



Iodine observed in  
new particle  
formation events

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# Iodine observed in new particle formation events in the Arctic atmosphere during ACCACIA

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## Abstract

Accurately accounting for new particle formation (NPF) is crucial to our ability to predict aerosol number concentrations and thus cloud properties, which is in turn vital in simulating radiative transfer and climate. Here we present an analysis of NPF events observed in the Greenland Sea during the summertime as part of the Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) project. While NPF events have been reported in the Arctic before, we were able, for the first time, to detect iodine in the growing particles using an Aerosol Mass Spectrometer (AMS) during a persistent event in the region of the coastal sea ice near Greenland. Given the potency of iodine as a nucleation precursor, the results imply that iodine was responsible for the initial NPF, a phenomenon that has been reported at lower latitudes and associated with molecular iodine emissions from coastal macroalgae. The initial source of iodine in this instance is not clear, but it was associated with air originating approximately 1 day previously over melting coastal sea ice. These results show that atmospheric models must consider iodine as a source of new particles in addition to established precursors such as sulphur compounds.

## 1 Introduction

In the Arctic, clouds are the dominant factor in the control of the incoming and outgoing energy balance at the Earth's surface and, here and throughout the troposphere, the largest single source of uncertainty in climate predictions. Understanding processes governing atmospheric aerosol concentrations is critically important to improved prediction of clouds and thus weather and climate (Boucher et al., 2013). The optical thickness and lifetime of clouds can be strongly influenced by the population of aerosol particles available to act as cloud condensation nuclei (CCN) (Haywood and Boucher, 2000). Particle concentrations in the summertime Arctic are typically very low (of the order of  $10^2 \text{ cm}^{-3}$ ) and therefore cloud properties in this region are highly sensitive

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to the mechanisms by which new particles are formed and are grown to viable CCN sizes (roughly 50–100 nm, depending on the cloud conditions) (Merikanto et al., 2009). Understanding these processes is crucial to our predictive capability of climate in the Arctic, as clouds can have a strong warming or cooling effect, depending on a variety of conditions (Hodson et al., 2013).

New particle formation (NPF) can dramatically increase aerosol number concentrations in the atmosphere (Kulmala et al., 2004; Spracklen et al., 2006), alongside direct emissions of particles (e.g. through combustion, sea spray, dust suspension etc.), contributing up to half of the global CCN burden (Merikanto et al., 2009; Yu and Luo, 2009). NPF generally occurs through the rapid photochemical production of low vapour pressure secondary material such that stable molecular clusters are able to grow to viable sizes (nm) (Kulmala et al., 2013, 2000). As they grow, the freshly nucleated particles act as sinks for the secondary condensable material, potentially shutting off the nucleation process. NPF has been observed in a variety of different environments across the world. The role of sulphuric acid (produced in the atmosphere from the gas phase oxidation of marine biogenic dimethyl sulphide or SO<sub>2</sub> from fossil fuel burning or volcanoes) and organic matter (chiefly from terrestrial biogenic sources) has been extensively studied (e.g. Laaksonen et al., 2008; Q. Zhang et al., 2004; Riccobono et al., 2014) and it has also been shown that ammonia and amines have very important roles in promoting NPF through ternary processes (Kirkby et al., 2011; Almeida et al., 2013; Berndt et al., 2014).

Because of the generally low number concentrations present, the Arctic CCN population is highly sensitive to NPF and thus the processes responsible, which are currently highly uncertain in this environment (Lee et al., 2012). NPF has previously been observed during Arctic ship-based measurements and given the lack of significant biogenic or anthropogenic sources of organic precursors, most efforts to model these observations have only invoked NPF from the oxidation of sulphur species (Korhonen et al., 2008). An alternative hypothesis is that rather than nucleation, the initial source

of the new particles is the fission of organic biogels in primary particles (Karl et al., 2013, 2012).

In coastal environments at lower latitudes, frequent daytime NPF events have been observed associated with gaseous iodine at low tide. This suggests that in these environments, iodine is the dominant source of nanoparticles, which have been observed to grow to larger sizes able to scatter radiation and contribute to CCN (McFiggans et al., 2004, 2010; O'Dowd et al., 2002; Whitehead et al., 2009; Yoon et al., 2006; Lehtipalo et al., 2010). Gaseous I<sub>2</sub> is produced in abundance by macroalgae species (*Laminaria digitata*, *Fucus vesiculosus* and *Ascophyllum nodosum* have been identified as being responsible for NPF) in response to being exposed to the atmosphere (Küpper et al., 2008; Huang et al., 2013) and is rapidly photooxidised to iodine monoxide (IO) and higher iodine oxides, which polymerise forming particles (Saiz-Lopez et al., 2012; Saunders et al., 2010). To date, iodine-initiated NPF has most commonly been observed in seaweed-rich coastal areas but the theoretical potential exists in low background aerosol conditions with associated high iodine fluxes (Mahajan et al., 2010). Atkinson et al. (2012) attributed NPF events observed in the Weddell Sea in Antarctica to iodine emissions from sea ice, although no data on aerosol composition was presented.

Here we present data on NPF events recorded aboard the RRS *James Clark Ross* in the Greenland Sea during the summer ACCACIA cruise. Through the analysis of a particularly strong and persistent case study, we show evidence for the role of iodine in NPF events in this region.

## 2 Methods

As part of the Aerosol-Cloud Coupling And Climate Interactions in the Arctic (ACCACIA) project, intensive measurements of aerosol composition and properties were made aboard the RRS *James Clark Ross*, an ice-hardened research vessel. The cruise

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(JR288) consisted of a number of traverses in and out of the sea ice margin in the region of Greenland and Svalbard during July and August 2013 (see Fig. 1).

The University of Manchester instrumentation was located in an instrumented sea container on the foredeck of the *James Clark Ross*, sampling air through a 5 m stack via a 3.5  $\mu\text{m}$  cut cyclone, as has been performed during previous measurement campaigns (Allan et al., 2009). No attempt to correct for size-dependent particle losses has been made for this work, as it will not affect the qualitative results presented here. No in-line drier was used, as the relative humidity (RH) in the container was maintained low due to the temperature differential. A number of instruments sub-sampled from the main inlet manifold.

A Differential Mobility Particle Sizer (DMPS) system was used to measure size-resolved particle number concentrations. This was built at the University of Manchester (Williams et al., 2007) using dual “Vienna” design Differential Mobility Analysers (DMAs) (Winklmayr et al., 1991) of different lengths (to cover different particle size ranges) with stepping voltages, selecting negatively-charged particles. TSI (Shoreview, MN, USA) model 3010 and 3025a Condensation Particle Counters (CPCs) were used to count the particles and the Wiedensohler (1988) charging parameterisation used to invert the data. The sheath air system uses a recirculating system dried to a low (< 20 % RH) humidity using a membrane drier linked to a dry compressed air system. Total number concentrations were provided by a TSI model 3776 CPC.

An Aerodyne (Billerica, MA, USA) Aerosol Mass Spectrometer (AMS) of the High Resolution Time-of-Flight (HR-TOF) design (Canagaratna et al., 2007) was also used to measure particle composition. Calibrations were performed using monodisperse ammonium nitrate for mass and NIST-certified Polystyrene Latex (PSL) Spheres (Thermo Scientific) for size. The data was collected in ‘V’ and ‘W’ mass spectral modes, which have  $m/\Delta m$  resolutions of 2100 and 4300 at  $m/z = 200$  respectively (DeCarlo et al., 2006), although only ‘V’ mode data is presented here due because of the low signal-to-noise ratio of the ‘W’ mode data.

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A Droplet Measurement Technologies (Boulder, CO, USA) Single Particle Soot Photometer (SP2) was used to measure black carbon. This was a version ‘D’ instrument, calibrated using monodisperse Aquadag, scaled by a factor of 0.75 as recommended by Laborde et al. (2012)

Sub-saturated particle growth factors were measured using a Hygroscopicity Tandem Differential Mobility Analyser (HTDMA). This was the second Manchester-built instrument (Whitehead et al., 2014), conforming to EUSAAR specifications (Duplissy et al., 2009). This uses two Brechtel Manufacturing Inc. (Hayward, CA, USA) DMAs housed in temperature-controlled boxes and linked in series. The first had dry sheath air (< 20 % RH) while the sheath air in the second was maintained at 90 % RH, using a membrane humidification system on a feedback loop with an Edgetech (Marlborough, MA, USA) dew point monitor. The particles were counted using a TSI model 3782 water-based CPC. Sizes were calibrated with PSL spheres and humidity was validated through the comparison of the measured deliquescence humidities of ammonium sulphate and sodium chloride with modelled values (Topping et al., 2005). Data were inverted using the method of Gysel et al. (2009).

Halocarbons were quantified in air and seawater using an Agilent 6850 gas chromatograph (GC) with 5975C MSD coupled to a commercial thermal desorption unit (TD, Markes Unity2-CIA8) (Andrews et al., 2014).

Back trajectory analysis was performed using HYSPLIT 4 (Draxler and Hess, 1998), employing GDAS reanalysis wind fields (NOAA Air Resources Laboratory, Boulder, CO, USA). Back trajectories were started at 20 m above mean sea level and run using modelled vertical velocities.

### 3 Results

During the measurements, a number of events were noted in the DMPS data whereby a large number of small particles contributed dramatically to the ambient population. Contributions from combustion sources (e.g. ship plumes) were eliminated by the lack

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of black carbon (BC) detectable by the SP2. The data were also filtered according to short-lived spikes seen with the CPC that could be associated with other sources from within the ship. The DMPS occasionally showed “open” distributions, whereby the peak existed at or below the lower size limit of the instrument (3 nm), providing evidence for NPF (Fig. S1.1). Time periods where a significant portion of the detected particles were smaller than 10 nm are identified on Fig. 1a, on an overlay of the cruise track and sea ice concentration data using the polar stereographic product from the Special Sensor Microwave Imager/Sounder (SSMIS) instrument on the Defense Meteorological Satellite Program (DMSP) F17 platform (Cavaliere et al., 2013).

These periods all occurred in proximity to coastal locations around Iceland, Greenland and Svalbard. The most significant and persistent of these events occurred between the 25 and 27 July, when the ship was close to the sea ice margin off the coast of northeastern Greenland and the air had previously travelled over the breaking sea ice off Greenland. (Fig. 1b). Markers for ice diatom activity in the form of  $\text{CH}_2\text{Br}_2$  were also observed to be elevated during this period (Fig. 1c). During this period, the nonrefractory aerosol composition was mainly organic, with only around  $0.1 \mu\text{g m}^{-3}$  of sulphate present (Fig. S1.2).

The candidate NPF events were also associated with elevated number concentrations, with transitions over periods of hours (Figs. 2 and S1.1 in the Supplement), indicating that the new particles last long enough to grow to the larger sizes that can contribute to CCN. It is likely that the transitions in the size distributions resulted from changes to the source footprint in relation to the position of the ship rather than in situ growth of the observed new particles. It is worth noting that the characteristic “banana” growth events observed at coastal sites and other locations (Kulmala et al., 2004; Ehn et al., 2010; Yli-Juuti et al., 2011), result from diurnal modulation of boundary layer dynamics and photochemistry, which is missing under the continuous insolation of the Arctic summer.

Aerosol Mass Spectrometer (AMS) data during the 25–27 July showed a signal at  $m/z = 127$  during the periods when larger particles were present (Fig. 3), which

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is identified as  $I^+$  ions by its precise mass/charge ratio of 126.90 (Fig. S1.3) (Wang et al., 2012).  $I^+$  has previously been reported as the largest peak in photochemically-produced iodine oxide AMS mass spectra in the laboratory (McFiggans et al., 2004; Jimenez et al., 2003), but this is the first time that it has been reported in ambient particles. In previous coastal studies, owing to the proximity to the initial source of iodine, particles did not grow to sizes large enough (around 30 nm) to be transmitted by the AMS aerodynamic lens inlet (Liu et al., 2007; Q. Zhang et al., 2004; X. F. Zhang et al., 2004).

To investigate whether the  $I^+$  signal could be associated with processes governing the formation of organic aerosols (Fig. S1.2.), Positive Matrix Factorisation (PMF) was performed on the data (Paatero and Tapper, 1994; Ulbrich et al., 2009). Once shipping emissions and misattributed sea salt are excluded, this analysis found that the organic matter detected by the AMS could be attributed to methyl sulphonic acid (MSA) (Phinney et al., 2006; Decesari et al., 2011) and highly-oxygenated organic material (McFiggans et al., 2005; Jimenez et al., 2009). The  $I^+$  signal was not represented in any of the factors derived, only manifested in the residual data, which implies that the particulate iodine had a source that was distinct from the processes controlling the formation of particulate organic matter (be they primary or secondary). A further implication of the MSA observation is that this compound was at least partly responsible for the reported sulphate concentrations, as this also produces  $SO^+$  and  $SO_2^+$  ions in the mass spectrum (Zorn et al., 2008). Therefore, the actual non-seasalt sulphate concentration is likely to be lower than what is reported, but the quantitative fraction of MSA is difficult to estimate, as the fragmentation behaviour is highly variable and not calibrated during this study. Further details are presented in section S2 of the supplementary material.

## 4 Discussion

To link the  $I^+$  signals detected by the AMS to the particles seen by the DMPS, the size-resolved data from the two instruments were quantitatively compared through the fitting



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of lognormal distributions. Shown in Fig. 4 are the DMPS volume-weighted size distributions from the period of peak  $I^+$  concentrations (26 July 22:50–23:45 UTC), together with the AMS Particle Time-of-Flight size-resolved data for  $I^+$ , organics and sulphate. The AMS data was of a low signal-to-noise ratio, due to the short averaging time and low signal levels (Allan et al., 2003), however the fits converged consistently using a standard Levenberg–Marquardt algorithm, with the peak centres, widths and heights allowed to vary freely. The DMPS distribution is bimodal, with the Aitken mode related to the  $I^+$  peak in the AMS data and the accumulation mode related to the sulphate and organic modes. The ratio of the fitted Aitken mode diameters yields a particle effective density of  $1.77 \pm 0.23 \text{ g cm}^{-3}$ , which is typical of an inorganic aerosol (Cross et al., 2007). Given that this quantity is a product of the material density and the Jayne shape factor (DeCarlo et al., 2004; Jayne et al., 2000), this may be an underestimate of the material density if the particles are nonspherical, as has been suggested by electron microscopy of laboratory-generated particles (McFiggans et al., 2004).

The shift in composition of these particles is also reflected in the HTDMA data, which shows that during this period, the growth factor of 50 nm dry particles at 90 % RH is 1.34, whereas for the rest of the cruise, values were always greater than 1.5 (Fig. S1.4). The low growth factor and the density estimate are consistent with a dense but low-hygroscopicity substance such as iodine oxide ( $I_2O_5$  has a material density of  $5 \text{ g cm}^{-3}$ ) making up a significant portion of the particulate volume. This is different to what would be expected of organic matter, which tends to be of a low density and low hygroscopicity, and inorganic salts and sulphuric acid, which are high density and high hygroscopicity.

While the data discussed above provides strong evidence for the presence of iodine in the particles during these events, it does not prove that iodine was responsible for the initial NPF, which will have occurred upwind prior to measurement. However, given the rapidity of the iodine oxidation process and the very low volatility of the products (McFiggans et al., 2010; Whitehead et al., 2009; Lehtipalo et al., 2010), it is reasonable to assume that the presence of iodine-based secondary particulate implies that iodine-

initiated NPF was also occurring. It is also worth noting that sulphate concentrations were low during the main case study, so this NPF event did not occur during a period of particularly strong sulphuric acid production.

The initial source of the iodine responsible for these events is not known. Macroalgae have been identified as an iodine source in midlatitude coastal studies (Küpper et al., 2008; Huang et al., 2013) and macroalgae beds containing kelps and wracks also occur on the northeast coast of Greenland (Borum et al., 2002), albeit with a different species composition. Elevated  $\text{CH}_2\text{Br}_2$  levels would be consistent with a macroalgal source (e.g. Laternus, 1996). However, the biomass density of the north-east Greenland kelp beds may be considerably less than found at temperate locations such as Galway Bay (Werner and Kraan, 2004), and ice scouring may reduce macroalgal density in shallower waters where the algae are more likely to be exposed to the atmosphere (Wiencke and Amsler, 2012; Borum et al., 2002).

A source of iodine from the ice sheets themselves is plausible and consistent with the findings of Atkinson et al. (2012). A potential biogenic source are microalgae such as ice diatoms, which have previously been shown to be a potential source of HOI and  $\text{I}_2$  to the Arctic atmosphere (Hill and Manley, 2009). The presence of elevated levels of  $\text{CH}_2\text{Br}_2$  in air compared to levels in seawater during the iodine particle event (Fig. 1c) is also consistent with this suggestion, as polar diatoms are known to be a strong source of bromocarbons (Sturges et al., 1992, 1993). Alternative abiotic suggestions for the source of iodine are chemical production from the ice surface promoted by the freezing of sea salt in the presence of nitrite ions (O' Driscoll et al., 2006) or high levels of iodide sometimes associated with the sea ice region (Chance et al., 2010) and oxidation to  $\text{I}_2$  and HOI on the sea surface (Carpenter et al., 2013). There are also sources of atmospheric iodine in the form of iodocarbons, but we do not consider these likely to be responsible for the observations, as it has been shown that these have a much lower NPF potential compared to  $\text{I}_2$  (McFiggans et al., 2004). It should be noted that no events were recorded near the ice margin to the northeast of Svalbard during the

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latter stages of the cruise, so it may be that this phenomenon is restricted to coastal areas or certain stages of the ice melt process.

## 5 Conclusions

Herein we show observations of new particle formation (NPF) over the Greenland Sea in summer. A persistent event, associated with air originating over the breaking sea ice off Greenland, featured NPF and particles growing to sizes in excess of 50 nm. During this period, iodine was unambiguously detected by an Aerodyne Aerosol Mass Spectrometer and measurements of hygroscopicity and effective density were consistent with iodine oxide comprising a significant portion of the particulate volume. This strongly implies that iodine had a role in the initial NPF events, which is a phenomenon previously associated with coastal locations at lower latitudes (Huang et al., 2013; McFiggans et al., 2004, 2010). The initial source of the iodine in this case is unlikely to be the macroalgae identified during previous studies, but could be speculatively related to other macroalgae species or microalgae associated with the sea ice, which would be consistent with the findings of Atkinson et al. (2012) based on measurements in Antarctica.

These results show that correct prediction of Arctic aerosol number concentrations requires knowledge of iodine processes in new particle nucleation and growth. Our observations suggest that the source of iodine is related to processes associated with coastal sea ice, so this could represent a potentially significant source of particles during periods of ice loss and thus a potential climate feedback mechanism. As yet we have insufficient data to predict how widespread these processes are, but if this phenomenon is limited to coastal areas, it would not explain the events above 80° N studied by Karl et al. (2012). More work is required to identify the initial source of the iodine and the exact mechanisms for iodine NPF at a molecular level (Kulmala et al., 2013).

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## Data Availability

Processed data are archived at the British Atmospheric Data Centre ACCACIA archive. Raw data available on request.

**The Supplement related to this article is available online at**  
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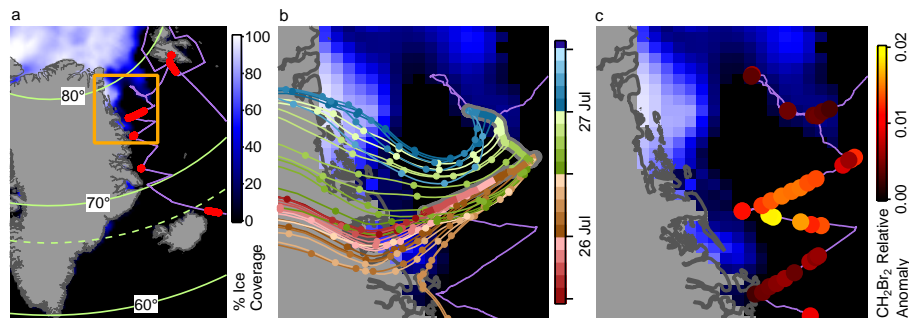
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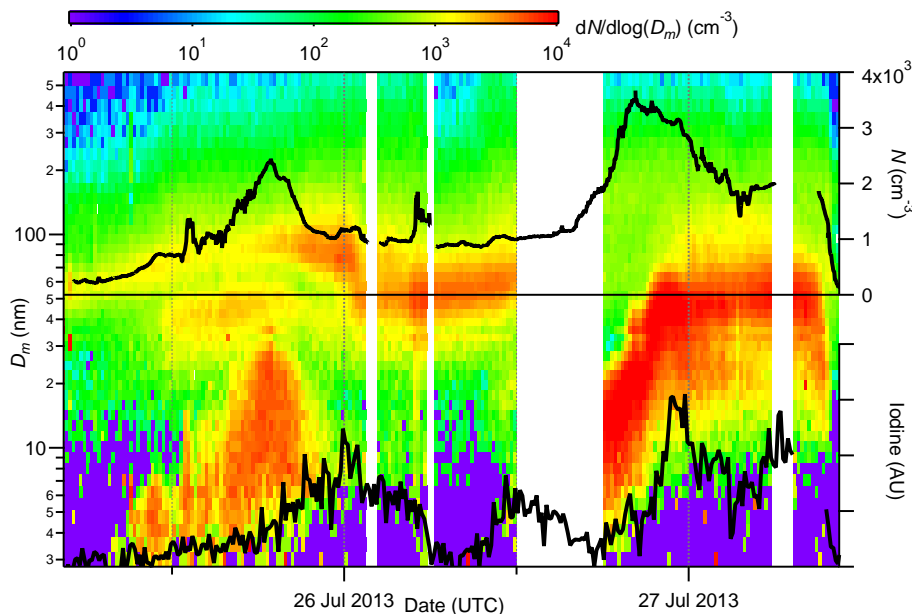


**Figure 1.** (a) Cruise track (purple), ice coverage and locations of NPF events (red), defined by the presence of a mode of particles smaller than 10 nm not attributable to combustion or ship emissions (parts of the cruise outside of the area depicted did not show evidence of NPF). (b) HYSPLIT back trajectories from cruise track corresponding to the 25–27 July case study. Markers are at 6 hourly intervals. (c) Ratio of atmospheric concentrations of  $\text{CH}_2\text{Br}_2$  (a product of ice diatom activity) to its saturation levels in seawater.

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**Figure 2.** Size-resolved number concentrations ( $dN/d\log(D_m)$ ) from a differential mobility particle sizer (DMPS) as a function of electrical mobility diameter ( $D_m$ ), total number concentrations ( $N$ ) from a condensation particle counter (CPC) and uncalibrated iodine ion concentrations from an Aerosol Mass Spectrometer (AMS) based on the signal at  $m/z = 127$  ( $I^+$ ) during the main case study. White areas in the DMPS data show periods of ship influence or when the instrument was not sampling ambient air.

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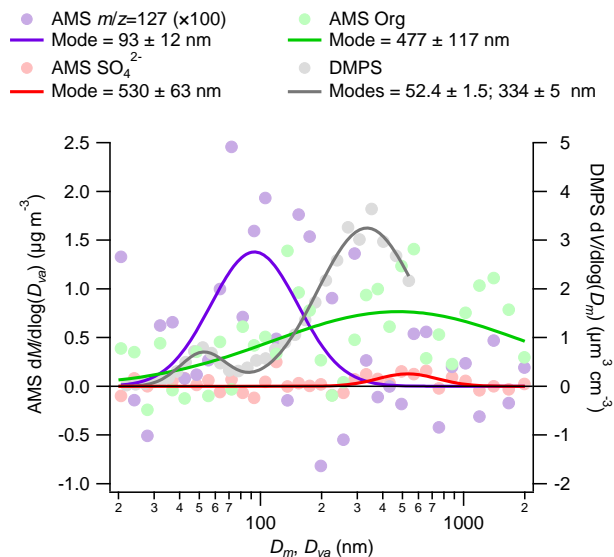
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**Figure 3.** Size-resolved volume ( $V$ ) and mass ( $M$ ) concentrations from the DMPS and AMS respectively, against electrical mobility ( $D_m$ ) and vacuum aerodynamic ( $D_{va}$ ) diameters, during the period of highest iodine loading (26 July 22:50–23:45 UTC) with lognormal least squares fits and associated standard errors. AMS mass concentrations are reported at  $m/z = 127$  ( $I^+$ ), sulphate and organic matter. The low signal-to-noise ratios of the AMS data are due to the low concentrations and short averaging time. Note that the widths of the AMS distributions should not be directly compared against the DMPS, as the AMS distributions are subject to broadening introduced by the chopper wheel and variations in particle density.