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

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S1: Nebulization drift tests

In this section, we test the extent of nebulization drift over an hour-long measurement period (i.e., the longest sample integration period performed in the manuscript). This was tested by directing a particle-laden airflow (nebulized from the monodisperse, 746 nm PSL liquid standard: $m_{PSL}(D_p$ = 746 nm) = $8.8x10^6$ PSL particles cm^{-3}; Sect. 2.3.3), to an optical particle sizer (OPS; MesaLabs Bios DryCal 220) and making continuous, one-second interval measurements over three separate ~1-hour long tests. The nebulization efficiency (ε_{neb}) was calculated as

$$\varepsilon_{neb}\left(D_p = 746\ nm\right) = \frac{n_{OPS}(D_p{=}746\ nm)\cdot F_{flow}}{m_{PSL}(D_p{=}746\ nm)\cdot V_{neb}} \qquad \text{(S1)}$$

where n_{OPS} is the PSL number concentration measured by the OPS, and the flow rates F_{neb}, V_{neb} and, F_{wet} are as described in the main text. The long-term drifts in nebulization, calculated as the linear percent change over the hour-long measurement interval, were determined in the three tests to be 22%, 9.2%, and -33 % ($\Delta\varepsilon_{neb}/\Delta t = 0.18\cdot10^{-5}\ s^{-1}$, $0.08\cdot10^{-5}\ s^{-1}$, and $-0.30\cdot10^{-5}\ s^{-1}$ for test #1, test #2, and test #3, respectively; Fig. S1.). Importantly, results of the three tests indicated that drift direction was not systematic, as both negative and positive drift biases occurred over the one-hour nebulization periods (Fig. S1, shown below). It is thus reasonable to view the drift uncertainty as a simple spread about the hour-long mean of the three tests, in this case equating to $\varepsilon_{neb} = 0.068 \pm 0.013$ (1 s.d.), or ~18% relative uncertainty.

It is equally important to note that in our study, calculation of particle mass-concentration (eq. 4) does not explicitly incorporate estimates of ε_{neb}, but rather estimates of the extraction efficiency, ε (eq. 5), determined experimentally and independent of ε_{neb}. However, via eq. 6 (now included in the main text; see C5 below), ε is shown to be a function of ε_{neb} and transmission efficiency, ε_{trans}. Since past studies (e.g., Cziczo et al., 2006) have shown PALMS transmission to be relatively stable, we thus take the uncertainty interval calculated for the extraction efficiency parameterization, ε (~30% relative uncertainty at $\varepsilon(D_p$ = 746 nm); eq. 5) to implicitly encapsulate uncertainties in nebulization efficiency.

References:

Cziczo, D. J., Thomson, D. S., Thompson, T. L., DeMott, P. J., Murphy, D. M: Particle analysis by laser mass spectrometry (PALMS) studies of ice nuclei and other low number density particles, Int. J. Mass Spectrometry, 258, 21-29. 2006.

Figure

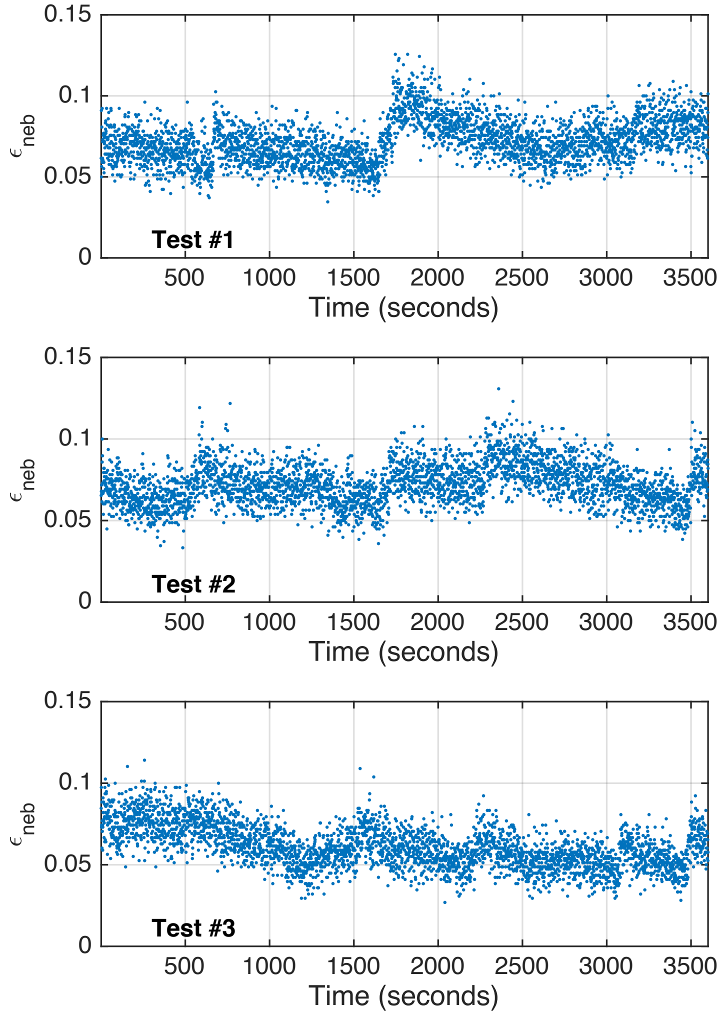

Fig. S1. Results of three separate nebulization drift tests.