Aerosol composition measured in the NH polar vortex using single particle mass spectrometry

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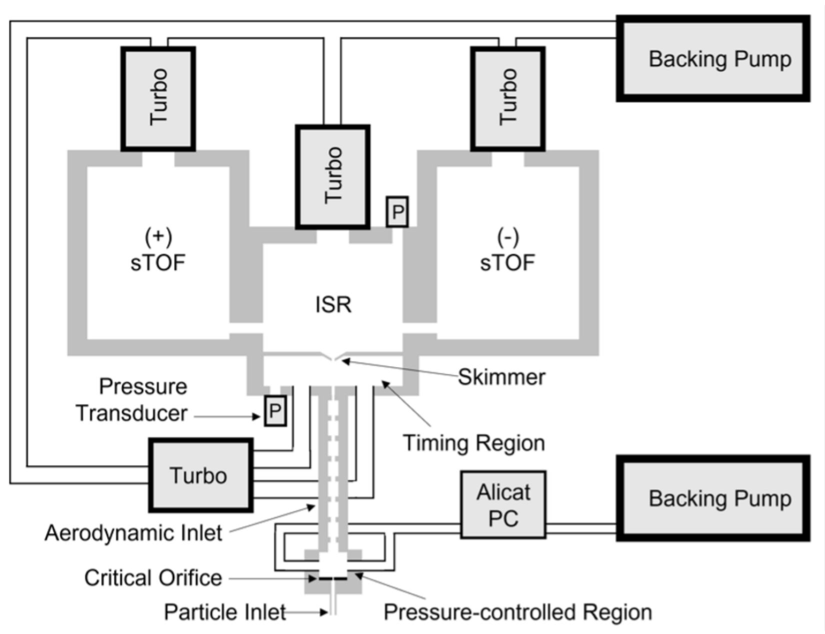
Stratospheric Aerosol: Properties and Processes

The stratospheric aerosol layer is an important component of the Earth's climate system. It scatters incoming solar radiation, a modest fraction of which returns to space, cooling the surface. It also absorbs and re-radiates infrared radiation, causing warming. Most of the mass of stratospheric aerosol is sulfuric acid, which gives stratospheric particles very different chemistry than less acidic tropospheric particles. The size, composition, and number of stratospheric particles determine their effects in the climate system.

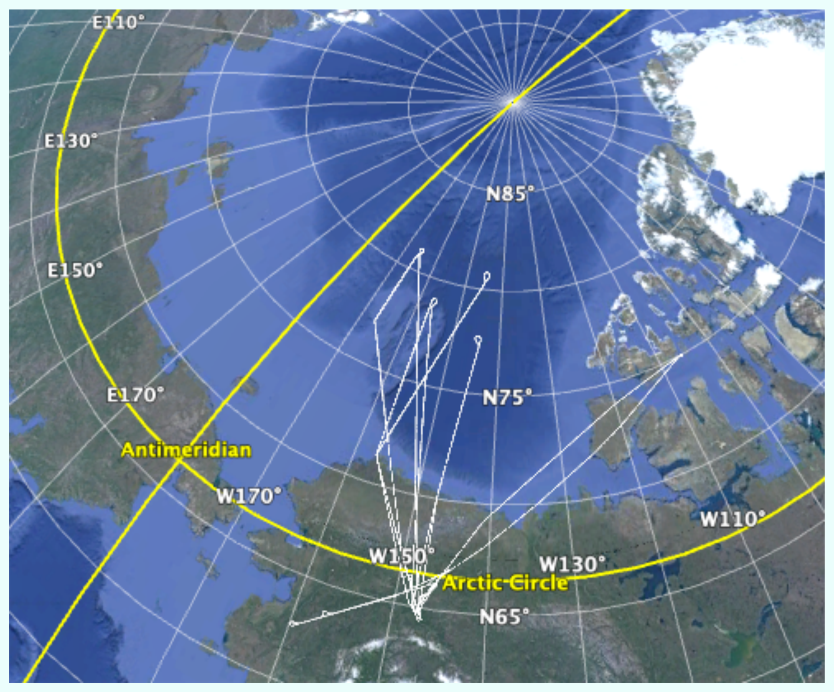
The processes controlling the formation and properties of stratospheric aerosol are not well-understood, making their properties difficult to predict. Several processes are likely important. There is limited transport of aerosol and aerosol precursors over the tropopause. Carbonyl sulfide photolyzes in the upper atmosphere to provide a source of sulfuric acid. Meteoric "smoke" formed from the ablation of meteors contributes metals to the stratospheric aerosol. New particle formation from gas phase precursors may contribute to particle number. Particle coagulation may grow particles.

Our goal is to constrain the relevant processes by measuring the size and composition of stratosphere particles and connecting these to air mass sources and properties.



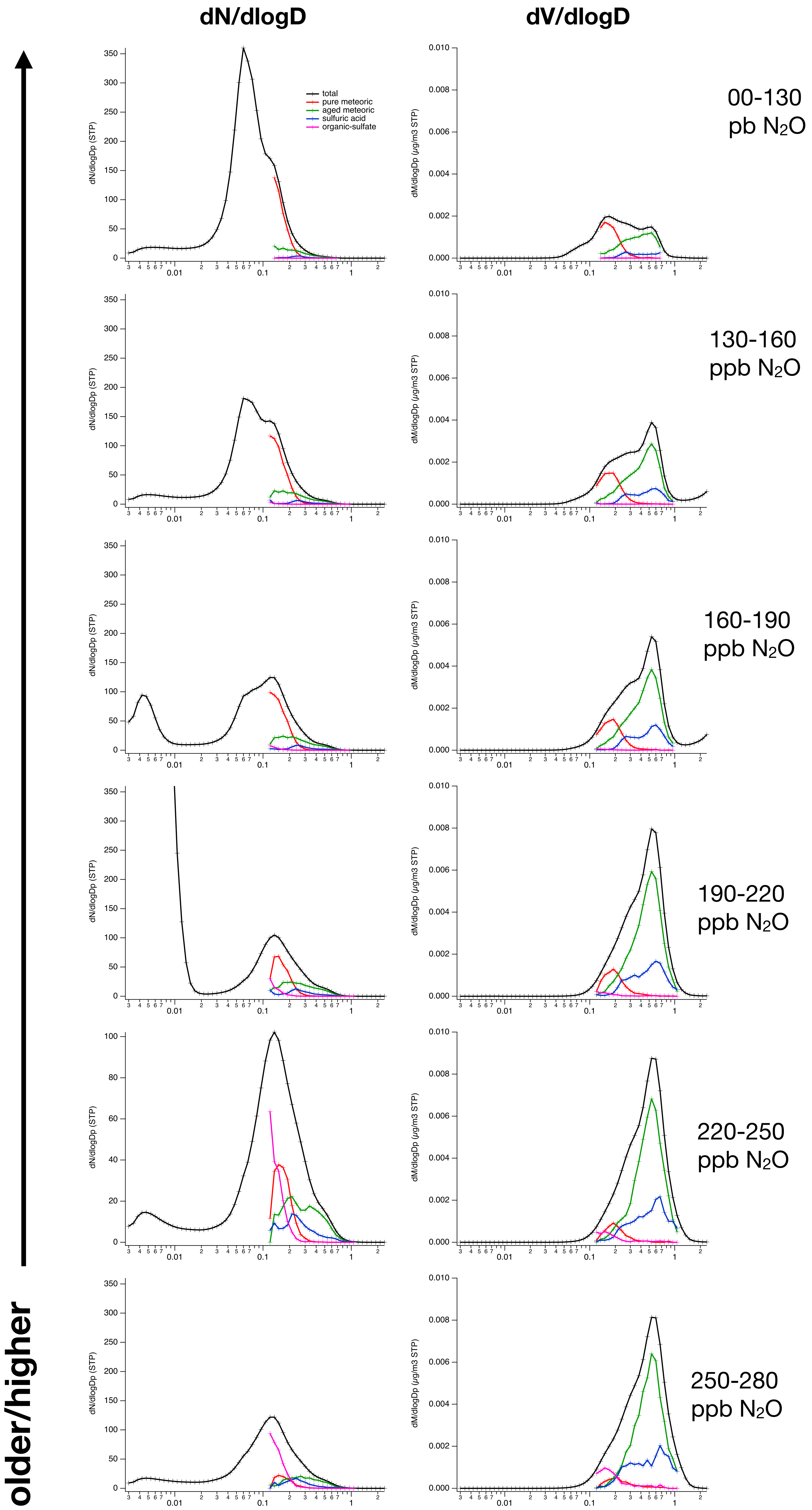
PALMS-NG particle composition

Particles were classified by their composition and sized using the Particle Analysis by Laser Mass Spectrometry-Next Generation (PALMS-NG) instrument. The instrument flies in the nose of the NASA WB-57 aircraft and analyzes individual particles of ~ 0.1-2 μm diameter in situ at a typical rate of 2-4 particles per second. Atmospheric particles are subsampled from a forward-facing duct at the very nose of the aircraft, then enter an aerodynamic inlet to form a particle beam. Particle aerodynamic diameters are measured by the transit time between two continuous wave lasers, the second of which triggers a UV (193 nm) excimer laser pulse which ablates and ionizes the particle. Positive and negative ions are sent to separate time-of-flight mass analyzers. Hit rates for laser triggers are very high, meaning low bias in particle types analyzed. Mass spectra are used to classify the particles into distinct composition types.



Study Area: Polar air over the wintertime Arctic

The particle composition data reported here are from flight measurements made March 13-23, 2023 out of Eielson AFB near Fairbanks, AK. Flight tracks from the period March 13-22, 2023 are displayed in the map above. During this period, the polar vortex was within range of the WB-57 and frequently sampled.



Size distributions of different particle types in different ages of stratospheric air

Five main characteristic particle types were identified and quantified in air in and near the polar vortex during March 13-23, 2023: sulfuric acid (>80% sulfuric acid), meteoric-sulfuric "pure" (~>99% sulfuric acid, with trace meteoric metals and extremely low organic content), meteoric-sulfuric with some organic (may contain a few weight % organic molecules), organic-sulfate (> ~ 50:50 organic:sulfate). At the highest altitudes, in the oldest air (lowest N_2O mixing ratios), almost all the particles contain meteoric metals. High numbers of small meteoric particles containing small amounts of organic material at high altitudes are suggestive of relatively recent formation aloft. The origin of the condensing organics is uncertain.

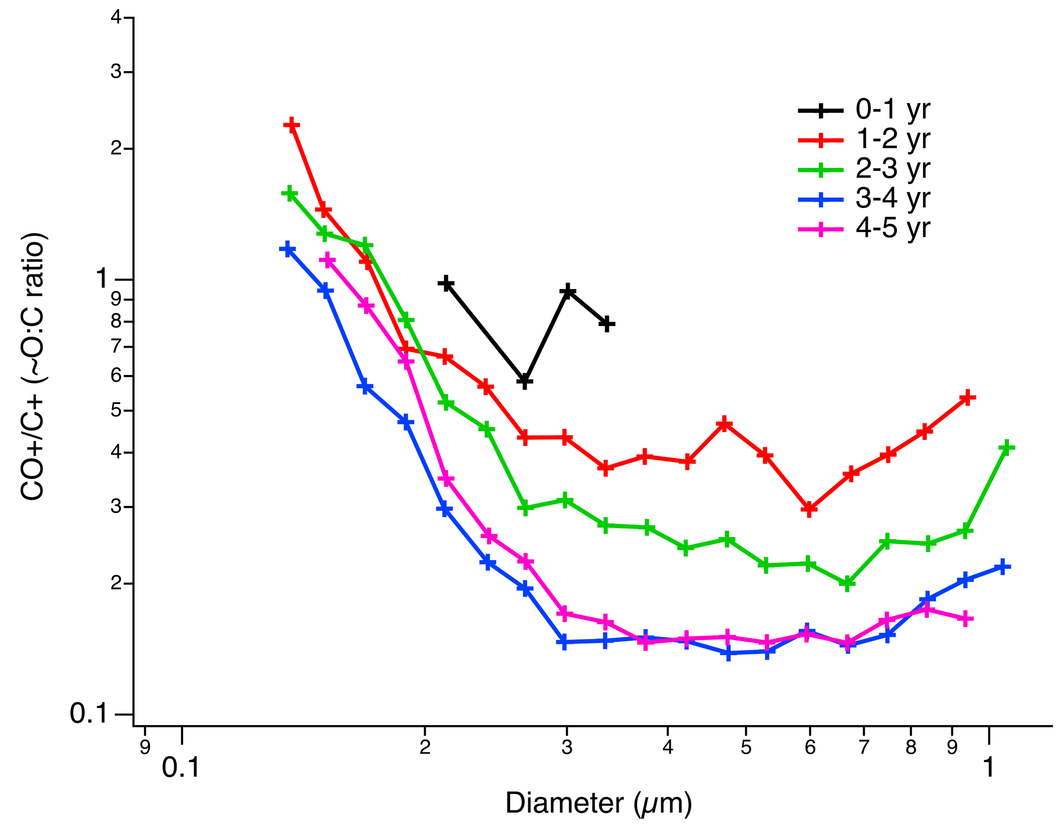
Preliminary Conclusions

1.The vast majority of particles in highly aged polar vortex air contain meteoric metals.

2. Some meteoric-sulfuric particles have extremely low organic mass fractions. These clearly have a high altitude source, consistent with formation in highly processed air that lacks condensable organic vapors.

4. Coagulation with background stratospheric aerosol is a major pathway to convert vortex pure meteoric particles into aged meteoric particles

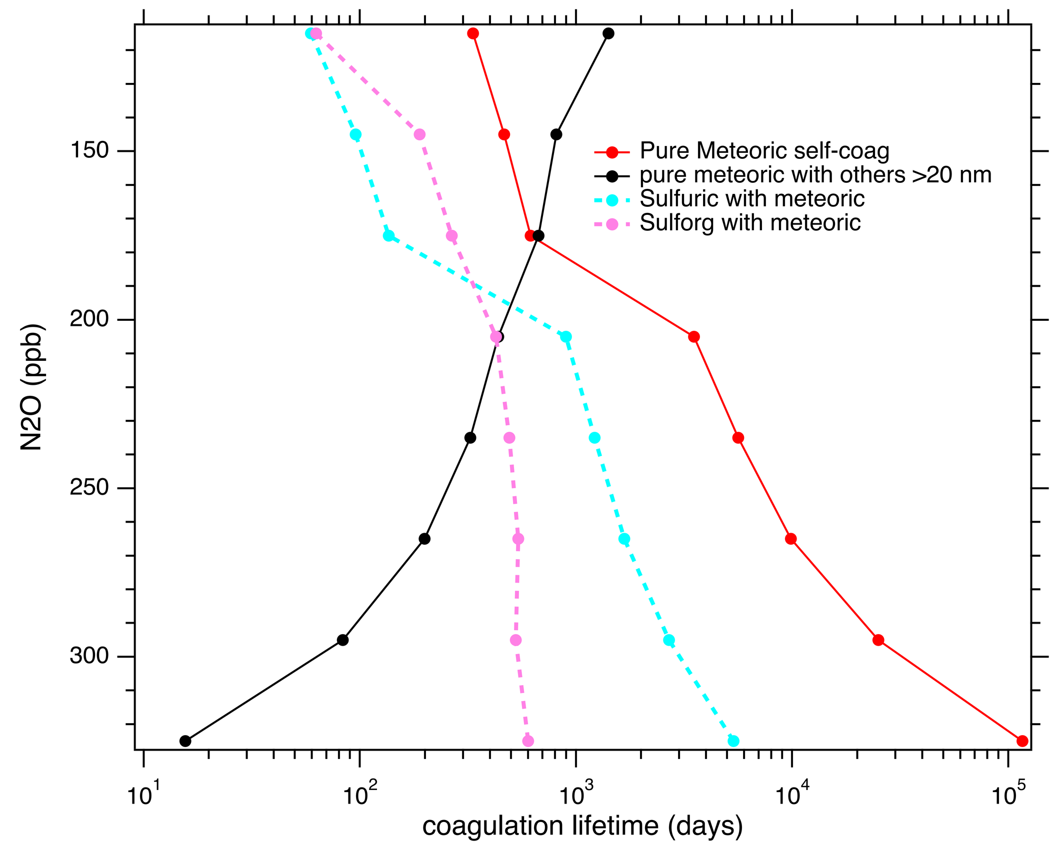
5. Organics in sulfuric acid particles move towards a lower oxidation state over time without apparently losing much carbon,



Oxidation state of stratospheric aerosol

The PALMS ionization process converts most of the mass in organic molecules into much smaller ions. The two main organic ions appearing in the positive mass spectra are CO^+ and C^+. Their ratio gives some indication of the O:C ratio of the molecules that were present in the analyzed particle.

Particles which are predominantly sulfuric acid but contain some organics show a decrease in O:C ratio as the particles age, but not much loss in total carbon. Aging is estimated using the mean age of air determined using SF6 as a tracer. This is consistent with condensation and oligomerization reactions occurring in the highly acidic particles,



Coagulation lifetimes of particles

To understand the fate of meteoric particles formed in the polar vortex, their lifetimes to coagulation were determined with respect to other important particle types (and to self-coagulation). While they persist for a long time in the low density, low particle air in the stratosphere, other particles relatively quickly (timescale of months) take up vortex-sourced particles and turn into "aged" meteoric particles.