Sources, distribution, and acidity, of sulfate-ammonium aerosol in the Arctic in winter-spring

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**Motivation**

**Sulfate** is a dominant component of winter/spring Arctic Haze and has direct and indirect impacts on climate.

**Acidic sulfate** aerosol can be neutralized by ammonium, with implications for atmospheric chemistry and climate. Arctic surface data show ammonium has been decreasing more quickly than sulfate, leading to increasingly acidic aerosol.

The sources of inorganic aerosol to the Arctic are highly uncertain, and observed concentrations can’t be reproduced by models.
Goal: Understand the sources and acidity of aerosol in the Arctic

Approach: Integrated analysis of sulfate-ammonium aerosols from:

1. **ARCTAS (NASA) and ARCPAC (NOAA)** aircraft campaigns during April 2008, based in Fairbanks, AK
2. **Long-term monitoring sites** in Alaska, Canada, Spitsbergen

3. **GEOS-Chem v8-02-03** full chemistry, GEOS-5 meteorology, 2°x2.5° horizontal resolution
   - **SO₂ emissions:**
     - EDGAR anthropogenic with regional overwrites (map)
     - FLAMBE biomass burning with corrections (Fisher et al. 2010)
     - AEROCOM volcanism
   - **NH₃ emissions:**
     - Bouwman et al., 1997 anthropogenic (with overwrites) & natural
     - FLAMBE biomass burning with corrections (Fisher et al. 2010)

**Additions to standard v8-02-03 model**
- Improved cold temp. wet scavenging (Q. Wang)
- Imposed aerosol dry deposition velocity over ice
- Imposed NH₃ seasonality over Asia
A diversity of sources contributed to aerosol in the North American Arctic during ARCTAS (April 2008)

Fossil fuel regions
- N. American Arctic
- Eurasian Arctic

Other sources:
- Ships
- Biomass burning
- DMS oxidation
- Volcanic emission
- Natural NH₃
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- E. Asia and Europe are major sources of sulfate and ammonium at all altitudes
- Below 2 km, E. Asian, European, and N. American sources have comparable influences on sulfate
- Natural sulfate sources (DMS, volcanic) are important at all altitudes
- Russian fires are an important source of ammonium above 2 km (in 2008)

**Sulfate**
- 5-10 km: E. Asia, Europe, N. America
- 2-5 km: West Asia/Siberia, N. America, Europe, E. Asia
- 0-2 km: West Asia/Siberia, N. America, Europe, E. Asia, Observations

**Ammonium**
- 5-10 km: E. Asia, Europe, N. America
- 2-5 km: West Asia/Siberia, N. America, Europe, E. Asia
- 0-2 km: West Asia/Siberia, N. America, Europe, E. Asia, Observations
Arctic spring aerosol during ARCTAS/ARCPAC ranged from very acidic to fully neutralized.
Different source regions show different neutralization signatures in the Arctic

- Aerosol from Europe and East Asia is mostly neutralized

- Aerosol from West Asia / Siberia and North America is significantly more acidic

- Free tropospheric aerosol will likely become more neutralized as SO\textsubscript{x} emission controls are implemented in East Asia

- However, opposite trend is occurring in the boundary layer, at least at Barrow...
Surface data highlight importance of West Asian/Siberian emissions

- West Asian emissions drive winter aerosol burdens in High Arctic
- Increasing aerosol acidity at Barrow likely due to energy production & growth in Siberia/Kazakhstan

Sulfate

Ammonium

Barrow (71.3°N, 156.6°W)

Denali (63.7°N, 149.0°W)