

We appreciate the careful consideration of our manuscript by this reviewer. We have carefully responded to all of the point-by-point comments and issues raised by the reviewer and have revised the manuscript accordingly. These revisions are described in detail below, with our responses given in blue.

Responses to Reviewer #2:

This manuscript describes measurements of low molecular weight organic acids (LMWOA) from the oil sands in northern Alberta, and uses a box model to evaluate the measurements. The estimates of emission ratios, and impacts of those emission ratios on concentrations downwind is a useful addition to the growing literature on organic acids in the atmosphere. I recommend publication following minor revisions.

We thank the reviewer for his/her positive comments. We agree that this paper will make a good addition to existing literature on this topic. We have addressed the specific issues raised by this reviewer below.

There are too many acronyms to keep straight! I suggest moving most of them into full words (e.g. OS -> oil sands), and reserving acronyms for extremely long word combinations ('LMWOA') or very commonly used acronyms (VOC, SOA).

We agree that there are many acronyms in this paper. We have taken the reviewer's suggestion and tried to use full words where possible. Specifically "OS" is now "oil sands", PAN is now "peroxyacetyl nitrate", FA is now "formic acid", AA is now "acetic acid".

The authors set up an excellent case for their study in the Introduction – a solid description of relevant literature, and lines of evidence regarding exactly why anthropogenic sources from oil production could be regionally and globally relevant. However, the last sentence of the introduction ("It is expected that the results of this study will be broadly applicable to the secondary formation of LMWOA from other anthropogenic activities with hydrocarbon emissions") does not follow this level of logic and reasoning. For example, other anthropogenic activities with hydrocarbon emissions can range from vehicle emissions to natural gas extraction to industrial solvent production to human-induced biomass burning activities, etc. The VOC precursor emissions from each of these sources can vary widely, and thus I expect the LMWOA emissions to vary as well. After explaining how unconventional the oil sands region is – and that the emissions from that region can be different from other areas – I don't see how these measurements will be broadly applicable to other regions. The authors have built up a strong case for their work, and broad applicability to other anthropogenic hydrocarbon sources is unnecessary to convince me that this work is important. I strongly recommend the authors delete the sentence.

We agree that "broadly applicable" is an overstatement. While we agree on this point, we note that it is the underestimate of LMWOA formation from the box model that is likely to be more broadly applicable even to urban areas, since the VOCs oxidation mechanisms used here are

not going to be significantly different for any other location (even though the amounts and types of LMWOAs formed will be different). Regardless, for clarity we have removed this sentence from the paper since as the reviewer states, it is not necessary.

Authors identify the deposition of organic acids downwind of the oil sands as a potential source of atmospheric acidity. My recollection of the acid deposition literature is that organic acids do not contribute to ecosystem acidity – formic and acetic acids are weak acids, in contrast to nitric and sulphuric acids, which are strong acids. Can the authors point to literature or back-of-the-envelope estimates to further back up this idea that deposition of organic acids could be relevant for ecosystems downwind of the study area? While I understand that this is not the focus of the paper, and a detailed estimation is beyond the scope of the paper, the suggestion that organic acids could influence ecosystem acidity is a large one, and I think that the authors would do well to provide a little more literature or evidence that this is a likely event.

We agree that strong acidity will dominate ecosystem acidity in many cases, and we have indeed noted this point in the manuscript already (lines 26-27): *“The strong acidity associated with sulfur deposition is likely to dominate downwind ecosystem effects. Consequently, critical load exceedances for highly sensitive aquatic systems in Northern Alberta have mainly been assessed from the perspective of sulfur and nitrogen”*

However, weak acidity has been considered as a possible acidifying agent particularly for remote areas where strong acidity is mostly absent. We have now added literature references for the consideration of weak acids in acidification (although there are admittedly few of them). Downwind of the oil sands, most of the highly sensitive lakes/rivers etc are more likely to be impacted by inorganic acidity. However the degree of LMWOA impact depends upon the critical load that can be tolerated by the ecosystem. In the event that some lakes are just slightly below their critical load (assessed from the standpoint of S deposition), an additional input of even weak acidity may bump the ecosystem into a critical load exceedance. This would of course need to be verified and explored further, but is possible. We have clarified the text to make this point clearer. The paragraph now reads:

“The strong acidity associated with sulfur deposition is likely to dominate downwind ecosystem effects. However, the impact of weak acidity on ecosystem acidification has been previously considered (Vet et al., 2014), and may be particularly important in remote areas (Stavrakou et al., 2012;Keene, 1983). The impact of weak acidity downwind of the oil sands specifically is not clear. Critical load exceedances for highly sensitive aquatic systems in Northern Alberta have mainly been assessed from the perspective of sulfur and nitrogen (Cathcart et al., 2016;Whitfield et al., 2016). While the impact of the large amount of weak organic acidity formed downwind of the oil sands has not been evaluated, it may have a relevant impact on ecosystem acidification in highly sensitive systems which are approaching their respective sulfur and/or nitrogen critical loads. These results warrant a further investigation of the potential impact of this LMWOA emission/formation in this context”

pg.13, line 6: misspelled Lagrangian.

We have fixed the spelling here.

Figure 4: Box D has the note that the blue square indicates the mean value; it is distracting to have this in the middle of the yellow bar – I suggest either removing the legend from the figure and merely describing it in the figure caption, or offsetting it from the data columns so that it cannot be confused with actual data

We have now removed the legend and simply described it in the figure caption.

Figure 9: Please specify that these results are from the photochemical box modeling, as opposed to observationally-derived for clarity

We have made the suggested change.