Arctic pollution sources and transport: Results from the ARCTAS aircraft mission and beyond

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With support from the NASA Tropospheric Chemistry Program
“Arctic Haze” - a long-standing air pollution problem

Discovered in the 1950s by pilots flying over the Arctic

Observed every winter and spring since the 1950s

Transported pollutants include aerosols, ozone, mercury, ...

Haze layers: Vertically thin (10m-1km)
Horizontally extensive (20-200 km)

Photo by Cam McNaughton
Arctic pollution has direct and indirect impacts on climate
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There is an urgent need to understand pollution in the Arctic atmosphere

Quinn et al., 2008
NASA/ARCTAS campaign designed to address this need

**ARCTAS:** Arctic Research of the Composition of the Troposphere from Aircraft and Satellites
- Spring/summer campaign in 2008
- Comprehensive in situ aircraft measurements
- Maximize value of satellite and surface data
- Improve models of atmospheric composition

**Along with:**

**Satellite Teams:** AIRS, CALIPSO, MISR, MOPITT, MODIS, OMI, TES

**Ground Teams:** ARC-IONS, NATIVE, UAF

**Model Forecasts/Analyses:** GEOS-5, GEOS-Chem, GOCART, LaRC, MOZART, STEM

**International Polar Year in-field collaborations:**
ARCPAC (NOAA), ISDAC (DOE), Pre-HIPPO (NSF), GRACE (Germany) POLARCAT-FRANCE (France)
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ARCTAS-A: 1-21 April; based in Fairbanks, Alaska and Thule, Greenland

MODIS fire counts, April 2008

Jacob et al., 2010
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*Jacob et al., 2010*

*Fisher et al., 2010*
ARCTAS observations complemented by other data sets

**Aircraft**
- High-resolution
- Vertical information
- Multiple species
- Infrequent campaigns
- Snapshot in time

**Surface**
- Long-term records
- Intensely studied
- Sparse coverage
- No vertical information

**Satellite**
- Continuous data
- Spatially dense
- Decadal-scale
- Limited sensitivity
- Many uncertainties
Models provide links between disparate data sets

- Aircraft
- Surface
- Satellite

Pollution source-receptor relationships
Inverse emissions estimates
Detailed chemical processing
GEOS-Chem chemical transport model links datasets

The GEOS-Chem chemical transport model (CTM):

- Global, gridded, 3-D model
- Simulates emissions, chemistry, transport, and deposition
- Driven by GEOS-5 meteorology with 0.5°x0.67° horizontal resolution, regridded to 2°x2.5° resolution
- Represent our best *a priori* understanding of processes
- Applied here to carbon monoxide and aerosols

Emissions:

- Anthropogenic
  Boumwan et al. 1997 for NH₃
  EDGAR with regional overwrites for CO, SO₂
- Biomass burning (FLAMBE)
- Volcanic (AEROCOM)
- Other natural sources
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Rahn and McCaffrey, 1980; Barrie, 1986
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Recent modeling studies disagree, suggest varying importance for Europe, Russia, East Asia

**ARCTAS Science Questions:**
- What are the transport pathways for different pollutants?
- What are the contributions from different source regions?
Carbon monoxide (CO) as a pollution tracer

**Source:** incomplete combustion (fossil fuel & biomass burning)

*Fisher et al., 2010*
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Model emissions adjusted to fit ARCTAS & ARCPAC data: ↑ anthropogenic, ↓ fires
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**Model source attribution:**
- Asian anthropogenic source dominates Arctic CO background
- European source important near surface
- Fires contribute to variability

Fisher et al., 2010
Combined data demonstrates ability of AIRS satellite instrument to see mid-tropospheric transport events

**AIRS Instrument**
- on polar-orbiting Aqua satellite
- measures CO in thermal IR
- cloud-clearing capabilities
- ~70% daily global coverage
- sensitive to mid-troposphere

\[
\varepsilon_{\lambda}(T_1) I_{\lambda}(T_0)
\]

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\[ \varepsilon_{\lambda}(T_1) \]

Absorbing Gas

\[ I_{\lambda}(T_0) \]

Surface

Fisher et al., 2010
AIRS 6-year record provides context for 2008 results

- Weakened Aleutian Low in spring 2008 decreased transport to Alaska
- Implies stronger Asian fossil fuel influence in El Niño years

**Fisher et al., 2010**
Sulfate is a dominant component of winter/spring Arctic aerosol pollution.
Sources and acidity of Arctic aerosol remain highly uncertain

Barrow, Alaska
Fraction of aerosol mass

Quinn et al., 2002 (adapted)

Sulfate is a dominant component of winter/spring Arctic aerosol pollution

But models can’t reproduce observed Arctic sulfate

In recent multi-model CTM intercomparison, models varied by factor of 1000!
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At Barrow ammonium has been decreasing more quickly than sulfate, leading to increasingly acidic aerosol

Opposite is true at Alert, suggesting different sources
Sources and chemistry of atmospheric sulfate

- Volcanoes
- Industry
- Phytoplankton
- Ocean
- Agriculture
- Fires

SO$_2$ + OH $\rightarrow$ H$_2$O$_2$, O$_3$

DMS $\rightarrow$ OH

H$^+$, SO$_4^{2-}$

Acidic
Sources and chemistry of atmospheric sulfate

Volcanoes, Industry, Phytoplankton, Ocean, Agriculture, Fires

SO₂, DMS, OH, H₂O₂, O₃

Acidic: H⁺, SO₄²⁻
Neutralized: NH₄⁺, SO₄²⁻
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The extent to which sulfate is neutralized by ammonium has implications for both atmospheric chemistry and climate.
Wet deposition data test GEOS-Chem simulation over source continents

April 2008

SULFATE

AMMONIUM

NADP

EMEP

EANET

Fisher et al., submitted
A diversity of sources contribute to aerosol in the North American Arctic in spring

Other sources: Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH₃
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Other sources:
- Ships
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- DMS oxidation
- Volcanic emission
- Natural NH$_3$

- 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
- Biomass burning is an important ammonium source in the troposphere
Surface data highlight importance of West Asian/Siberian emissions

Economic growth in Kazakhstan and increased energy production in Siberia may lead to increased Arctic aerosol in winter.
Spring aerosol ranged from very acidic to fully neutralized.
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Different source regions show different neutralization signatures in the Arctic

- Simulation of April aircraft data shows the most neutralized aerosol originates from Europe and East Asia.
- Aerosol from West Asia & Siberia is significantly more acidic.
- Future short-term changes in neutralization of Arctic aerosol will depend on relative emissions growth, especially in East Asia & West Asia.
- Long-term, free tropospheric aerosol will likely become more neutralized as SOₓ emission controls are implemented in East Asia but NH₃ emissions continue to grow.
April aerosol neutralization shows a vertical gradient in April

Most acidic air at surface: influence from West Asia, Norilsk/Kola Peninsula, Prudhoe Bay

More neutralized air in free troposphere: influence from East Asia, biomass burning
In winter, the free troposphere is much more acidic

Very low aerosol concentrations in winter, mainly from natural sources

Extremely limited NH$_4^+$: major inputs from East Asia, biomass burning don’t occur until spring
Take-home messages

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Future changes in global emissions distributions will drive changes in both the composition and the climate impacts of Arctic aerosol.