

Title: Single Particle Composition via STEM/EDS

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1.0 Data Set Overview:

These data files contain the percent by number of aerosol particle types collected at different flight levels on the Gulfstream-V aircraft and analyzed via Scanning Transmission Electron Microscopy Energy-dispersive X-ray Spectroscopy (STEM/EDS) during SOCRATES. Samples were collected via impaction below cloud at approximately 150 m, in cloud after droplet evaporation, and above cloud in two dry size ranges as described below. Data were analyzed from only a selected number of flights. Samples in drizzle or in precipitating clouds were excluded. Particles in the smaller size range were only analyzed for later flights when they were collected with heaters turned off after they were found to be partially volatile in the heated inlet and sample line. The data are further presented and discussed in *Twohy et al.* [2020].

2.0 Instrument Description:

Aerosol particle samples were collected below and above clouds, as well as in clouds using a Counterflow Virtual Impactor (CVI) inlet [Noone *et al.*, 1988; Twohy *et al.*, 2003]. The CVI inlet and porous tube were composed of titanium and the sample lines were stainless steel. The CVI inlet was heated to ~50°C during cloud sampling in order to evaporate cloud droplets larger than 7-8 microns diameter, and the sample stream was maintained at 40°C to prevent condensation prior to measuring the associated water vapor with a tunable diode laser hygrometer. Ambient aerosol particles were collected above and below cloud by periodically turning off the CVI counterflow airstream and, for later flights, turning off the heaters to minimize losses of volatile species during sample collection. Particles were collected with a two-stage impactor onto carbon-coated electron microscope grids and silicon nitride membrane windows in two dry diameter ranges of about 0.1-0.5 μm and 0.5-5 μm (these physical diameters assume particle densities of 2 g cm⁻³ at 1000 mb). Impactor substrates were stored immediately after flights and held for off-line analysis below 0°C. The 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 Colorado State University, using a low background Beryllium sample holder.

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. For each sample, the grid or window was scanned for representative sampling areas where particles were sufficiently far from the jet center to avoid

overlap, but heavily loaded enough to minimize any potential artifacts. Fifty or more particles per sample in the smaller size range were imaged and analyzed for elemental composition, while for more sparsely loaded samples in the large size range, fewer particles were usually analyzed. For samples with particles visibly volatile under the beam, groups of particles were analyzed together to minimize energy input per unit area per time so major elements could still be detected. Particles were separated into categories based on spatial mapping of their detected elements and morphology as given in Table 1. In-cloud composition percentages have been corrected for particles volatile in the CVI inlet and sample lines (and for droplets below the CVI cut size) by comparing the CVI residual number concentration from the UHSAS to the cloud droplet number concentration measured by the CDP as described in *Twohy et al.* [2020], Supplemental Information.

Table 1. STEM-EDS Particle Classification Scheme

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

4.0 Data Format:

There are two short csv data files, SingleParticleCompositionSmall.csv for the 0.1-0.5 micron diameter particles and SingleParticleCompositionLarge.csv for the >0.5 micron diameter particles. The column variables include:

Flight: flight number as used by the NCAR flight facility in SOCRATES, e.g., RF11 for Research Flight 11

Date: date in Day-Month-Year format for which sample was collected in UTC (not necessarily start date of flight)

Sample: sample number as used by CVI group in SOCRATES

Diameter: approximate dry diameter in microns, with 50% aerodynamic diameters cut size converted to equivalent spherical size assuming a density 2 g cm^{-3} at 1000 mb

Level: There are three levels of samples: approximately 160-180 m (below cloud), in cloud in the marine boundary layer or above cloud at the higher altitudes specified (GPS altitude from the aircraft data system was used, rounded to nearest 10 m). Each altitude range is listed in separate blocks in the file.

Start UTC: start time that samples were collected in HH:MM:YY UTC

End UTC: end time that samples were collected in HH:MM:YY UTC

N: number of particles used in each sample to calculate composition fractions

Remaining nine columns: fractional composition; i.e., 0.375 means 37.5% of the particles analyzed had the composition named in column header (see Table 1).

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 in the >0.5 micron diameter range [Twohy *et al.*, 2020]. Sulfur-based particles, which were almost exclusively in the >0.1-0.5 micron diameter range, are also likely to have some organic components that cannot be detected by STEM/EDS above the substrate background, particularly if they are volatile. However, based on the discussion in Twohy *et al.* [2020], we expect sulfate mass to dominate over MSA and other organic mass in the >0.1-0.5 micron particles, which represent most of the sub-micron particle mass and much of the cloud condensation nuclei number concentration.

6.0 References:

- McInnes, L. M., D. Covert, P. K. Quinn, and M. S. Germani (1994), Measurements of chloride depletion and sulfur enrichment in individual sea-salt particles collected from the remote marine boundary layer, *J. Geophys. Res. Atmos.*, 99(D4), 8257-8268.
- Noone, K. J., J. A. Ogren, J. Heintzenberg, R. J. Charlson, and D. S. Covert (1988), Design and calibration of a Counterflow Virtual Impactor for sampling of atmospheric fog and cloud droplets, *Aerosol Sci. Technol.*, 8(3), 235-244.
- Twohy, C. H., J. W. Strapp, and M. Wendisch (2003), Performance of a counterflow virtual impactor in the NASA Icing Research Tunnel, *J. Atmos. Ocean. Tech.*, 20(6), 781-790.
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