

Summary/recommendations

This is a generally interesting paper that shows promise to be of use to the community in providing a new method for tackling the poorly-constrained problem of dry deposition. However, there are a significant number of details missing in the methodology that lead me to be unable to fully evaluate the accuracy and use of the measurement techniques presented in this work. I lay out these concerns and then comment on a number of smaller issues. The authors may have internally considered and accounted for each of these issues, and if so details should be provided to strengthen the validity of their work. I recommend that this work be published in ACP after the following suggestions. Due to my concerns listed below, I am currently unable to recommend publication in ACP Letters, but would be willing to consider the revised manuscript as a candidate for ACP Letters.

The authors did not provide enough uncertainty analyses. I go into further details here:

- The authors do not discuss whether there are other possible sources of SO_x/TOS. They mention that they include additional NO_y sources but it's unclear whether those additional sources are being added in at appropriate spatial points in their analyses.
- Relatedly, have the authors considered bidirectional fluxes? See e.g. Fulgham et al., 2020. In that work it was noted that water films and droplets drive equilibrium partitioning, of particular importance for acids. As this study focuses on inorganic acids, there may be importance here. Can the authors discuss and perhaps provide some estimates of bidirectional flux's contribution to measured gas-phase concentrations throughout the flight path? It may be irrelevant in this system (fairly polluted; potentially little partitioning back to the gas phase) but worth doing the calculation and commenting on.
- The authors do not discuss partitioning between SO₂/SO₄ and within various N species. For example, "loss" of SO₂ to SO₄ may be misinterpreted as dry deposition losses. The methods used here to determine effects of partitioning should be explained further, along with any uncertainties that arise.
- The velocity deposition parameter V_d is hard to interpret, given that there are both gas- and particulate-phase species. Gases and particles deposit at different rates, and particles themselves have a size-dependent deposition rate (Emerson et al., 2020). The authors only use [SO₂] in their calculation for V_d but apply this to TOS. What uncertainties are introduced by ignoring [pSO₄], even though [SO₂] is estimated in this work to account for ~90% of dry deposition? My guess is that the uncertainty is small-but it is worth addressing.
- Using an AMS for particle phase species does not provide the "total" particulate mass of a given species. For example, pSO₄ could occur in coarse and fine-mode particles outside of the AMS measurement range. (The authors even state that this is a dusty region, which indicates that it is likely that an amount of nitrate and sulfate will be in the coarse mode.) The authors should discuss this and provide some uncertainty bounds on this issue.

- NOy is a notoriously tricky measurement(s) to make. I was not able to easily determine how the authors define NOy. They state on lines 136-138 “Measurements are discussed in terms of total oxidized sulfur (TOS, the sulfur mass in SO₂ and particle-SO₄ (pSO₄)) and total reactive oxidized nitrogen (TON, the nitrogen mass in reactive oxidized nitrogen species, often denoted NOy)”--it’s unclear if they are including pNO₃ from the AMS in this definition. Chemiluminescence has known interference issues (e.g. Dunlea et al., 2007)-do these interferences impact the TON calculation made here, or are they lumped into TON and not important to separate out? It’s also unclear how the chemiluminescence portion of the NOy set-up is being run and more details should be provided.
- How may deposition rates of different N species vary, and how may this contribute to overall uncertainty in the calculations for TON? The authors discuss how pH influences deposition, and the different N species being considered have different pK_as. The relative pK_a to the pH of the surface will impact a molecule’s dissociation in solution (which in turn impacts how quickly more molecules will be pulled into solution).

Dunlea, E. J., Herndon, S. C., Nelson, D. D., Volkamer, R. M., San Martini, F., Sheehy, P. M., Zahniser, M. S., Shorter, J. H., Wormhoudt, J. C., Lamb, B. K., Allwine, E. J., Gaffney, J. S., Marley, N. A., Grutter, M., Marquez, C., Blanco, S., Cardenas, B., Retama, A., Ramos Villegas, C. R., Kolb, C. E., Molina, L. T., and Molina, M. J.: Evaluation of nitrogen dioxide chemiluminescence monitors in a polluted urban environment, *Atmos. Chem. Phys.*, 7, 2691–2704, <https://doi.org/10.5194/acp-7-2691-2007>, 2007.

Fulgham, S. R., Millet, D. B., Alwe, H. D., Goldstein, A. H., Schobesberger, S., & Farmer, D. K. (2020). Surface wetness as an unexpected control on forest exchange of volatile organic acids. *Geophysical Research Letters*, 47, e2020GL088745. <https://doi.org/10.1029/2020GL088745>

Emerson, E. W., Hodshire, A. L., DeBolt, H. M., Bilsback, K. R., Pierce, J. R., McMeeking, G. R. and Farmer, D. K.: Revisiting particle dry deposition and its role in radiative effect estimates, *Proc. Natl. Acad. Sci. U. S. A.*, 117(42), 26076–26082, doi:10.1073/pnas.2014761117, 2020.

General comments

Section 2.2 Since the work pins upon NOy and SO₂, I recommend including limits of detection for each species and for relevant particulate AMS species.

Line 112-113: “Other NOy species are expected to be greater than that of HNO₃.” Do the authors mean greater as in concentration or greater as in conversion efficiency? Please clarify this statement. Similarly, clarify what is meant by low in “Species like NO₃ radical and N₂O₅ are expected to be low” (line 113).

Line 132-135: The discussion about the collection efficiency of the AMS is a little vague. I suggest adding supplementary information-perhaps a figure showing time-varying CE. I assume the same CE was applied to each AMS species?

Line 156-160: I suggest including the time resolutions of the met/aircraft state parameters.

179: what elevation (height) are the stacks at?

Methods: Please provide units for each quantity/equation you introduce (especially as you are not using SI base units). It was confusing to follow along with each equation. How did you arrive at the units of dry deposition flux F , equation 5, to be $t\ S\ km^2\ hr^{-1}$? Should this be $t\ S\ km^{-2}\ hr^{-1}$, assuming D_{TOS} to be in units of t/hr and area to be in units of km^2 ? I believe this is a typo and that you meant km^{-2} . As verification, eqn 6 for dry dep velocity only works if the dry deposition flux has units of $t\ km^{-2}\ hr^{-1}$. Providing and checking through units for each metric should avoid any typographical of this sort. Note that this unit for F comes again on line 361.

Line 257: I suggest providing a very brief description (1-2 sentences) on what an inferential algorithm is. This will make the paper more accessible to a broader audience.

Line 266: “To assess the potential for a general underestimation of V_d across different inferential deposition algorithms...” Why do the authors assume a model underestimation in V_d a priori?

Results & discussion section: the authors heavily rely on variable abbreviations (e.g. T , TOS , TON , E_{TOS} , E_{TON} , etc) throughout this section. I suggest using words frequently to remind the reader what each variable means & to improve readability. For instance, line 361 “ D ” could be redefined in words.

Line 326: Why did flight F20 only show 2 distinct plumes for TON , and not TOS ?

Lines 338-340: I found this statement to be a little confusing. Is the 92% dry deposition of SO_2 supposed to be for the observed cumulative deposition? I suggest rewording to be more clear.

Section 3.5 This is a really interesting analysis. Did the authors run any model simulations using an adjusted V_d (say by changing pH) to see how much improvement is made in other variables studied in this work, such as TOS and F_{TOS} ? This may equate to a significant amount more of work but would make the paper stronger. Alternatively, the authors may consider speculating/rough calculations on estimated model improvement of other variables by improved V_d .

Results in context for a longer timescale? What is total time scale of measurements (e.g. X hours after emission)

Technical comments

Line 76: First mention of AOSR, needs to be defined here

Line 86: ‘determine emissions’... from? The oil sands specifically or the entire region? I suggest being more explicit here to help define the scope of the field campaign.

Line 147: Define E/N and Td.

Figures and tables

Figure 1: The resolution of the figure is rather low. I suspect this will be fixed in the final version but in the meantime, the color bars are hard to read. I recommend defining TOS and TON in the figure caption.

Figure 3: The figure caption needs more information. Explain what each grouping of subplots mean.