

Response to the review of # acp-2013-915 by Zhang et al.

We thank Reviewers for their very thoughtful comments, criticisms, and detailed suggestions. Each comment has been addressed below (review comments in black; author responses in red).

Reviewer 1

Does the paper address relevant scientific questions within the scope of ACP?

1. Does the paper present novel concepts, ideas, tools, or data?

The paper compares evaluates different methods to estimate regional fluxes, none of them novel, although applying the EQ method to CH₄ and N₂O is a first ever. The comparison does have considerable value however.

2. Are substantial conclusions reached?

Yes, the relative uncertainties and merits of the various methods are well discussed. However, since the top down methods are presented here as an independent means to verify bottom up reported missions, I believe it is a missed opportunity that the authors did not include the emissions reported in the national GHG inventory system that must be available for the region. I would love to see also the bottom-up NIR estimates according the IPCC guidelines, because its verification is the main motivation for the work presented here.

We concur with the reviewer that various top-down methods used in this paper can provide important information to verify and constrain the bottom-up reported emissions. Therefore, we compared the flux estimates of three major greenhouse gases using top-down methods (including EQ and EC) with a fossil fuel emission inventory from CarbonTracker and the EDGAR inventory, the latter of which are widely used anthropogenic GHG inventories based on the IPCC guidelines:

1) We used the flux aggregation (FA) method (section 2.5) to estimate the bottom-up CO₂ flux, which includes the biogenic fluxes measured at four major land cover types and the anthropogenic fluxes from a fossil fuel emission inventory used in Carbon Tracker. The comparison indicates that EC (a top-down method) and FA (a bottom-up method) have relatively good agreement with the regional CO₂ flux, while the EQ method underestimated the CO₂ uptake in July. The detailed comparison was discussed in Sections 4.1 and 4.2.

2) We compared the CH₄ and N₂O flux estimates from the EQ method with the EDGAR inventory, and found that the EDGAR inventory may underestimate CH₄ and N₂O emissions significantly. Details were presented in section 4.5.

Inspired by the reviewer's comments, we also looked into the national GHG inventory system developed by the United States Environmental Protection Agency (US EPA, 2014). The details of this comparison are presented in Section S6 of the supplementary materials and the key results are discussed in the paper. See section 4.5 of the main text.

3. Are the scientific methods and assumptions valid and clearly outlined?

If any, this is the only weak point in the paper. The methods are not always exhaustively described. I would prefer to learn more about the details perhaps in the form of supplementary material. See 5 below.

4. Are the results sufficient to support the interpretations and conclusions?

Yes. Although after the methods/results sections many questions remain, these are largely answered by the very good section 4 discussion.

5. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

No. A few examples:

Tall tower EC: it is unclear how exactly the storage term is computed (F_s in eq 1). which observation heights? interpolated?.

Why the (arbitrary?) limit of $-4 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for discarding F_s in the morning transition?

We have added Section S1 in the supplementary materials that address the questions related to the storage term in the tall-tower eddy covariance method.

was the EC system ever above the night time SBL?

It is possible that the top of the Stable Boundary Layer (SBL) was lower than the EC system installed on 100 m level especially during the winter months. The median nocturnal boundary layer height at the site is about 200 m (Griffis et al., 2013). It is well known that the eddy covariance method does not perform well in such stable atmospheric conditions. Therefore, we used the friction velocity (u_*) as a quality control (Davis et al., 2003; Goulden et al., 1996), to discard the flux data measured during such stable conditions.

how exactly monthly mean composite diurnal variation (equation)?
effect of this compared to gap filling (in discussion section)?

We estimated the monthly flux from the hourly fluxes using a “diurnal composite” method with the following steps:

1. Compute an averaged diurnal cycle for a month by averaging all of the available data within each hour (from 0:00 to 23:00).
2. The monthly mean flux, therefore, represents the average of the diurnal composite. Further, the annual flux represents the average of the monthly fluxes.

Therefore, when no data gap exists, the monthly value from the “diurnal composite” method is the same as the monthly mean of all available data within the month. To evaluate the uncertainty associated with data gaps, we performed a Monte Carlo simulation following Griffis et al. (2003): We randomly removed 30% of the data for each month, and recorded the calculated monthly and annual fluxes following the same data processing procedure described above. By repeating this simulation 5000 times, we determined the standard deviation of the annual flux estimates.

This approach avoids the propagation of uncertainties associated with gap filling procedures (i.e. assumptions and uncertainties related to light-response and respiration functions). This approach is usually more stable than semi-empirical gap-filling methods, therefore, it can provide a relatively good estimation even when large percentage of data is missing (Falge et al., 2001) Further, at this time, we do not know of a reliable method for gap filling strategy to deal with N₂O and CH₄ fluxes. For consistency, we treat each GHG the same.

in method section it is claimed no gap filling is done, in section 3.1 suddenly there is gap filling (but only for one specific month?)

We thank the reviewer for pointing out this problematic statement. We changed the statement to “We estimated the monthly mean from the diurnal composite of the CO₂ flux based on valid observations (Section S2 in supplementary materials).” In the context, this statement explains how we calculate the monthly flux from the hourly flux.

However, the gap filling that we describe in section 3.1 was carried out in order to estimate the annual flux for 2009, when one month of data was missing. The monthly flux for June, 2009 was not available due to problems with instrumentation. Therefore, we estimated the flux for June, 2009 by assuming the CO₂ flux trend from May to July in 2009 was similar to the trend in 2008 and was calculated as:

$$\frac{F_{5,2008} - F_{6,2008}}{F_{6,2008} - F_{7,2008}} = \frac{F_{5,2009} - F_{6,2009}}{F_{6,2009} - F_{7,2009}}$$

In the equation above, F_{5,2008}, F_{6,2008}, F_{7,2008} are mean CO₂ fluxes for May, June, July, of 2008; while F_{5,2009}, F_{6,2009}, F_{7,2009} are mean CO₂ fluxes for 2009.

EQ method: how is time averaging done of c+ and cm? what does `composite diurnal variations` here mean (equation)?

The time averaging from the hourly mean c to monthly c was carried out using the same “diurnal composite” approach described in Section S2 of the supplementary materials for the CO₂ flux.

CT give more detail on the version/product used

We added additional information to the method section 2.4.3 regarding the CT product. More information about the product can be found at http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2011_oi/documentation_CT2011_oi.pdf

The FA method: the regional flux is a simple area weighted average of the fluxes of the respective land cover types, or is it footprint weighted. Either way give equation. Either way give a table in section 2.1 with the fractional cover of all significant land cover classes. Now, it is unclear how much of the area is covered by grass/pasture and or forest, only that together they make up 40%. Fires are implicit in CT; any estimate from FA?

We improved Section 2.5 to provide a more detailed description, and hope it will address questions raised by the reviewer.

The CO₂ emission from Fire was not included in our FA method, because it was less than 1% of the CO₂ emission from fossil fuel for the tall tower region in 2009.

How exactly is the footprint map used ? Why only for EQ and not for FA? is this footprint the same as for EC? (the footprint is first mentioned only in section 3.1; move this to methods section; give footprint map (e.g. in SM)

We hope the improved Section 2.5 will address the questions here.

Briefly, we used two types of footprint to calculate the FA flux and compared the result with the estimates from top-down methods (EC and EQ flux). The footprint map derived from STILT analyses is for the concentration measured at 200 m. This footprint is not the same as that derived for the eddy covariance flux measured on 100 m.

We have added a footprint map for the concentration measurement on 200 m to the Supplementary materials according to the reviewer's suggestion.

in 3.3 somehow an annual CH₄/N₂O flux is computed? it remains completely unclear how that is done. details! equations!

We have added further details to section 3.3 to make this section more transparent. Additional information regarding the methodology can also be found in Zhang et al., (2014).

I suggest to add a supplementary section describing such details.

We thank the reviewer for pointing out the lack of clarity in the method part. We have now added supplementary materials to the paper.

6. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

yes

7. Does the title clearly reflect the contents of the paper?

No. The title suggests the paper is only about PBL methods, implicitly assuming their bottom-up method (Flux Aggregation) or the direct Eddy covariance at larger elevations can and should be taken as references. The title should better reflect the fact that there is no a-priori best method to determine regional fluxes and that this paper is about a more general methods comparison including both bottom-up and top-down methods.

We now change our title to "Estimating regional greenhouse gas fluxes: An uncertainty analysis of planetary boundary layer techniques and bottom-up inventories"

8. Does the abstract provide a concise and complete summary?

Yes

9. Is the overall presentation well structured and clear?

Generally yes, but see my point 4 above: quite some elements now very well discussed in section 4 come a bit late in the whole narrative. Now section 4 is really the core of the paper. Considerable parts could be integrated in the results section, which would make that part stronger

10. *Is the language fluent and precise?*

Yes

11. *Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?*

yes but... see above

12. *Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?*

I suggest to add a supplementary section describing some of the methods in more detail see 5 above.

In table 1 do not only present r^2 and NSE but also the (annual) flux magnitude of each method and the RMSE. The high values for the first two only show that the seasonal cycle is followed by each method.

We have added supplementary materials according to the reviewer's suggestion.

According to Moriasi et al. (2007) NSE suggests how well the plot of two sets of data (i.e. observed and simulated data) fits the 1:1 line, therefore, a high NSE value for EC fluxes and FA fluxes (i.e. $NSE > 0.75$) does not only suggest the two datasets share the similar seasonal cycle but also suggests that the absolute difference between EC and FA fluxes (considered as "noise") are relatively small comparing to the variance of the EC (or FA) fluxes (considered as "information").

We consider the magnitude of the annual flux and the NSE cannot reflect how well the monthly flux data from EC and FA methods compare. As reported in the paper, "in 2009, the tall tower's annual average EC flux was $-0.35 \mu\text{mol m}^{-2} \text{s}^{-1}$ ($-131 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$), while the seasonal variation was about $6 \mu\text{mol m}^{-2} \text{s}^{-1}$ ", about 16 times higher than the annual average". A small difference in the monthly flux will lead to a relatively large RMSE comparing to the annual flux.

Table 1 focuses on the comparison of monthly fluxes between methods, while Section 4.1 and Table 2 are dedicated to the comparison of annual fluxes between methods.

13. *Are the number and quality of references appropriate?*

Generally yes

14. *Is the amount and quality of supplementary material appropriate?*

No see 5 and 12 above

Miscellaneous

p3239, l 6 water vapor mixing ratio on the tall tower...which level?

The water vapor flux was measured at the 100 m level using the EC system, while the water vapor mixing ratios were measured at the 200 m level. We have added these details to the manuscript (The line below equation 4).

p3241, l12-17 This is too simple a discussion of the fetch of the EC observations. More sophisticated methods to determine the footprint exist e.g. Kljun et al 2002/2004, Vesala et al various reviews. Bring this in line with the better footprint estimate made a few paragraphs later for EQ

We used two types of footprint method. One is an equally-weighted circular footprint, and the other is derived from the Stochastic Time-Inverted Lagrangian Transport model (STILT, Lin et al., 2003) We have improved section 2.5 to address the reviewer's concerns.

p3244, l11-12 '...no prevailing wind direction...' strange statement, the more so because on p3248 l10-15 an explicit advection estimate is made considering a northwest prevailing wind

The statement in p3244 L11-12 is for the wind condition for the whole year 2009; while the statement in p3248 L 10-15 is for wind condition during the intensive observation period (September, 2009).

section 4.2 I am not an expert on this EQ method but eq 2 to me suggests that the flux at the PBL top considered here is a vertical advection term only, subsidence, $W \neq 0$. It neglects turbulent entrainment due to shallow cumulus, convection, $W=0$, $w' \neq 0$. However, you even confuse it more by combining eq2 with 3 and 4 respectively. In the first case, eq2+3, implicitly you account for all transport terms that cross the PBL top. Combining eq2 with 4 you do not! This justifies more discussion at least!

We agree with the reviewer that the ρW terms in equations 3 and 4 are different. The first one (equation 3) includes all of the transport terms while the other only includes the vertical advection term. However, the basic assumptions of the EQ method are that over long time scales (e.g. weeks) the boundary layer is in equilibrium and the horizontal advection and storage are negligible. Further, Helliker et al. (2004) reported that the large scale synoptic subsidence dominates the exchange at the top of the boundary layer. Therefore, the ρW term in equations 3 and 4 are essentially the same under these assumptions.

p3246, l22-23. 'If the EQ method' Bullshit statement. Delete whole sentence

As suggested we have removed this sentence.

p3247, l20-end nowhere in the paper is the effect of PBL dynamics discussed wrt the 200m observations; are these always in the mixed layer? can they be above the SBL at night? etc

According to the EQ method, the diurnal dynamics of the planetary boundary layer are ignored, because the EQ method assumed that over long timescales (e.g. weeks), the large scale atmospheric processes dominates, and the boundary layer is in statistical equilibrium (Griffis et al., 2013).

The uncertainties related to using the CO₂ concentration at 200 m as the CO₂ concentration in the mixing layer was discussed in Section 4.2.

p3248,l22-23 Direct measurements of these terms.... How ? Suggestions?

We have added a sentence to this paragraph, suggesting direct measurement such as aircraft or drones.

fig 2. the plotted error bars appear to span the range of the three annual estimates (the lines pass through their end points) this cannot be the standard deviation. More the stdev is not a very meaningful parameter for $n=3$. Better show this figure as a bar graph plotting the bars for each of the 3 years

We adjust the graph and the caption to using the error bar to show the lower and upper boundary of the CO_2 flux for each month from 2007 to 2009.

fig 3 since you use FEC as reference throughout the paper I suggest you make that line thick

We have noted the line with triangles. We are afraid that the F_{EC} line will be confused with the F_{FA} line if we make the F_{EC} line thicker.

Fig4 why show the lines for NIWOT ridge no variability for CO_2 and CH_4 and so little for N_2O

The CO_2 and CH_4 mixing ratio for NIWOT was monthly average while the N_2O mixing ratio was hourly average.

Reviewer 2

1. Does the paper address relevant scientific questions within the scope of ACP?

Yes.

2. Does the paper present novel concepts, ideas, tools, or data?

Regional flux estimate has been a persistent knowledge gap. The novel idea of this research is to conduct a comprehensive uncertainty analysis in multiple methods (inverse modeling, PBL equilibrium approach, tower-flux upscaling....) for estimating regional fluxes of multiple greenhouse gases (CO_2 , CH_4 , N_2O). Such an analysis is valuable and useful for studies with intends to upscale the local tower-flux measurements to a regional scale.

3. Are substantial conclusions reached?

Yes.

4. Are the scientific methods and assumptions valid and clearly outlined?

I agree with review #1's comments. I think that the basics of each method are clearly outlined. However, some descriptions and reasoning on assumptions need more efforts because the whole point of this research is to study uncertainties in different methods for regional flux estimations. Particularly, very large uncertainty is associated with the equilibrium approach. To improve the manuscript, I have a few suggestions for authors to consider.

(1) One more uncertainty sources associated with EQ approach might be in using the concentrations measured at Niwot Ridge (NWR, $40^{\circ}3'11''\text{N}$) as the proxy data of free-tropospheric CO_2 data ($c+$) at the KCMP tower site ($44^{\circ}41'19''\text{N}$). Although both sites are in the Ferrel cell with prevailing west winds aloft, CO_2 concentration has a prominent increase towards to high latitudes in northern hemisphere (Denning et al.,

1996). I suggest adding discussion on the above uncertainty source for EQ approach in the discussion section.

Denning, A.S., Fung, I.Y., Randall, D.A., 1995. Latitudinal gradient of atmospheric CO₂ due to seasonal exchange with land biota. *Nature* 376, 240–243.

We agree with the reviewer that using the CO₂ concentration measured at NWR site as the proxy of c+ at the KCMP site leads to uncertainties in the EQ method. We have added a paragraph to discuss this uncertainty (Page 3247 L21-P3248 L2) and refer to the work of Denning et al. 1995.

(2) I also suggest one more option that the Marine Boundary Layer CO₂ (http://www.esrl.noaa.gov/gmd/ccgg/globalview/co2/co2_description.html) measured at the same latitude as the KCMP tower site is located can be used as background-free-tropospheric CO₂, i.e. c+ in equation (2). The real values of free-tropospheric CO₂ (c+) can be provided by aircraft measurements as shown in Figure 2 in Yi et al. (2004).

Yi, C., K. J. Davis, P. S. Bakwin, A.S. Denning, N. Zhang, A. Desai, J. C. Lin, and C. Gerbig, The observed covariance between ecosystem carbon exchange and atmospheric boundary layer dynamics at a site in northern Wisconsin, *Journal of Geophysical Research*, 109, D08302, doi:10.1029/2003JD004164, 2004.

GLOBALVIEW-CO₂: Cooperative Atmospheric Data Integration Project – Carbon Dioxide. CD-ROM, NOAA ESRL, Boulder, Colorado, also available on Internet via anonymous FTP to <ftp.cmdl.noaa.gov>, last access: August 2011, Path: [ccg/co2/GLOBALVIEW](ftp://ftp.cmdl.noaa.gov/ccg/co2/GLOBALVIEW), 2006. If authors can find aircraft CO₂ data available to use, it would be incredibly helpful.

We thank the reviewer for this suggestion on using aircraft measurements or Marine Boundary Layer (MBL) CO₂ as the free tropospheric CO₂ (c+) at the KCMP site. We first compared the CO₂ mixing ratio at the NWR site with the Marine Boundary Layer CO₂ at the latitude of 44°N and aircraft measurements at three aircraft measurement sites close to the KCMP tower (Figure S3); then we use those mixing ratios as the free tropospheric CO₂ (c+) at the KCMP site to calculate the CO₂ flux with the EQ method (Figure S4).

We found that the CO₂ mixing ratio at the NWR site was not significantly different from the aircraft measurements at the three sites near KCMP, but was higher than the CO₂ mixing ratio in the MBL from July to September. Therefore, the annual CO₂ budget estimates using the CO₂ mixing ratio at NWR site are always in the middle of the uncertainty range determined by different data sources for c+. Therefore, we use the CO₂ mixing ratio at NWR site as c+ for EQ method for comparison with other top-down methods and report the details about the uncertainty analysis in section S4 of the Supplementary Materials

(3) Authors have estimated rW by several approaches, comparisons are valuable. I suggest authors using a figure or a table to summarize the comparison across the approaches.

We have added a figure of ρW to the supplementary materials.

5. Are the results sufficient to support the interpretations and conclusions?

Yes.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

See suggestions in 4th question.

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

Generally yes, but a few references below are necessary:

Denning, A.S., Fung, I.Y., Randall, D.A., 1995. Latitudinal gradient of atmospheric CO₂ due to seasonal exchange with land biota. *Nature* 376, 240–243.

Yi, C., K. J. Davis, P. S. Bakwin, A.S. Denning, N. Zhang, A. Desai, J. C. Lin, and C. Gerbig, The observed covariance between ecosystem carbon exchange and atmospheric boundary layer dynamics at a site in northern Wisconsin, *Journal of Geophysical Research*, 109, D08302, doi:10.1029/2003JD004164, 2004.

GLOBALVIEW-CO₂: Cooperative Atmospheric Data Integration Project – Carbon Dioxide. CD-ROM, NOAA ESRL, Boulder, Colorado, also available on Internet via anonymous FTP to ftp.cmdl.noaa.gov, last access: August 2011, Path: ccg/co2/GLOBALVIEW, 2006. If authors can find aircraft CO₂ data available to use, it would be incredibly helpful.

We have added Denning et al. and Yi et al. to the manuscript; we used the following reference instead of GLOBALVIEW-CO₂ as requested by the Cooperative Global Atmospheric Data Integration Project.

Cooperative Global Atmospheric Data Integration Project. 2014. Multi- laboratory compilation of atmospheric carbon dioxide data for the period 2000-2013 (obspack_co2_1_CARBONTRACKER_CT2013_2014-05-08). Compiled by NOAA Global Monitoring Division: Boulder, Colorado, U.S.A. Data product accessed at <http://www.esrl.noaa.gov/gmd/ccgg/obspack/>.

8. Does the title clearly reflect the contents of the paper?

It would be better if the title is changed into something like “Uncertainty analysis in multiple planetary boundary layer methods for estimating regional fluxes of greenhouse gases”.

9. Does the abstract provide a concise and complete summary?

Yes.

10. Is the overall presentation well structured and clear?

Yes.

11. Is the language fluent and precise?

Yes

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

Yes.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

See my suggestions in question 4.

14. Are the number and quality of references appropriate?

See my suggestion in question 7.

15. Is the amount and quality of supplementary material appropriate?

I agree with reviewer #1’s suggestions.

Miscellaneous

Page 3249, line 3, remove “other”.

We made the correction.

“our tall tower” has been used many times throughout the paper. It would be better if “our tall tower” is replaced by “the KCMP tower”.

We made the replacement

Reference

- Davis, K. J., Bakwin, P. S., Yi, C. X., Berger, B. W., Zhao, C. L., Teclaw, R. M., and Isebrands, J. G.: The annual cycles of CO₂ and H₂O exchange over a northern mixed forest as observed from a very tall tower, *Global Change Biol*, 9, 1278-1293, doi:10.1046/j.1365-2486.2003.00672.x, 2003.
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- Goulden, M. L., Munger, J. W., Fan, S. M., Daube, B. C., and Wofsy, S. C.: Measurements of carbon sequestration by long-term eddy covariance: Methods and a critical evaluation of accuracy, *Global Change Biol*, 2, 169-182, doi:10.1111/j.1365-2486.1996.tb00070.x, 1996.
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- Zhang, X., Lee, X., Griffis, T., Andrews, A., Baker, J., Erickson, M., Hu, N., and Xiao, W.: Quantifying nitrous oxide fluxes on multiple spatial scales in the Upper Midwest, USA, *Int J Biometeorol*, 1-15, doi: 10.1007/s00484-014-0842-4, 2014.