / Sacramento? Authors: This is discussed later in the paper.

Line 18: any R^2 calculation has to be associated with a statistic significance test. Please calculate and give the p-values associated to the R^2 coefficients. Authors: True, but in this case, where the coefficients of determination (r^2) are so high, the p-

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values are <0.0001 in most cases, excepting, of course, those cases where r^2 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

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of the CO₂ 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.

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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/lons. 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 CO_{2ff} is calculated using RCO:CO_{2ff} and CO values, the uncertainty is usually dominated by the uncertainty in RCO:CO_{2ff}, and the uncertainty scales with the magnitude of CO_{2ff}. For low values of CO_{2ff}, however, the uncertainty is dominated by the uncertainty in the CO measurement itself, which results in the lower limit on the CO_{2ff} 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 for the insert (insert instead of inset). Authors: Fixed.

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

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