Atmos. Chem. Phys. Discuss., 13, C8364–C8367, 2013
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An airborne assessment of atmospheric particulate emissions from the processing of Athabasca oil sands" *by* S. G. Howell et al.

Anonymous Referee #1

Received and published: 23 October 2013

The paper discusses aerosol particle and trace gas measurements made from the NASA DC-8 and P-3B aircraft around the oil sands facilities near Fort McMurray, Alberta on a few days in the summer of 2008. This work extends previous work on trace gas emissions to estimate fluxes and processes related to particulate quantities. The main results are 1) flux estimates of some aerosol particle quantities and trace gases, 2) the conversion rate of SO2 to sulphate in the plumes is significantly higher than estimated based on reasonable OH concentrations, and 3) little or no organic aerosol was observed at the closest sampling points relative to the plume origins while the measured sulphate concentrations were significant. The measurements are useful,

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but some of the interpretation requires significant revision.

1. The observations suggest that in close to the stacks the fine and ultrafine particles are composed almost entirely sulphate and some BC. There is an absence of significant measured OM in close to the stacks, but the OM increases as the plume ages, evidenced by the observations further downwind. The result of this is that the authors hypothesize that the OM downstream results from sulphuric acid-catalyzed reactions with VOCs. Certainly it seems possible that acid-catalyzed reactions contributed to the increase in OM downstream of the stack (the authors should also consider referencing some of the work on this subject by the paper's editor). However, the evidence for a lack of OM in close to the stack is simply that the AMS did not measure any, yet clearly there were many small particles and at least one form of carbonaceous aerosol (black carbon) present in close to the stack (your Figure 4). The authors need to present a stronger case for the absence of OM. The AMS observations are explained if the OM is in particles too small to be transmitted by the AMS. Liu et al. (Liu, Peter S. K., Deng, Rensheng, Smith, Kenneth A., Williams, Leah R., Jayne, John T., Canagaratna, Manjula R., Moore, Kori, Onasch, Timothy B., Worsnop, Douglas R. and Deshler, Terry. 2007, Transmission Efficiency of an Aerodynamic Focusing Lens System: Comparison of Model Calculations and Laboratory Measurements for the Aerodyne Aerosol Mass Spectrometer, Aerosol Science and Technology, 41: 8, 721-733) show that the AMS lens transmission efficiency at the lower end of the size range drops below 100 nm diameter, and at 50 nm diameter it is about 11%. Thus, if many of the smaller particles that were emitted were less than 50 nm, and it is clear from your Figure 11 that most particles were smaller than 50 nm (indeed, even at 10 km, most were still below 30 nm diameter), then the AMS would not see them and it is possible that they were largely composed of OM. Although no significant volume is evident in the particles <50 nm at 10 km, the observations made in close to the stack appear to indicate that the number concentrations of smaller particles were from 50-100 times more concentrated, depending on how much the real concentrations were suppressed by the counting problems of the CPCs. If the authors insist that OM was not a factor in the

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direct emissions, then they need to undertake proper closure of their chemical and physical measurements to improve their argument.

2. Page 21310, lines 23-24 – dust particles smaller than 1 um diameter have been observed. The statement that the 1 um mode is too small for mechanical generation needs either more qualification.

3. Page 21314 – There are a lot of processes going on over 10 km (and of course you are comparing different days). If the BC particles are initially small, then light absorption will be reduced. As the plume ages, coagulation will enhance the absorption by the BC mass by increasing the effective size of the BC. Coatings may also play a role, but there is ambiguity in that process. This needs a better discussion.

4. Top of page 21313 and Figure 8 caption – What is the difference between the thin dark blue and red curves versus the thick dark blue and red curves? In the caption, you say "despite the obvious convection", but the clouds pictured in Figure 2 do not indicate strong convective, consistent with your profile. The apparent differences between your upwind profile data and the plume profile data from the lidar are likely due to the combination of the elevated stack, the plume rise and the wind shear (discussed at beginning of section 3). Also, indicate what Zm is in the caption.

5. Page 21314, lines 6-17 - If you use a constant 1 m/s for the exhaust from the "mixed layer", you are clearly and significantly overestimating the impact of the clouds. The clouds pictured in your Figure 2 are weak and scattered. The mean updraft across one of those clouds would not be 1 m/s, and there are very few clouds. A value of 0.1 m/s will be closer, but even that is more likely to be an overestimate of the cloud effect. Also, the clouds do not completely vent.

6. The "clearly visible" "Convection out of the mixed layer" referring to Figure 7a is curious. If it were such, the plume at that point should show more continuous mixing, and why does no cloud show up over top of it? Why would there also not be such lifting under the cloud at 1806-1809, which is the most significant entity in Figure 7a? It is not

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evident that the aerosol above 3 km from 1802-1806 is directly connected to the plume that is below 1.5 km. The aerosol signal above 3km could be the beginning or ending of cloud. The picture is much more complicated than the authors suggest.

7. Indicate the altitude of the aircraft in the image or caption of Fig 7a.

8. Page 21315 – "7 July" ? do you mean 10 July? If not, please explain

9. Page 21316, lines 17-20 – If some sulphate was scavenged by dust particles, then the reduced transmission of the AMS above 600 nm would result in a lower sulphate measurement. This is an explanation that needs to be added.

10. In Figure 10 and its caption: what does "minimal plume period" mean? Did your PSAP filter transmission drop about 1930 on June 28?

11. Page 21319, line 19 – change "appear to" to "may also".

12. Conclusions - For reasons of cloud venting discussed above, your flux estimates need to be recalculated. It is also important to indicate that these are only one daytime estimate.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 21301, 2013.