Review: Ryoo, AMTD, 2018

General Comments:

This manuscript includes emission estimates of CO2 and CH4 from the Sacramento, California, area from aircraft measurements on three different days. It presents some important and interesting investigations on the sensitivity of flux estimation toward the mass balance method used, the treatment of wind measurements, the choice of background, the inclusion of entrainment at the top of the boundary layer, and different interpolation and extrapolation methods. Still, I think the manuscript needs improvement in the structure, explanation of the methods used as well as the presentation of obtained data and results. The manuscript needs major revisions before it can be accepted for publication in AMT.

The structure of the manuscript could be improved with respect to the different sensitivity studies. I recommend choosing one "best-conduct" approach, explaining and using it for the flights first, and then doing the sensitivity studies and relating their results to this "best-conduct" approach to see each choices influence on the flux calculation individually. Thus there would be one section each on the mass balance method used (Gauss vs. downwind curtain), the treatment of wind measurements (raw winds vs. mass-balanced winds), the choice of background (min vs. avg), the inclusion of entrainment at the top of the boundary layer, and different interpolation (kriging, vertical interpolation, …) and extrapolation methods (kriging, constant, exponential weighting function, Gaussian fit).

The presentation of measured data differs for the three flights and the two compounds CO2 and CH4. Please choose the same set of figures for each flight, making sure all important data used in the flux calculation (like wind speed) is shown for all flights.

The results of using different treatment of input data are the different flux estimates. These numbers are often only named in the text. I think a tabular representation of results for each sensitivity study (or two combined studies, like in Table 1) would increase the readability of the manuscript. I really liked Table 1 and its discussion.

I am not sure if all of your flights are well suited for flux estimation. Generally you would look for a well-mixed boundary layer in order to decrease the uncertainty of flux below the lowest flight height. On November 17, 2015, the winds are quite different from one flight level to another and very weak in the lowest level. Furthermore, detecting the highest CO2 concentration in an upwind part of the flight path shows that this day is not suitable for flux estimation. Also the local flux estimates on July 29, 2015, show low wind speed with changing direction.

Your calculation of mass-balanced wind is interesting. It seems as if the mass-balanced wind in Fig. 5 is constant with height. Shouldn't it vary with height because you use the average wind for each level? What is your surface condition? Why is kriging used? Please explain in more detail how the "mass-balanced" wind field is generated? The difference in the flux estimate does not surprise me (l. 359). You have very good wind measurements, and you definitely see a change in wind speed with height in Fig. 5b. Low wind speed with high concentrations and high wind speed with low

concentration might result in the same flux. Thus, Fig. 5c looks quite logical to me. If you remove all your information on the vertical wind speed change, as you did in Fig. 5e, then of course the flux only represents the concentration measurement. Why do you neglect your information on the wind situation? Did you ever calculate the mass-imbalance of the raw winds? Is it significant?

Your use and understanding of the divergence method seems flawed. Please review the Gauss theorem and describe it correctly in l. 257. I do not believe that a background is necessary for this method. You simply calculate all fluxes through the surface of the cylinder (outflow – inflow) and thus receive the change of mass within the cylinder. Please check the publication of Conley et al. (2017), which nicely explains the application of Gauss theorem on aircraft mass balance flights. For your second method (the curtain downwind of the sources) you definitely need a background and here the influence of choice of the background value on the flux estimate is quite interesting.

Your calculation of the flux through the top of the cylinder is useful, but I do not understand how you determine the surface flux. First, with the Gauss theorem you need to assume that all the mass change inside the cylinder (e.g. what leaves through the surface) comes from the sources on the ground, thus what you determine is the surface flux. Second, on the ground the vertical wind speed is zero. How can the surface flux calculated with your method then be different from zero?

Finally, could you calculate an overall uncertainty of your flux estimates from the different calculation methods and treatment of input parameters?

Please improve the consistency of your terminology. For example you defined the two ways of treating your wind measurements as “raw wind” and “mass-balanced wind”. In the following manuscript you then repeatedly use mean wind, measured wind, averaged wind, area-mean wind…

I also recommend grammar checking by an English native speaker and thorough checking of references to Figures and Sections. Furthermore, please increase the size of the axis labels and color bars on most of your plots. They are not readable.

Specific Comments and Technical Corrections:

l. 43: Which meteorological factors?

l. 46: “emissions fluxes” should become “emission fluxes”. Please check the whole manuscript.

ll. 48-49: This should be reformulated due to the low winds on Nov. 17, 2015, and the high concentrations in the upwind part of the flight pattern.

ll. 49-50: The wind variability and seasonality has not been investigated in this study. Please reformulate.

l. 51: Where do you show the influence of the distance to the emission sources?

l. 58: What is your “modeling strategy”? Do you do any modeling?

l. 61: Why don’t you mention your investigation on background and wind treatment in the Abstract?

l. 65: Introduce abbreviations once and then use during remainder of manuscript (e.g. GHG).

l. 66: Is air-quality important in this study?

l. 69: Check your use of “give rise to”…

l. 72: What is the “role of human behavior in altering the emissions”? Do we need to know it for national emission estimates?

l. 76: What are indirect emissions?

l. 78: Please give an example of a bottom-up inventory using proxy data to achieve fine spatial resolution.

ll. 83-86: What about flux estimates of European cities?

l. 94: What do you mean with “those efforts reach general agreement on emission inventories across the cities”?

l. 106: Supplementary Material

l. 108: Do you really mean “uniform vertical mixing”, or maybe “uniform distribution of trace gases”?

l. 110: Does this sentence (“These studies…”) apply to the first or second category, or maybe both??

l. 112: Does the “single-screen multi-transect method” really depend on constant wind speed? You could also use average wind at each transect or even raw wind at each measurement point.

l. 119: The cylinder pattern should be “around” a source and not only “near”.

l. 126: Why are the additional point sources considered sources of uncertainty?

l. 131: I think we all got the concept of three-dimensional space (delete the parentheses).

l. 134: The PBLH is not hard to measure. It is relatively easily determined from a vertical profile of temperature and humidity as you have done in this study. You even stated that the different approaches you used led to similar results. So what is difficult with respect to the PBLH? It is certainly difficult to model correctly, like you stated that substantial differences exist between models and reanalysis data. Also consider the large diurnal variability of PBLH.

l. 135: Please don’t use “observed” in connection with models. This might confuse.

l. 139: Do you really think the execution of flights is a goal of the study? Or is it merely necessary for the other goals?

l. 145: Which “value”?

l. 153: Describe the three flights here and mention figure 1. It is not mentioned at all in the text.

l. 154: How many whole-air standards do you use for calibration?

l. 159: Take off time is not sufficient. Please give the total flight times in UTC. Using Pacific Standard Time and Pacific Daylight Time here needs more explanation on why you give take-off times in different ways.

Sect. 2.2: Review order of sections: I would first discuss the interpolation method and then the extrapolation.

l. 163: Reformulate: “Because the lowest flight level was typically between 250 m and 380 m above the surface …”

l. 165: Sentence needs restructuring: The “unmeasured values” lead to uncertainty whether or not a “well-mixed layer assumption” is made. Split sentence!

l.168: Refer to Figure 2 here. There is no reference to it in the text.

l. 169: What do you mean by “elevated” plume? Is it lifted of the ground or are there large enhancements of the concentration?

l. 170: How exactly do you derive the background level? What is the “lowest flight measurement”? Has it got anything to do with the “lowest flight level” which you use in the formula? Is the background only determined from the lowest flight level? Why are you talking about background at this point? It is a section on extrapolation to the ground. Do you also extrapolate the background values? What do X and t stand for?

l. 172: Do you mean that the details of the method are described in Gordon et al. (2015)?

l. 173: How is the Gaussian distribution of the plume dispersion calculated?

Sect. 2.3: Consider renaming the section to “Measurement interpolation”.

ll.176-186: Should this be a separate section called: Projection of data to cylinder surface?

l. 182: What is Y?

l. 190: Refer to Fig. S4 as you show these differences there. Consider overplotting the measurements on the kriged and interpolated fields for better assessment of your result that kriging better captures individual plume features. What altitude range do the elliptical cylinder plots cover? Ground to PBLH? Please state in the figure caption.

ll. 214-227: Consider a separate section on uncertainties.

l. 216: Not only downwind interpolated values induce uncertainty. Upwind values as well.

l. 226: Add “observations” behind “direction”.

l. 229: Remove “to the choice of background value and” because this is not the topic of this section. You do not investigate the wind characteristics but the treatment of wind measurements.

l. 230: Remove “In one”

l. 231: What are “measured points”? How did you measure them?

l. 233: Stick to one tense (averaged, equaled).

l. 233: “By assuming non-divergence, mass can be balanced.” This is correct, but is this really what you need here?

l. 242: Do you assume PBLH to be constant during your flights? At what time during the flight did you measure the profile?

l. 243: Is the boundary layer “growing” during your flights? How do you know?

l. 248: How is C(t,z) determined? How can one point surround the top of the cylinder? How is the background defined here?

l. 252: Is the entrainment calculated from the kriged data?

l. 263: Flux is defined through a surface. Thus it cannot be “inside” the cylinder.

ll. 265-280: See my comment in the General Comments section on the use of a background value with the Gaussian divergence theorem. If you consider inflow and outflow, you do not need a background. In your formula the result should be invariant to the value of background mixing ratio chosen if you consider positive contributions as outflow and negative contributions as inflow.

l. 291: Use present tense.

l. 295: Remove “concentration”.

l. 299: How is the kriged estimate less arbitrary in an area far away from measured values? What assumptions is it based on? Is the state of the PBL (stable/unstable) taken into account?

l. 300: You don not mention the Gaussian fit method depicted in Figure 2 at all.

Sect. 3.1: What is the influence of the different choice of interpolation and extrapolation on the flux estimate? Here a table similar to Table one would be great.

l. 304: Remove “gap of the”.

ll. 314-320: Please mark all the locations mentioned in the text on a map so the reader can confirm your statement.

l. 325: Present tense.

l. 327: Please check “a farther”.

l. 330: Maybe use the last sentence of this paragraph as its first. Good introduction.

l. 350: The PBLH you determine from the vertical profile might have an uncertainty of <1%, but is this value representative for the whole measurement area with this accuracy? What about changes over time and with the location? How does a less defined PBLH influence the uncertainty?

Fig. S6: Looking at your method of estimating PBLH there seems to be a possible error of more than 1 % as well. In Fig. S6d it becomes clear, that you use the 50 m averaged values for checking the gradients. Then you place it at the top of the layer with highest gradient. Here it is visible, that this point is easily 40 m above the layer where a 20 m averaged profile would see the gradient. Thus your uncertainty is around 50 m, which would be almost 10 % for a PBLH of 600 m.

l. 355 ff: See my comment on the treatment of “mass-balanced wind” in the General Comments section.

Sect. 3.3: Please already refer to your Table 1 when naming the results.

l.267: Why is there an “actual” location of the rice field? Pleas show locations on a map rather than just giving coordinates. This is very hard to visualize for a reader.

l. 370: “the local emissions are attributed to these high flux estimates”. Did you mean: “The high flux estimates are attributed to the local emissions”?

l. 374: Formulation: “mean wind vector at the dominant wind direction (positive and one direction) and speed”. How is this calculated?

l. 381: There is no Table 2.

l. 387: Raw wind is displayed in the bottom two lines.

ll. 390-391: This sentence is incomplete and not logical.

l. 394: Table 1 shows a range of 3.68 - 26.58 Mt CO2 yr^{-1} for the whole city.

l. 396 ff: Here you investigate the difference between using the complete ellipse and only the downwind part. This should be a separate section and the results should be presented in another table.

l. 399: Change “From this study,…” to “According to these calculations…”

l. 401: Table 1 gives a range of 13-92 Mt CO2 yr^{-1} for Nov. 18, 2015.

l. 402: Please indicate “Region-3” on a map.

l. 405: Is vi) the same as i)?

l. 405: Which of these does “This” refer to?

ll. 415-422: “Note … Table 2).” All this is repetition to before and not about the topic of this section which is “vertical mass transfer”.

l. 428: Remove “First,”

l. 431: Specify: “different flux calculation methods”

l. 453: There is a contradiction here “the final flux estimates become similar”, because the beginning of the sentence states that the background value is a major source of uncertainty.

l. 459: Insert “that” after “suggesting”.

ll. 460-468: This section is a general overview of the flight results and should be plasec earlier in the Conclusions.

l. 463: An overview of wind conditions should also be placed in the Results section.

l. 464: This result (isolated high concentrations of CO2) has not been shown in the Results section either.

l.470: Why did you expect sources to be concentrated on the downwind side?

l. 471: “Furthermore” does not fit here.

l. 471: Wind variability definitely influences the flux estimates, not only during different times of the year. So this seems logical. It would be much more interesting how large the uncertainty due to this is assumed to be.

l. 475: The size of the ellipse is another factor that appears here for the first time in the manuscript. There is no data given on how large your ellipses were and what the influence is in the Results section.

l. 480 and 481: Remove two of the three “further”.

l. 482: Do you really want to assess: “seasonality of sensitivity of emission estimates”? Just start with seasonality of emissions first.

l. 484: Where do you show the sensitivity of emission estimate uncertainty to temperature and potential temperature?

ll. 490-491: This sentence needs some revision and focus.

Figures

Fig. 1: There is no shading visible in Fig. 1c.

Fig. 2: What is the “altitude of lowest flight data”? Please indicate the location of these measurements on a map, giving coordinates is not very helpful.

Fig. 3d: Is the ellipse shown from the ground to the highest flight level or which altitude range?

Fig. 4: Please provide headings with the date of the flight for the left and the right column.

Fig. 5: Why is the mean wind kriged? This has not been mentioned in the text.

Fig. 7: Why is there this large space between the two sets of bars? What is “area-mean”?

Table 1: Tables normally have their description above not below them.

Supplementary Material:

There appear to be bits and pieces of text strewn throughout the Supplement. Please give them a heading and a number so it becomes clear where they belong and you then may also refer to them from the main manuscript.

Fig. S1: Figure b color bar label is missing.

l. 7: I am not sure if you can say that emissions are “accumulated” downwind. They are transported downwind, but accumulation would mean that there is very slow wind only.

ll. 11-12: This is not true. With a curtain flight it is also possible to detect emissions from more than one point source within the city, throughout the city and downwind. It gets problematic if there are sources further upwind of the city that get mixed with the city plume and cannot be separated from it.

ll. 15: You mention three types of flight patterns in the main manuscript but only show two of them here.

Fig S2: Reformulate “throughout the altitude”. Color bar labels are missing.

l. 25: “accumulated” s.a.

l. 28: Why is air at lower wind speeds less dispersive?

l. 28 ff: Reformulate sentence “Both flights …”

l. 30: Who uses continental scale wind for flux estimates?

Fig. S3: This figure is not mentioned in the main manuscript. Please add flight dates to the left of the plots.

l. 36: Consider “falling”. For methane the dashed line is blue. Remove “observation”.

Fig. S4: Why are there “boxes” or vertical cuts visible in (d)? Does this have to do with gridding? What is the grid size? Could you plot the measurements on top of the interpolated fields? This way it is easier to assess your statement “kriging reflects the individual plume characteristics better”. Could you show the extrapolated fields to the ground as well? Which step is performed first: interpolation or extrapolation? Is this described in the text?

Also: Use the same color bar range for all plots.

l. 57: Don’t (b) and (d) also show only the subset of the ellipse? Could you change direction of these plots? Then this arrow would not be necessary.

l. 82: Remove the sentence: “The CH4 enhancement was localized near the landfill.” This is obvious.

l. 83: Also remove “…, and we … case.” This is also obvious.

Fig. S7: This figure is not mentioned in the main manuscript.

Conley, S., Faloona, I., Mehrotra, S., Suard, M., Lenschow, D. H., Sweeney, C., Herndon, S., Schwietzke, S., Pétron, G., Pifer, J., Kort, E. A., and Schnell, R.: Application of Gauss's theorem to quantify localized surface emissions from airborne measurements of wind and trace gases, Atmos. Meas. Tech., 10, 3345-3358, 10.5194/amt-10-3345-2017, 2017.