

Referee Comments for Osman, M. et al.,

“Real time analysis of insoluble particles in glacial ice using single particle mass spectrometry”

General Comments

The authors implement a PALMS to successfully employ nebulizer+instrument techniques previously used by single particle soot photometers to make measurements of particle classification, size and concentration in ice core samples from Greenland. Though limited by a low particle transmission that is currently inherent to the PALMS instrument, the methodology still manages to result in new, interesting measurements that to my knowledge have not been realized before, making this a very worthy manuscript for publication after addressing some relatively minor concerns. The results also provide substantial motivation to continue to push the transmission-efficiency capabilities of the PALMS instrument, the results of which would make the methods used here a much more viable path towards measurement of particle concentrations and size distributions in ice cores.

The presentation of the goals, setup, methodology and results are generally very clearly stated, with very few exceptions (notes below). Further, the manuscript seems to have been carefully prepared, as I struggled to find any typos, spelling errors or poor grammar. Below, I suggest some minor corrections to be addressed, including explanation of their monitoring of potential background contamination levels and system stability. Also, I suspect there may be an error in their calculation of nebulization efficiency (though it results in only a small change in the quoted number). Finally, I recommend more carefully explaining the differences in interpreting PALMS vs SP2 measurements of black carbon / soot.

Specific Comments

Page 3, line 18: Also see Katich et al., 2017 (doi:10.1080/02786826.2017.1280597), which provides a lengthy closely-related discussion on aerosolizing particulate from snow and ice.

Page 7, line 5,6: Did the authors intersperse regular measurements of ‘blanks’ (i.e. ultra-pure water) to quantify the average background level of particulate seen by PALMS when using a ‘clean’ nebulization system? There is mention of looking for background from the stainless steel band saw (not what I’m concerned about here) and of sonicating the parts between samples. But I wonder if there is a quantification of average background levels due to any residual particulate in the nebulizer lines? How does this compare to signal levels? Negligible?

Page 8, line 19: Please clarify the phrase “rate of liquid nebulization” ... same as rate at which liquid is fed to the nebulizer i.e. liquid uptake rate? Does this occur at a user-controlled pump rate, or is it self-aspirating? If you have control over the pump rate, this could be another way to tweak the rate of particulate delivered to the PALMS inlet.

Page 9, line 23: Agreed, the nebulization efficiency can drift over time, even substantially, depending on the solution being nebulized. Was this monitored by occasionally measuring transmission of the 8.8e6 particles/cc solution in between samples? If so, perhaps show a summary of nebulization stability in supplemental material?

Page 11, line 6: The statement, “Scaling the efficiency curve by the ratio of excess-flow to the PALMS inlet flow”, I believe should read “Scaling the efficiency curve by the ratio of **TOTAL**-flow to the PALMS inlet flow.”

Further, I don't think I agree with the calculation of the nebulization efficiency, where it is achieved simply by making the correction due to particle loss from low PALMS sample flow. I would argue the following: If a nebulizer's efficiency is defined as the ratio of 'the rate of particles emerging in aerosol from the nebulizer' (call it $R_{aerosol} = N_{aerosol}/sec$) to 'the rate of particles introduced to the nebulizer' (call it $R_{introduced}$, which is known from your known PSL concentration and liquid uptake rate), then to know $N_{aerosol}$, you have to work backwards from the number of particles that PALMS sees.

$$\varepsilon_{nebulization} = \frac{R_{aerosol}}{R_{introduced}} = \frac{N_{aerosol}/sec}{R_{introduced}} = \frac{N_{aerosol}/sec}{m_{psl} \cdot V_{neb}}$$

working backward from the number of particles need by PALMS...

$$(N_{aerosol}/sec) = \underbrace{\left(\frac{N_{particles\ detected\ by\ PALMS}}{sec}\right)}_{(1)} \cdot \underbrace{\left(\frac{N_{particles\ introduced\ to\ PALMS}}{N_{particles\ detected\ by\ PALMS}}\right)}_{(2)} \cdot \underbrace{\left(\frac{F_{wet} + F_{dry}}{F_{inlet}}\right)}_{(3)} \underbrace{\left(\frac{F_{flow}}{F_{dry}}\right)}_{(4)}$$

where

(1) = f_{PALMS}

(2) = ε_{trans}

(3) = the correction introduced in the text

(4) = needed to correct for the fact that only 2/5 of the flow into PALMS has gone through the nebulizer (i.e. F_{dry} is just dilution air)

thus...

$$\varepsilon_{neb} = \varepsilon_{PSL} \cdot \frac{1}{\varepsilon_{trans}} \cdot \left(\frac{F_{wet} + F_{dry}}{F_{Dry}}\right) = \frac{\varepsilon_{PSL}}{\varepsilon_{trans}} \cdot \left(\frac{F_{flow}}{F_{Dry}}\right)$$

Plugging in some maximal numbers from figure 2, I get:

$$\varepsilon_{neb(max)} = \left(\frac{0.004}{0.15}\right) \cdot \left(\frac{5}{2}\right) \cong 6.7\%$$

it's not far off the quoted 4%, but as a matter of correctness, should be changed (assuming I've not erred).

Page 16, line 6: Regarding the 0.8 g/cm^3 density used for soot...What density are you referring to? Void free rBC? All recent literature that I know of uses either 2.0 g/cm^3 (a bit of an old number) or 1.8 g/cm^3 , so maybe this is a typo, or maybe you have actually used the wrong number, or maybe you are referring to a density that isn't clarified. Please comment...

Figure 4: It is important to note somewhere that the soot size distributions here are not directly comparable to typical rBC size distributions shown in literature that are measured via incandescence, i.e. SP2 measurements. My understanding is that PALMS will only measure the size of the entire soot-containing particle (via a scattered-light signal), which includes any 'coating' that is combined with the BC particle, and is not a measurement of the 'core' refractory BC mass. On the other hand, SP2 measurements will separate the rBC core mass (or volume-equivalent-diameter, VED) from the coating associated with an individual rBC particle. This rBC core VED distribution is what is typically shown in literature. So one could not compare the soot distributions shown here to, say, Schwarz et al. 2013 ("Black Carbon Aerosol Size in Snow"). A slightly expanded discussion on the interpretation of SP2 vs PALMS measurements of soot/rBC is recommended.

Technical Corrections

Page 3, line 15: 'Schwartz' should be Schwarz.