

This paper describes a top-down emission rate retrieval algorithm that the authors employ on two sample flights around an oil sands processing facility in Alberta Canada. The estimation methods, in principle, appear to be sound but the treatment of uncertainties elides many sources of error, and erroneously adds them arithmetically. The 20% overall uncertainty quoted in the abstract is misleading and inaccurate. Because the components of the emission rates are added together, the end result can be a small differential number derived from several much larger ones (in the case of CH₄ the leading terms are 1-2 orders of magnitude larger than the overall result.) Therefore the relative error magnitudes for each term cannot be simply added together to get an overall relative error.

The results from the SO₂ analysis seem to be reasonable (the aforementioned error in equation 11 notwithstanding) due to the fact that the plumes are lofted and clearly visible by the aircraft on the downwind sides of the facility. On the other hand, the methane results are very tenuous, and should receive an independent and revamped error analysis.

There are several serious concerns with the methane emission estimate. The first and foremost is that the majority of the methane is assumed to be passing through the lowest ~150 m where there are no measurements. As the authors point out, their method is extremely sensitive to the method of extrapolation, but then they choose a blended approach for reasons that are not very clear. Fluxes of methane from boreal forests are not known to be extremely large (the authors present no reference for this, but Shoemaker et al. 2014 report ~ 1 nmol/m²/s, which gives rise to increasing methane concentration in the boundary layer of about 0.1 ppb/hr.) This is nowhere near the ~100 ppb enhancements derived from the exponential extrapolations used in this work. In any event, Figure 5 shows results that do not jibe with the analysis as presented. On the August 20 flight the major plumes are seen (below the lowest flight leg) on the East and NE corner of the pattern, which is the downwind side on that day. Similarly, on September 2 the methane plume appears (mostly below the lowest measurement height again) on the south side of the facility, which again is the downwind side. But Table 6 indicates that on both flights there is a surplus of inflowing methane over what is outflowing ($E_{C,H,in} > E_{C,H,out}$).

This then leaves the possibility of horizontal convergence (lower concentrations but higher wind speeds on the upwind side), and indeed the manuscript mentions that there is very strong convergence in the test volume giving rise to mean vertical velocities of ~ 0.10 m/s. The authors make mention of this being potentially possible in complex terrain, but the terrain slopes down to the east (with the direction of the wind on the August flight), so this cannot be orographic uplift. And if this is simply because the downwind face of the volume is larger than the upwind area (because of lower terrain), there is no mention of taking this differing area into account in the flux budget. Unless there is a small 'urban heat island' effect from the facility, it is not clear why such a vigorous updraft would be ever-present at the site. Lenschow et al. [1999] outline the measurement requirements to measure horizontal flow divergence, and thus mean vertical velocities, by aircraft. Following their analysis of the uncertainty in the vertical velocity based on uncertainties in the horizontal wind measurements (specifications from the Aventech AIMMS-30 probe, a generation beyond the one used in this study, indicate an accuracy of all wind components to be 0.5 m/s) leads to an approximate error in the vertical velocity based on divergence

measurements of ~ 0.10 m/s. This means that the error in the third term of Table 6 is of order 100%! Further error in this term appears because the box-top concentration is only sampled at the perimeter of the flight pattern, but the enhancements from the local sources are presumably occurring in the middle of the facility. If the flow is truly that strongly convergent then the updraft should be lofting concentrations representative of the middle of the facility, not those along the perimeter.

There is no mention of the boundary layer height in the manuscript, but given the high latitude and ample latent heat fluxes, I would assume that it's not much above 1000 m (asl, ~ 700 m agl). Typically the surface layer (where the flux-gradient similarity relationship that the authors use for the wind extrapolation to the surface is valid) is about one-tenth the boundary layer height, or valid up to about ~ 70 m above the surface. Extrapolation of these relationships up to 150 m is dubious, and no fit parameters were given for the relationships observed by the RASS to assess the validity of the extrapolations.

Finally, as explained earlier, because the overall result is a small difference between three large leading terms (Table 6), it is not the case that errors in each of the terms can be treated as relative (%) and then added in quadrature at the end to get a relative error in the final result. In short, Equation 11 is incorrect. The errors need to be added together in their native units (t/hr), and when compared with the emission estimates they will likely be several times as large as the estimates themselves.

References:

D.H. Lenschow, P.B. Krummel, And S.T. Siems, Measuring Entrainment, Divergence, and Vorticity on the Mesoscale from Aircraft, *J. of Atmos. Ocean. Tech.*, v16, 1384–1400, 1999.

Shoemaker, J. K., T. F. Keenan, D. Y. Hollinger, and A. D. Richardson, Forest ecosystem changes from annual methane source to sink depending on late summer water balance, *Geophys. Res. Lett.*, 41, 673–679, doi:10.1002/2013GL058691, 2014.