Title: SOCRATES and CAPRICORN-2 Single Particle INP Composition via STEM/EDS

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1.0 Data Set Overview:

This data file contains the size, type and fraction of all aerosol particle types acting as ice nucleating particles (INPs) that were collected following processing (see below) in the Colorado State University Continuous Flow Diffusion Chambers (CFDCs) during NSF-supported ship and aircraft-based measurements over the Southern Ocean (SO) in 2018. Aircraft samples were processed and collected during real-time sampling at different flight levels on the National Science Foundation/National Center for Atmospheric Research Gulfstream-V aircraft during the Southern Ocean Clouds, Radiation, Aerosol Transport Experimental Study (SOCRATES). Ship samples were processed and collected during real-time sampling on the Australian Marine National Facility's RV Investigator that was deployed over the SO region from Hobart, Tasmania to ~65°S latitude during the SOCRATES flight period in January and February, 2018 in the related Clouds, Aerosols, Precipitation, Radiation and atmospheric Composition Over the southeRN ocean (CAPRICORN-2) campaign. Activated ice crystal residual particles were analyzed via Scanning Transmission Electron Microscopy Energy-dispersive X-ray Spectroscopy (STEM/EDS). These data are further presented and discussed in *Twohy et al.* [2021].

2.0 Instrument Description:

Aerosol particles were sampled via ambient inlet systems on the G-V (HIAPER modular inlet) and the RV Investigator [Humphries et al., 2019]. Some sampling on the G-V also included sampling of cloud particle residuals using a Counterflow Virtual Impactor (CVI) inlet [*Noone et al.*, 1988; *Twohy et al.*, 2003]. A mix of ambient aerosol and cloud sampling for INP activation and analyses from the CSU CFDCs was necessitated on the G-V by the nature of the mission for characterizing both aerosol and cloud properties, and the statistical limitations of sampling and accumulating low concentrations of INPs. Consequently, although INPs collected on a given flight may have been dominated by sampling within the marine boundary layer or clouds topping that layer, listed altitudes of sampling may include the full tropospheric range spanned by the G-V on a given flight. Lowest flight levels were typically 150 m and transits south from Hobart, Tasmania were typically done near 6 km MSL. The CFDC instruments process sampled particles via confining them and exposing them to a nearly steady-state temperature and relative humidity [*DeMott et al.*, 2017]. INPs are detected through freezing and growth of ice crystals during the residence time in the CFDC, followed by evaporation of liquid water and optical detection of ice crystals as a distinct mode of the size distribution at approximately >4 microns. Due to this optical detection scheme, a pre-impactor is used with the CFDC to limit large particles (also pre-dried with diffusion driers) from entering the instrument and confounding ice crystal versus aerosol detection. On the G-V, a two-stage pre-impactor with a 50% cut size of 2.5 μm aerodynamic diameter was employed. On the RV *Investigator* in CAPRICORN-2, a 1.5 μm pre-impactor was used due to concerns about drying larger sea spray particles in the atmospheric boundary layer close to the ocean surface. The ice crystal impactors used at the base of the CFDCs had identical single-stage 4 μm cut sizes, intended to capture only activated ice crystals. Ice crystals were impacted onto 3-mm carbon-formvar-coated electron microscope grids and silicon nitride membrane windows (indicated by "w" in data file). Impactor substrates containing dried INP residues were stored cold in clean containers immediately after flights and held for off-line analysis. The single-particle elemental composition of selected samples was measured on a JEOL JEM-2100F 200 kV scanning transmission electron microscope with Oxford Max 80 Energy Dispersive Spectroscopy (EDS) system at the Colorado State University Central Instrument Facility, using a low background Beryllium sample holder.

Six samples of INPs collected behind the CFDC on the ship cruise and four samples from the G-V aircraft were analyzed. The sizes of collected INPs were measured on the substrates (as imaged from the electron microscope), and ranged from about 0.1 to 1.5 microns diameter, the largest size accepted due to the sampling configuration as discussed in Section 3 below. The CFDC processing temperature ranged from 241K to 246K. Because the numbers of ice nucleating particles found and analyzed were low (n=87), the compositions of all INP samples were averaged together for analyses presented in Twohy et al. [2021]. For the same reason, this archive file includes both ship and aircraft samples.

3.0 Data Collection and Processing:

Blank grids were first analyzed to assess any particle types that might be considered artifacts and if present, these were not included in the final data set. Particles were separated into categories based on spatial mapping of their detected elements and morphology as given in Table 1, Supplemental Information.

Table 1. STEM-EDS Particle Classification Scheme

Category	*Characteristic signature*
Crustal dust	Silicates (rich in Si, variable Na, Mg, Al, K, Ca, and Fe), carbonates (Mg and/or Ca with large C peak), phosphates (rare), sometimes mixed with C, usually with irregular edges
Sulfur-based	Round shape, primarily S, O, may be volatile under the electron beam. (Sulfuric acid, ammonium sulfate/bisulfate or MSA)
Carbonaceous	*Organic:* C above background and may contain O, N, S, K, Na also
	Soot: C only plus characteristic chain aggregate morphology
Metals	Metals such as Al, Fe, Cr, Ti, Mn, Co, Zn, Cu, sometimes mixed with C
Biomass Burning	Amorphous mixture of K, S, may also contain detectable C
Sea-spray Na	NaCl with minor Mg, S, K, Ca, may also contain detectable C. Usually with crystalline structure
Sea-spray high S	Primarily Na with S>Mg by atomic weight, usually with K, Ca. May or may not contain detectable Cl and C
Other Salt	K, Ca, or Mg with minor Na relative to sea-spray, with Cl or S

Table 1 Notes. Chemical and morphological features used to identify different types of particles via scanning transmission electron microscopy and energy-dispersive X-ray analysis.

In order to correct acquired INP sizes and numbers to those present in the ambient atmosphere, In order to and to reject particles that were considered to have impacted the grid or window at aerosol diameters that exceeded those that might have existed as “wet” aerosols rather than ice, additional analyses were required. A number versus size correction was needed for all ship CFDC INPs that were collected while operating an aerosol particle concentrator (used for enhancing INP counting statistics) that possesses a size-dependent concentration factor [e.g., Tobo et al., 2013, but recalibrated prior to this study]. This resulted in the computation of a weighting factor as a function of INP size for all INPs collected when using the concentrator, reducing numbers of larger INPs that are enhanced by 150 times in the aerodynamic diameter range > 0.8 μm relative to the numbers of INPs in the aerodynamic diameter range below 0.5 μm that are enriched by less than 10 times. In correcting data from sampling via the aerosol concentrator, account was necessary for the fact that particles entered the concentrator at the ambient relative humidity in the ship sampling stack. For this purpose, the water uptake properties of sea salt were assumed for all particles. Another correction was required for both ship and aircraft collections to account for the size-dependent transmission efficiency curves of the CFDC pre-impactors, determined based on separate laboratory calibrations. Finally, if an apparent INP was detected dry on grids at a size exceeding 1.33 μm, these were omitted from analyses as being ambiguous as INPs because they were at sizes that would have potentially been captured by the ice crystal impactor at their wet aerodynamic diameters. Once data were assembled for all 87 INPS, each particle was assigned a corrected count based on its size-dependent sampling efficiency. The corrected count represents that particle's effective frequency relative to the sampled INP population. These corrected counts were then divided by the total counts to obtain an effective number fraction for that size and type of particle, relative to the sampled population.

4.0 Data Format:

The file format is csv. The data file is titled SingleINPComposition_SOCRATES_CAPRICORN2.csv. The column variables include:

RF or Ship ID: flight number as used by the NCAR flight facility in SOCRATES, e.g., RF11 for Research Flight 11, or Ship sample ID, e.g., Inv01 is sample 1 from the voyage. A “w” indicator refers to silicon nitride window collections rather than standard TEM grids.
Date start: start date in Day-Month-Year format for which sample was collected in UTC
Date end: end date in Day-Month-Year format for which sample was collected in UTC
Time start: Start time of collection in hours and minutes UTC
Time end: End time of collection in hours and minutes UTC
Altitude: altitude in m MSL over which collection was made
Diam: approximate dry diameter in microns, inferred from average of maximum and minimum linear sizes of particles in analyzed TEM images
Type: Classified particle type (Table 1)
Frac: Fraction of corrected particle counts of all analyzed particles represented by each particle, based on the size-dependent sampling efficiencies. These fractions define an INP size distribution and its composition for all INP analyzed from ship and aircraft during SOCRATES

5.0 Data Remarks:

Sodium-based sea spray includes both particles comprised of mostly NaCl (with other inorganic and organic sea-salt components), as well as those in the sea-spray “high S” category, which also have sodium but are enriched in sulfur and depleted in chlorine due to uptake and condensation of sulfur gases [*McInnes et al.*, 1994]. Organic coatings were commonly detected on sea-spray particles [*Twohy et al.*, 2021]. Sulfur-based particles are also likely to have some organic components that cannot be detected by STEM/EDS above the substrate background.

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