Supplement of

Aircraft measurements of High Arctic springtime aerosol show evidence for vertically varying sources, transport and composition

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1 Supplementary Methods

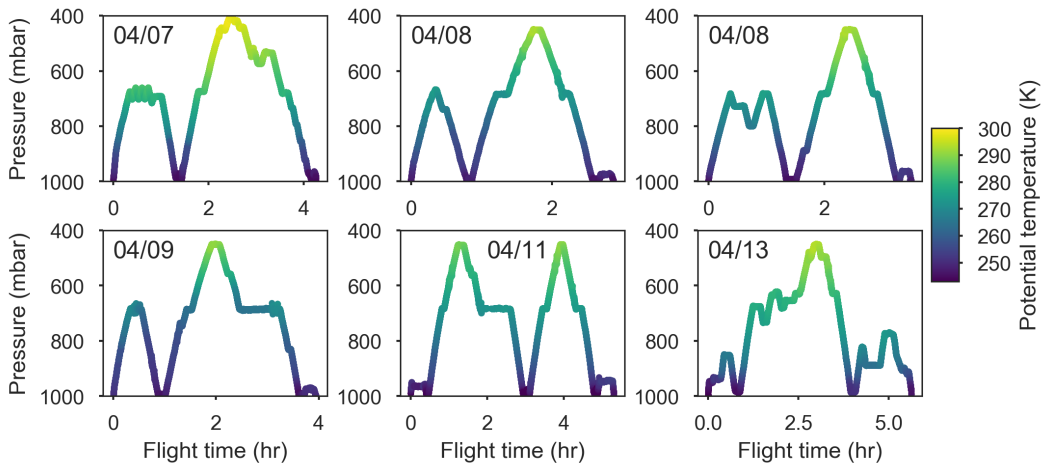

Figure S1: Pressure versus time since take-off, coloured by measured potential temperature, for all six High Arctic NETCARE flights during 7 – 13 April 2015. Note that the full flight is shown in each case; however, this work only considers measurements made at pressures higher than ∼640 mbar (i.e., below ∼3.5 km) because the ToF-AMS was not run at lower ambient pressures.

		April 7, 8 (leg 1), 9, 11	April 8 (leg 2), 13
Menu 1	Mode	Ensemble MS, SP laser on	Ensemble MS, SP laser on
	Duration	10s (5s open, 5s closed)	10s (5s open, 5s closed)
Menu 2	Mode	Ensemble MS, SP laser off	Ensemble MS, SP laser off
	Duration	10s (5s open, 5s closed)	10s (5s open, 5s closed)
Menu 3	Mode	epToF, SP laser on	Event Trigger, SP laser on
	Duration	10s	10s

Table S1: Summary of ToF-AMS Operation Modes during NETCARE 2015

ROI (m/z)	Threshold (Ions/Extraction)
46	1
64	1
45 - 150	3

Table S2: Event Trigger Regions of Interest

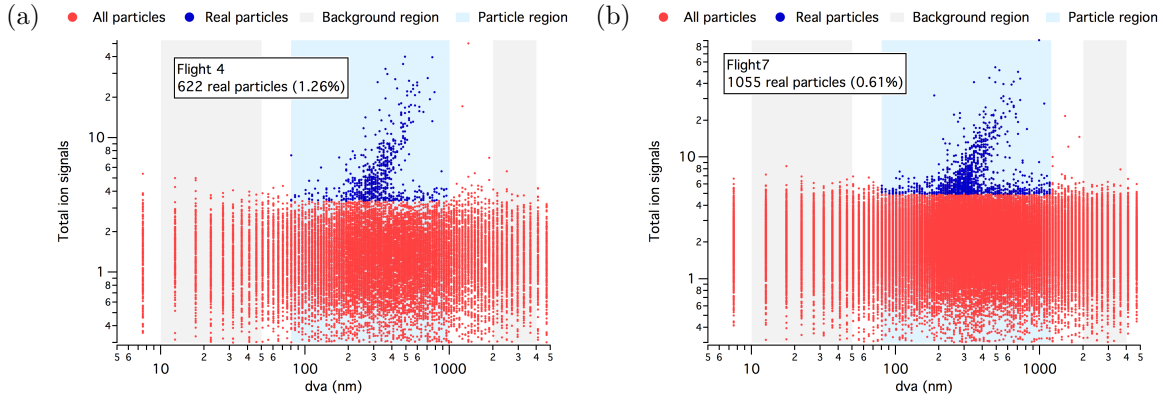

Figure S2: (a) Total aerosol ion signal, excluding air peaks, for all ETSP mass spectra collected during flight 4 on 8 April, 2015 (red points). Blue points represent particle spectra identified as "real" based on setting a threshold of the mean aerosol ion signal in background regions (grey shading) plus three times its standard deviation. Blue shading indicates the size range over which "real" particles are selected. 622 MS spectra were identified as associated with "real" particle events during flight 4, corresponding to 1.26% of total spectra collected during this flight. (b) As in (a) but for flight 7 on 13 April, 2015. 1055 MS spectra were identified as associated with "real" particle events during flight 7, corresponding to 0.61% of total spectra collected during this flight.

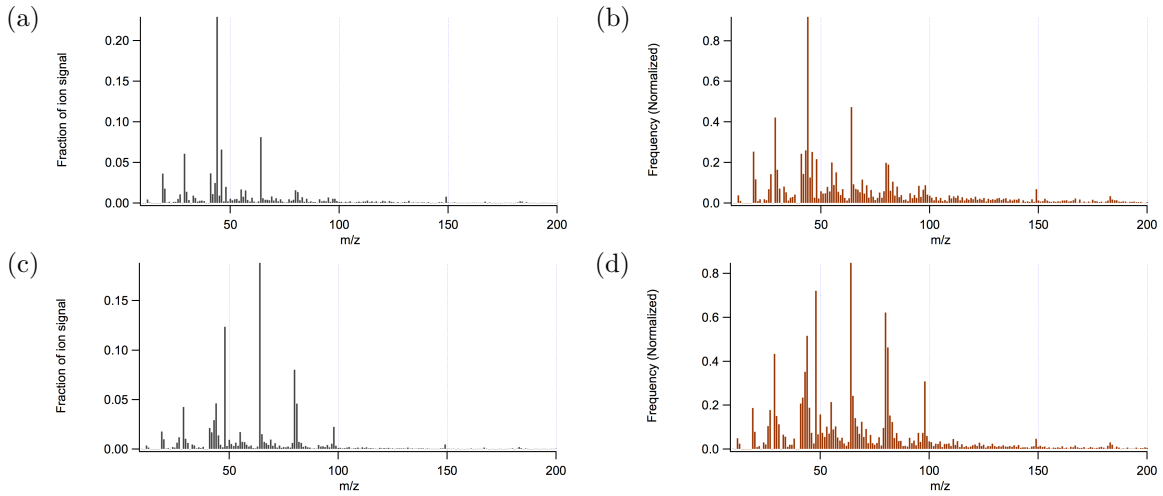

Figure S3: (a) Mean mass spectrum and mass spectral histogram (b) for particle class 1 of the two cluster solution. (c) Mean mass spectrum and mass spectral histogram (d) for particle class 2 of the two cluster solution.

2 Supplementary Results

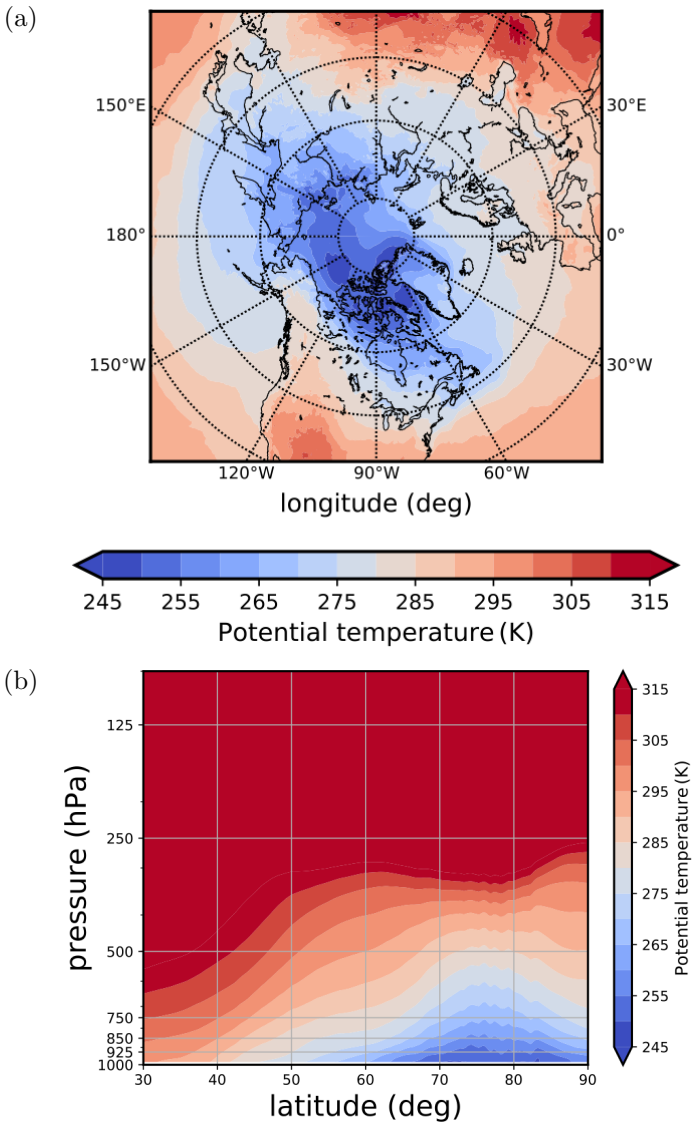

Figure S4: (a) Map of mean potential temperature during 7 – 13 April 2015 from ECMWF. (b) Zonal mean potential temperature from ECMWF during 7 – 13 April 2015, averaged between -110°W and -60°W.

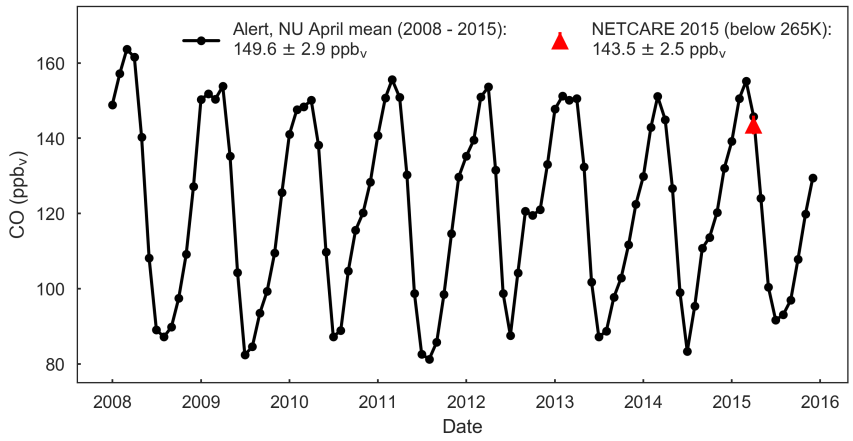

Figure S5: Monthly mean carbon monoxide concentrations at Alert, NU during 2008 – 2015 (black) (Novelli et al., 2016). The mean (± standard deviation) CO concentrations in April 2008 – 2015 at Alert was 149.6 ± 2.9 ppbv. The mean (± standard deviation) CO concentration measured during NETCARE 2015, from the surface up to 265 K (∼1500 m), is shown with the red triangle (143.5 ± 2.5 ppbv).

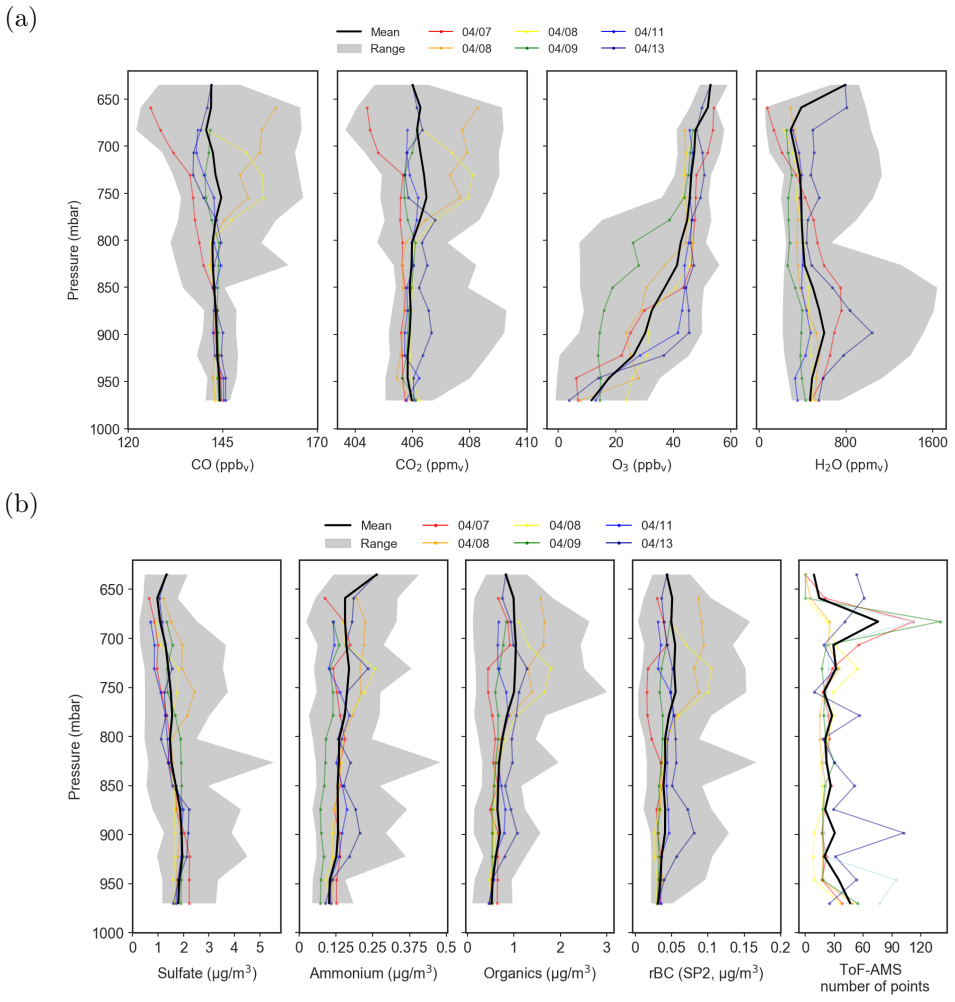

Figure S6: (a) Mean pressure profiles of trace gases in the polar dome observed during 7 – 13 April 2015, including carbon monoxide, carbon dioxide, ozone and water vapour. (b) Mean pressure profiles of sub-micron aerosol composition in the polar dome observed during 7 – 13 April 2015, including sulphate, organics and ammonium from the ToF-AMS and refractory black carbon (rBC) from the SP2. The profile of number of points represents the number of measurements from the ToF-AMS, which had the slowest sampling rate of all instruments deployed during NETCARE 2015.

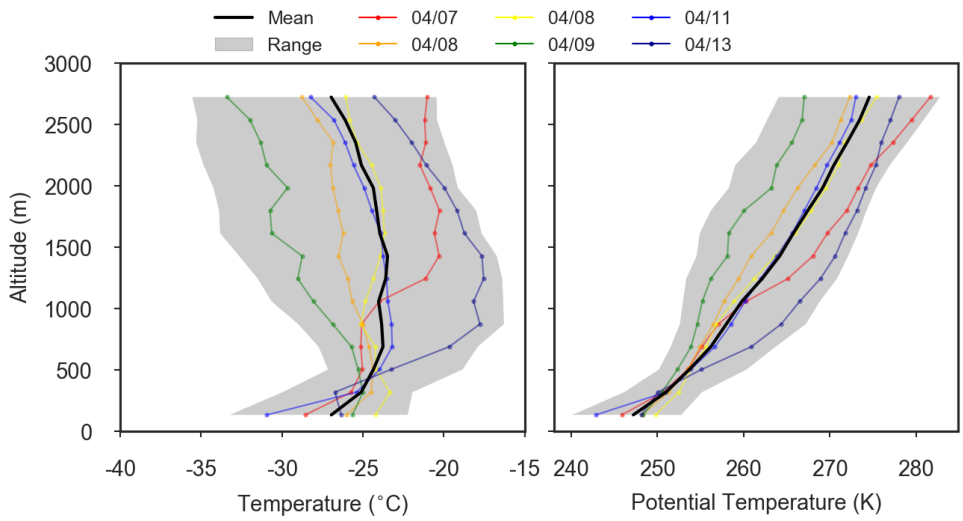

Figure S7: Mean altitude profiles of temperature (left) and potential temperature (right) in the polar dome observed during 7 – 13 April 2015. Coloured lines indicate the mean profile for each flight, the black line represents the mean profile over all flights, and gray shading shows the range of observations in each altitude bin.

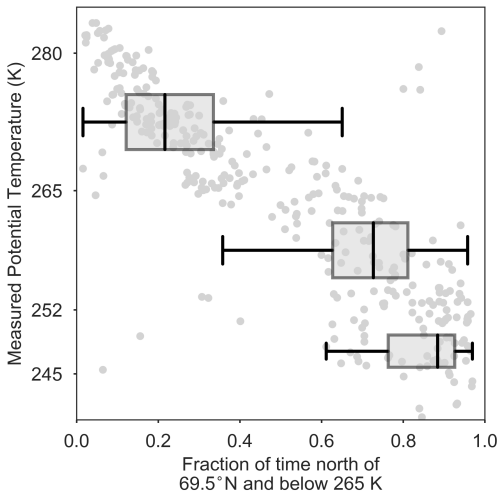

Figure S8: Observed potential temperature (K) as a function of FLEXPART-ECMWF predicted fraction of the past 10 days in the mid-to-lower polar dome (i.e., below 265 K and north of 69.5°N). The FLEXPART-ECMWF relative residence time is binned in the lower (245 – 252 K), middle (252 – 265 K) and upper (265 – 280 K) polar dome.

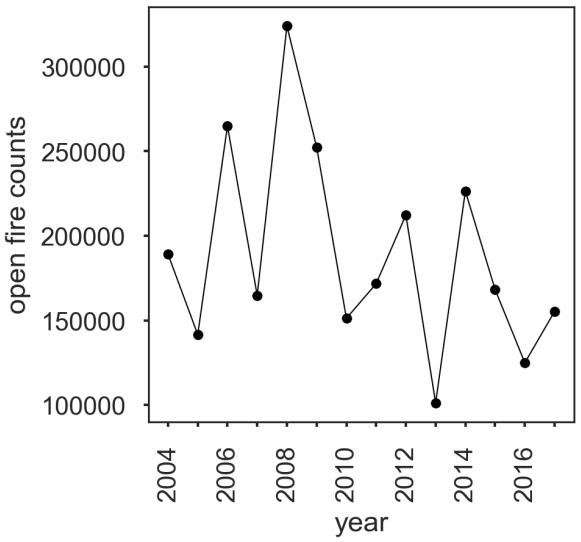

Figure S9: Active fire counts in the Northern Hemisphere north of 35°N during March to May for the period 2004 – 2017 from MODIS C6 (obtained from https://firms.modaps.eosdis.nasa.gov/download/)

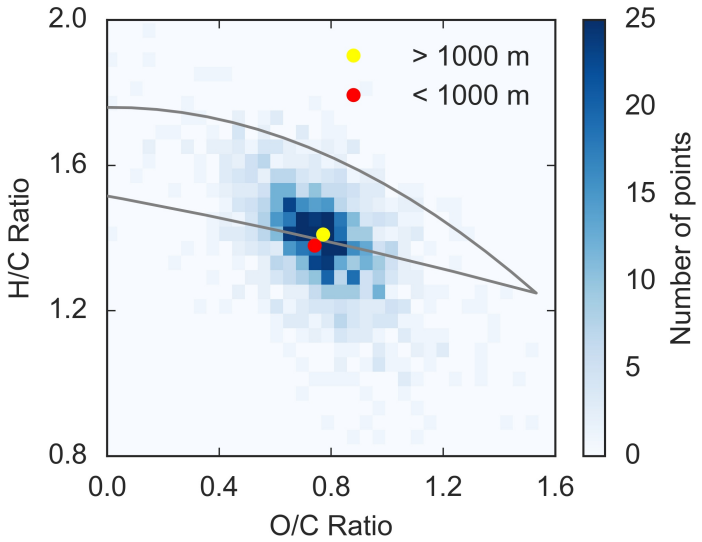

Figure S10: Two dimensional histogram showing the relationship between the oxygen-to-carbon (O/C) and hydrogen-to-carbon (H/C) ratio estimated by the ToF-AMS according to the method described in Canagaratna et al. (2015), where the color scale represents the number of observations. Grey lines represent the ambient range observed by Ng et al. (2011). Red and yellow circles represent the mean O/C and H/C ratios below and above 1 km, respectively, showing no discernible difference within error.

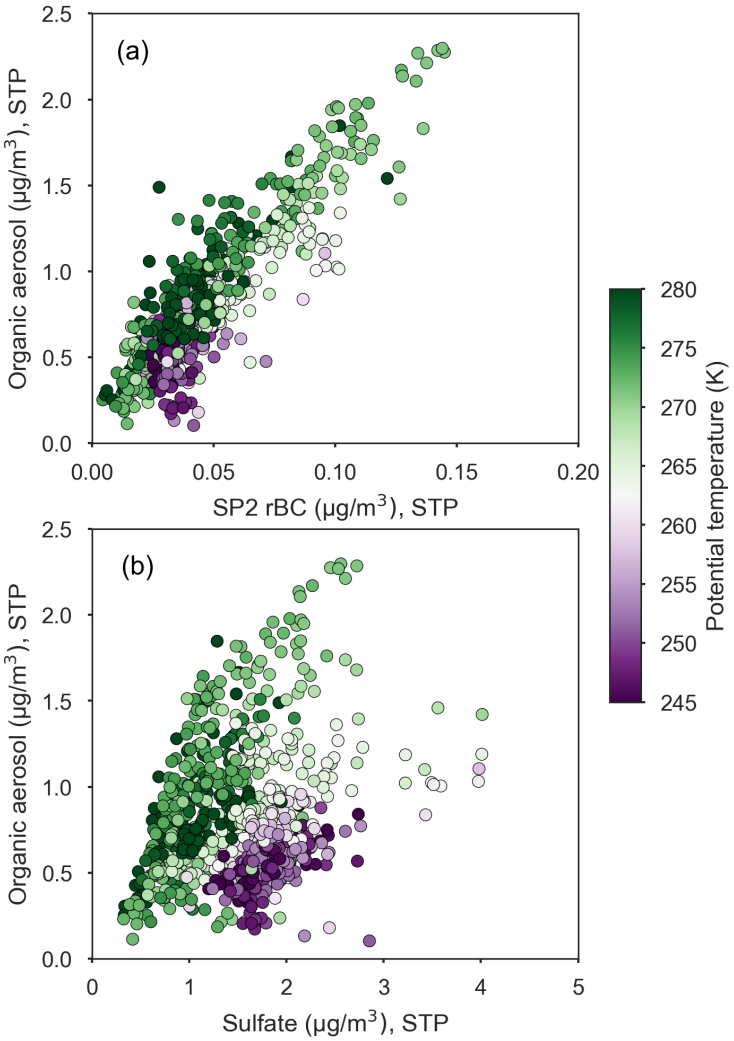

Figure S11: (a) ToF-AMS organic aerosol versus SP2 refractory black carbon (rBC), coloured by measured potential temperature. (b) ToF-AMS organic aerosol versus ToF-AMS sulphate, coloured by measured potential temperature.

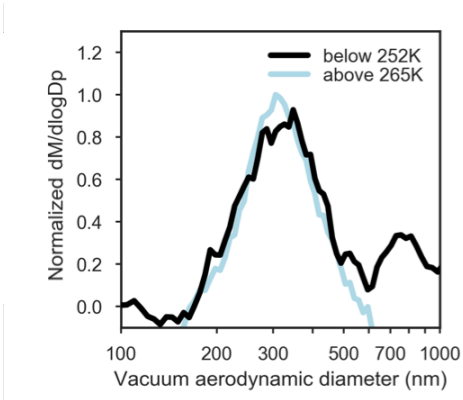

Figure S12: Normalized mean ToF-AMS size distributions of organic aerosol subset by observed potential temperature: below 252 K (black), above 265 K (light blue).

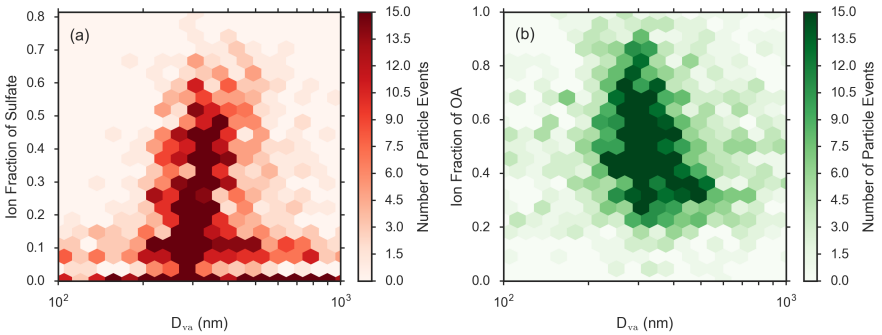

Figure S13: (a) Ion fraction of sulfate in single particle (ETSP) mass spectra as a function of particle size. (b) Ion fraction of total organic aerosol in single particle mass spectra as a function of particle size. The colour scale in both plots represents the number of real particle events observed. A total of 1677 real particle spectra were obtained from the combined observations on two flights (8 April and 13 April, 2015).

References

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