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Interactive comment on “Determining air pollutant emission rates based on mass balance using airborne measurement data over the Alberta oil sands operations” by M. Gordon et al.

Anonymous Referee #3

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Review of "Determining air pollutant emission rates based on mass balance using airborne measurement data over the Alberta oil sands operations" by Gordon et al., submitted to AMT.

This paper describes and outlines a specific methodology for calculating emissions of pollutants from point sources (for example, specific facilities) using aircraft concentration and wind measurements. To my knowledge, this is the first attempt at standardizing this method, which is often used but with slight differences between different research groups. It is a suitable subject matter for an AMT publication, as it focuses on the method developed for this estimation.

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Overall, this is a well written and clear paper that outlines a method for calculating emissions rates using aircraft measurements of pollutants and is appropriate for publication in AMT after some specific comments are addressed. My one concern is the treatment of uncertainties which is not as rigorous as it could be. The uncertainty of each component is generally estimated based on several cases (trying different extrapolation methods for example) and always stated as approximate (using approximately equals signs). The authors should be more definitive, even though they do state that the uncertainties are being "estimated" and not really calculated. I felt that a paper specifically describing a method should go the extra step and define uncertainties in a more systematic way, and I would like to see an attempt at this. Or, if the method is retained, some overall description: "for the TERRA method, we estimate uncertainties for each term by calculating each term using different assumptions and using the largest difference between two different results from the different methods" or something to that effect, to show at least methodological consistency.

Specific comments:

Line 25: In addition to Wratt et al., others have used this budgeting approach, for example see Gatti et al., Nature, 2014 for using aircraft profiles upwind and downwind to estimate fluxes in the Amazon.

Introduction does a nice job of outlining the different similar but slightly different methods that have been used in the past - this is a nice overview for the reader. See comment at the end of this review on perhaps differentiating between estimates of point source emissions vs. regional emissions over a larger area and measurements farther downwind.

p4774 L1: define IMU - is altitude not measured via GPS, but rather only converted from pressure altitude? What is the accuracy of this, usually pressure altitude conversion assumes some ideal atmosphere, etc.? How do you have lat/lon without GPS?

p4774 L1-5 I would also be interested in the frequency of the measurements on the

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aircraft. p4774 L15 - I cannot find the Picarro G2204 on their website - what is the frequency of measurements and is this a flight-ready analyzer?

By time delay, are you referring to how long it takes for air to go through the inlet line on the aircraft, or actually internal to the analyzer? This seems quite long - what is your flow rate on the Picarro unit?

Regarding the precision, I understand why you want to define it based on five different calibrations, but it seems like the short-term precision might be more important given your application. You are differencing upwind and downwind, so your uncertainty in that difference is only tied to the drift of the analyzer within a flight, not across the whole project. I would hypothesize that will be much smaller than 1.3%, which is very high. I need to read on as to how you fold this into an uncertainty on the emission flux, because 1.3% is not actually the precision of the methane measurement, but of the slope. How different are actual mole fractions you measure for each time you calibrate? At 1900 ppb, 1.3% is nearly 25ppb error - I doubt your calibration standards were measuring 25 ppb different from one calibration to another.

How does this uncertainty propagate into your final uncertainty? It is never addressed how this or other measurement uncertainty (in winds for example) affects the final uncertainty.

p4774 Line 10 - since this is a methods paper, how do you run those 5 calibrations for both gases? What range of concentrations (mole fractions) are used as your references?

p4779: what is the frequency of your wind measurement?

p4779, prior to interpolation using any of the 3 methods, what is the native resolution of the measurements along S? Do you average your wind measurements prior to interpolating? My understanding of wind measurements is that there is a lot of "noise", i.e. short term variability that would need to be averaged before interpolating (real

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atmospheric variability due to turbulence).

p4781 line 21 and 23: deposition velocity is sometimes V_d and sometimes VD , keep consistent.

It seems to me that separating the turbulent and advective fluxes means that you are averaging the concentrations you have measured (and the winds) so that you are not resolving turbulent eddies, and only capturing mean advection. Otherwise you are double counting. Am I understanding that?

p4781 L28 what is the E_a, V term?

p4785. It would clarify the different methods for extrapolating the concentration measurements down to ground level if you had a figure, or refer to Figure 6 here.

p4786, 20-25. Can you clarify how you compare the three interpolation methods to the original synthetic data to evaluate them? (rms error and correlation coefficient are defined how exactly?). The caption in Table 4 explains a little bit more, but I'm not sure if it says - you are comparing actual emissions calculations (which take the wind and the concentration into account both?). What is the interpolated average? (average emission, or average concentration?), A little more information would be good here, perhaps in the text.

p4786 If you can add a figure, I would like to see the concentration screens after the interpolation based on the flight data locations for all three interpolations and cases (i.e. the corresponding figure to Figure 3 but after you've re-sampled and interpolated the data) to get a qualitative feel for how this works. Since you ran so many cases, even just showing it for one case (perhaps Kriging which you determined was the best fit) would be nice.

p4787 L26: the figure caption and the earlier text indicate that you are referring to the zero-to-constant extrapolation, not the linear extrapolation here. Otherwise, this is confusing.

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p4787 section 3.3 - Which interpolation method is used in Figure 4? is this the Kriging method, and is that what is also plotted in Figure 5 and 6, because it was the best in your evaluation of the three methods? Please note this in the text and captions.

p4793 Line 27, this is only true if n are *independent* measurements of the same quantity. At 1 Hz, these are not independent because you have correlation length scales in the atmosphere much longer than your measurement frequency. I am guessing that the mean measurement of the upwind side of the top box is much different than of the downwind. I would argue that N might only be equal to 4 (for the four sides of the box top). Otherwise, the time traces of the measurements will tell you the correlation length/time scale, and then one can calculate N. This is what you would do if you had a circle for example. The 95% confidence interval in this mean is larger than you state.

p4794 L9: What highest uncertainties? you mean for each delta? In the text, values are given for each delta but not the highest or any range, so it's not clear what is meant by "highest" here.

p4795 section 4.2 - how do these wind shifts affect your uncertainty estimate? (should uncertainties be higher on days when the plume may shift?).

p4797 general comment: One thing should be mentioned either in the introduction or here, which is that other methods (e.g. single transect) that assume perfect mixing in the vertical direction (and thus, the extrapolation to the surface is constant), make that assumption only far enough downwind of the source so that the plume has had time and distance to mix vertically within the PBL. The TERRA method could also certainly be used farther away from a source or for calculating emissions from a larger region, but it is most useful when emissions are being calculated from a single source whose location is known and surrounded by the flight path. Some of the references in Table 1 use this type of method for significantly greater source regions (Turnbull, Peischl, Karion, Mays and Cambaliza).

Another general comment: What kind of uncertainties do you claim for the TERRA

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method in general? Is it dependent on the specific flight - i.e. would the same uncertainty analysis you have conducted (multiple methods comparisons, generally) need to be conducted every time TERRA is used to calculate emissions, or can it be generalized somehow?

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 4769, 2015.

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