

## ***Interactive comment on “Quantifying fugitive gas emissions from an oil sands tailings pond with open-path FTIR measurements” by Yuan You et al.***

**Anonymous Referee #1**

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Review of “Quantifying fugitive gas emissions from an oil sands tailings pond with open-path FTIR measurements” by You et al., submitted for publication in Atmospheric Measurement Techniques (amt-2020-257). This manuscript presents results from a field study conducted in the oil sands region of Canada, where gas emissions of CH<sub>4</sub>, NH<sub>3</sub>, and alkanes were measured from a tailings pond using micrometeorological techniques.

### General Comments

The strength of this work is the scientific significance of the emission measurements that have been documented. The subject matter is of broad interest due to the large scale of industrial operations in the oil sands region of Canada, the use of large tailings ponds in these operations, and a paucity of information on gas emissions from these

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sites. However, the scientific quality of the work is mixed. Several methodological details need to be clarified. And while some aspects of the study are well-explored, others are not given sufficient attention. Finally, the overall presentation of the manuscript should be improved. Basic information that should be in the manuscript is missing and the reader is forced to look to a companion paper and to supplemental material for that information. It is difficult for the reader to understand this paper without a good deal of effort. My overall recommendation is that the flux calculation details need to be significantly clarified and documented within the manuscript. This work could then make a valuable contribution to our understanding of the environmental footprint of oil sands operation.

### Specific Comments

1) Questions related to FTIR measurements. I have two questions regarding the FTIR concentration retrievals and the related flux calculation.

a. The authors state (line 108-109) that “... temperature and pressure dependent reference files were used for fitting and retrieving mole fractions.” It is unclear how this was done. My concern is that with vertical temperature gradients at the measurement site (as implied by Fig. 4 in the companion paper by You et al.), different temperatures should be used for retrievals for the different FTIR path heights. If not done, is there potential for “false” flux signals? The authors should comment on this.

b. The second question regards the concentration measurement used to calculate flux in Eq. 1. Following the classic WPL flux corrections for flux-gradient formulae (Webb, et al. 1980. Quart. J. R. Meteorol. Soc. 106, 85–100), should the concentrations be the mole fraction with respect to dry air (mixing ratio)? This should be clarified.

2) The exact procedure for calculating the FTIR fluxes is unclear. In one section the authors indicate the critical tracer diffusivity ( $K_c$ ) for the FTIR calculations was extrapolated from  $K_c$  measured from point concentrations (Eq. 2), and at another point they state that  $K_c$  is determined from the measured eddy diffusivity  $K_m$  (Line 314). Looking

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over this paper, the companion paper, and the supplemental material, I am uncertain as to what was done. The methodology needs to be better explained.

3) I am concerned about how tracer diffusivity ( $K_c$ ) is calculated in Eq (2). The calculation assumes a linear change in  $K_c$  with height, but the general view is that diffusivities (heat, momentum, moisture) are non-linear except in neutral stratification. This may have an impact on the calculated fluxes. Consider the standard definition of  $K_m$ :

$$K_m = k_v * u_* * z / PHI_m,$$

where  $PHI_m$  is the non-dimensional wind shear. A commonly used  $PHI_m$  relationship is given by Hogstrom (1996) for unstable conditions:

$$PHI_m = (1 - 19*z/L)^{-0.25}$$

If we redo the calculation outlined in the manuscript Eq. (2) to determine  $K_m$  for the FTIR gradient ( $z = 1, 12$  m) using the  $K_m$  calculated from the point measurements ( $z = 8, 32$  m), with the above expressions and assuming  $L = -20$  m, then  $K_{m\_FTIR}/K_m = 0.25$ . This is 25% lower than the 0.325 value the authors calculated assuming a linear relationship. This implies the fluxes calculated by the authors may overestimate the fluxes in unstable conditions. While this example is for the case of  $K_m$ , one would expect a similar non-linear relationship for  $K_c$ .

4) There should be a deeper discussion of the relationship between the different flux measurement techniques. I have four questions/concerns in this regard.

a. How much are the flux-gradient (FG) measurements constrained to match the eddy covariance (EC) measurements? It strikes me that calculating the tracer diffusivity ( $K_c$ ) from the concentration gradient and the EC flux, and then using that  $K_c$  in the FG calculation will act to force the FG and EC fluxes to be equivalent. Yet in several places (lines 180-210) the authors discuss how the FG and EC fluxes are different. How do we reconcile those two things?

b. As mentioned before, it is unclear how the FTIR fluxes were calculated in this study.

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Based on the companion paper, I suspect the turbulent Schmidt number ( $Sc$ ) was used to estimate  $K_c$  from the eddy diffusivity  $K_m$  (in the companion paper the authors give an interesting evaluation of  $Sc$  and conclude that over a broad stability range  $Sc = 0.923$ ). A conclusion of this manuscript is that the FG fluxes from the pond are 40% lower than the EC fluxes (Line 192). This difference between FG and EC could be erased with a smaller  $Sc$  (for which there is good evidence in the literature). In looking at the  $CH_4$  fluxes from the three different measurement techniques (Table 2), could the average from all three be statistically identical if  $Sc = 0.6$  is used in the FG calculations?

c) The authors discuss the difference in fluxes from the FG and the EC techniques in terms of their different measurement footprints. I am not convinced the difference is large. The authors state that the mole fraction footprint (FG) is larger than the flux (EC) footprint (Line 206). It is true that a concentration footprint is much larger than the corresponding flux footprint, but an FG footprint is given by a difference in two concentration footprints (two heights), which is not so different from the flux footprint (if the two FG heights are not too far apart). In other words, a distant source contributes almost exactly the same concentration at both the top and the bottom measurement heights, so the footprint difference for distant sources is zero. I think the difference between the FG and EC measurements may have another explanation.

d) The authors compare the fluxes calculated using their FG formula with those using the slant-path FG formula from Flesch et al. (2016), and found the slant-path calculations give fluxes that are 27 to 56% lower. This result is difficult to understand, given that both calculations start with the same underlying FG calculation approach and use the same concentration gradients. The difference could be due to the assumed  $Sc$ , or how the path integration of the FTIR concentration and  $K_c$  is handled. There is a difference in  $Sc$  between the two calculations. But because the  $Sc$  used in the slant-path formula (0.64) is smaller than used in this study ( $\sim 0.923$ ), the slant path flux magnitudes should be higher, not lower. I suspect the difference is related to the diffusivity calculation discussed in comment 3 above. The authors should explain how these two

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different FG formula can lead to such different results. This is important for confidence in their FG methodology, as the slant-path formula is a more rigorous expression of the FG relationship for path-integrated measurements than their working formula.

#### Technical Corrections

Line 75: it is unclear what is meant by “turbulent fluxes” in this context. Gas fluxes?

Line 85: “Other experimental details of the project can be found in You et al. (2020)”. This is one of several places in the manuscript where critical pieces of information are missing and should be included: e.g., a figure showing the equipment locations with respect to the tailings pond. I noted that Lines 128, 150, and 160 refer the reader to critical details in You et al. (2020). I would like to see some of that other-source material moved into this manuscript.

Line 117. “For these trace gases at this site, the detection limits of this open path system were insufficient . . .”. This is very interesting.

Line 122. Does the flux calculation really fit the usual description of a modified bowen ratio (MBR) method? It’s difficult to know because of uncertainty as to how the fluxes are calculated. Usually an MBR measurement means the unknown flux of the gas of interest is related to the known flux of another tracer gas, plus the gradient of that tracer gas, and the gradient of the gas of interest . No theoretical flux gradient relationships are needed. In this study, it is likely that the tracer diffusivity  $K_c$  is estimated from a stability corrected  $K_m$  measured with a sonic anemometer, I would say the technique is better described as an aerodynamic flux-gradient approach.

Line 160: “. . . the warm pond surface . . . resulting in continuing transport of pollutants . . . without significant diurnal variation.” When the pond is warm relative to the air, does this mean there are no diurnal changes in atmospheric stability over the pond? I doubt it. This statement also implies that aerodynamic resistance is the limiting factor controlling pond emissions.

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Line 314: “In our modified Bowen ratio approach,  $K_c$  is derived from a measured and stability corrected  $K_m$  . . .”. This sentence highlights the confusing description of the methods used in this paper: I do not think  $K_m$  has been defined to this point in this manuscript; and Eq. (2) suggests that  $K_c$  for the FTIR is calculated from  $K_c$  taken from point measurements (and not from  $K_m$ ).

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