Atmospheric pollution in the Arctic: Sources, transport, and chemical processing

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“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, ...

Photo by Cam McNaughton
Arctic pollution has direct and indirect impacts on climate

Quinn et al., 2008
Arctic pollution has direct and indirect impacts on climate

There is an urgent need to understand pollution in the Arctic atmosphere

Quinn et al., 2008
Previous understanding of Arctic pollution sources

Surface measurements in 1970s-80s suggested most sources in Europe & Soviet Union

Rahn and McCaffrey, 1980; Barrie, 1986
Previous understanding of Arctic pollution sources

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Recent modeling studies disagree qualitatively and quantitatively, suggest varying importance for Europe, Russia, East Asia

*Klonecki et al., 2003; Stohl, 2006; Shindell et al. 2008*
Previous understanding of Arctic pollution sources

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My dissertation questions:

• What are the contributions and transport pathways of different sources of Arctic pollution?
• How do these contributions vary as a function of altitude and species?
• What drives seasonality, interannual variability and decadal-scale trends of Arctic pollutants?
Aircraft data provide new look at Arctic troposphere

NASA/ARCTAS-A and NOAA/ARCPAC
- International Polar Year field campaigns
- Based in Fairbanks, Alaska in April 2008
- Comprehensive in-situ platforms
- 100+ scientists in the field including flight, satellite, ground, and model teams
- International collaborations with DOE (ISDAC), NSF (Pre-HIPPO), Germany (GRACE), France (POLARCAT-FRANCE)
Aircraft 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
The GEOS-Chem chemical transport model (CTM):

- Global, gridded, 3-D model
- Simulates emissions, chemistry, transport, and deposition
- Driven by input meteorology from an assimilated GCM
- Represents our best *a priori* understanding of processes
GEOS-Chem chemical transport model links datasets and provides added value

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Data and models work best together!

*We use data to:*
- Validate model concentrations
- Test our understanding of sources and processes

*We use the model to:*
- Link disparate data sets
- Test impacts of different sources and processes
- Draw broader implications beyond the time and space of the observations
I. Carbon monoxide (CO) as a pollution tracer

Source: incomplete combustion (fossil fuel & biomass burning)

Fisher et al., 2010
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**Model source attribution:**
- Asian anthropogenic source dominates Arctic CO background
- European anthropogenic source important near surface

![April 2008 Emissions](image)

![Mean CO Profile](image)

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

April 2008 AIRS CO anomaly

Lower-than-average CO over Alaska

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
II. Sources and acidity of sulfate-ammonium aerosol

Volcanoes

Industry

Phytoplankton

Ocean

Agriculture

Fires

\[ \text{SO}_2 \rightarrow \text{H}_2\text{O}_2, \text{O}_3 \rightarrow \text{OH} \rightarrow \text{H}^+, \text{SO}_4^{2-} \rightarrow \text{Acidic} \]
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SO$_2$ → OH

H$_2$O$_2$, O$_3$

Volcanoes Industry Phytoplankton Ocean Agriculture Fires

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

Acidic

Neutralized

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

DMS

OH

Volcanoes Industry Phytoplankton Ocean Agriculture Fires
II. Sources and acidity of sulfate-ammonium aerosol

The extent to which sulfate is neutralized by ammonium has implications for both atmospheric chemistry and climate.
Sulfate is a dominant component of winter/spring Arctic aerosol pollution.
Sulfate-ammonium aerosol in the Arctic

Barrow, Alaska
Fraction of aerosol mass

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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Quinn et al., 2002 (adapted)

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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
A diversity of sources contribute to aerosol in the North American Arctic in spring

Fossil fuel regions

N. American Arctic

European Arctic

Other sources:
Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH₃

Fisher et al., 2011
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Fossil fuel regions
- N. American Arctic
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- Ships
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**Sulfate**
- ARCTAS (SAGA)

**Ammonium**
- ARCTAS (SAGA)

Fisher et al., 2011
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**No single source is dominant**

**Graphs:**
- **SO$_4^{2-}$ (nmol m$^{-3}$ STP)**
- **NH$_4^+$ (nmol m$^{-3}$ STP)**

**Altitude (km):**
- 0-2
- 2-5
- 5-10

**Models:**
- ARCTAS (SAGA)
- GEOS-Chem Observations

*Fisher et al., 2011*
A diversity of sources contribute to aerosol in the North American Arctic in spring

- Fossil fuel regions: N. American Arctic, European Arctic
- Other sources: Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH$_3$

- No single source is dominant
- Below 2 km, E. Asian, European, and N. American sources have comparable influences on sulfate

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- Above 2 km, growing influence from E. Asian and biomass burning sources

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Fisher et al., 2011
A diversity of sources contribute to aerosol in the North American Arctic in spring

- No single source is dominant
- Below 2 km, E. Asian, European, and N. American sources have comparable influences on sulfate
- Above 2 km, growing influence from E. Asian and biomass burning sources
- Natural sources are important at all altitudes

Other sources: Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH$_3$

Fisher et al., 2011
Surface data highlight importance of West Asian emissions

Sulfate

Ammonium

Barrow (71.3°N, 156.6°W)

Denali (63.7°N, 149.0°W)

West Asian emissions important in High Arctic

Economic growth in Kazakhstan and increased energy production in Siberia may lead to increased Arctic aerosol in winter

Fisher et al., 2011
Spring aerosol ranged from very acidic to fully neutralized

Aerosol is very acidic, sulfate is primarily as H$_2$SO$_4$

Sulfate is primarily as (NH$_4$)HSO$_4$

Aerosol is fully neutralized, sulfate as (NH$_4$)$_2$SO$_4$

Fisher et al., 2011
Spring aerosol ranged from very acidic to fully neutralized

Aircraft Observations

GEOS-Chem

Fisher et al., 2011
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.
- Recent growth in West Asia may explain trends of increased aerosol acidity at Barrow.
- 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.

Fisher et al., 2011
III. Mercury in the Arctic Ocean is an ecological and human health concern

High mercury concentrations have been observed in the tissues of many Arctic species, including those that make up traditional northern diets.

Since the industrial revolution, both wildlife tissue mercury concentrations and atmospheric mercury emissions have increased substantially, but the links between the two remain uncertain.

Understanding atmosphere-ocean mercury exchange is critical to understanding the role of the atmosphere in biogeochemical mercury cycling and mercury toxicity.
The atmosphere-ocean mercury cycle
The atmosphere-ocean mercury cycle

Mid-latitudes

Arctic

Hg^0 \rightarrow Hg^{II} \rightarrow Hg^0

Geogenic, Anthropogenic, Biomass burning, Vegetation, soil, snow

Ocean surface

Ocean subsurface

Snow/ice
The atmosphere-ocean mercury cycle

**Mid-latitudes**

**Arctic**

Diagram showing the atmospheric ocean mercury cycle with different pathways and components.

- Hg$^0$ (elemental mercury)
- Hg$^{II}$ (dissolved mercury)

Pathways include:
- Geogenic sources
- Anthropogenic sources
- Biomass burning
- Vegetation, soil, snow

Ocean surface and subsurface interactions are depicted.
The atmosphere-ocean mercury cycle

**Mid-latitudes**

- Geogenic
- Anthropogenic
- Biomass burning
- Vegetation, soil, snow

**Arctic**

- Snow/ice

**Ocean subsurface**

- Hg^0 -> Hg^II

**Ocean surface**

- Hg^II -> Hg^0

**Mid-latitudes**

- Hg^0

**Arctic**

- Hg^II
Testing our current understanding: model comparison to atmospheric observations

**Observations:** multi-year mean over three Arctic sites

**Model:** GEOS-Chem at these sites in 2008

**Spring minimum:** Atmospheric Mercury Depletion Events

**Summer maximum:**
Testing our current understanding: model comparison to atmospheric observations

The observed summer peak in boundary layer Hg⁰ is significantly underestimated. Can we explain it using our standard understanding of mercury cycling as a source from:

• the atmosphere?
• the snowpack?
• the ocean?
Hypothesis 1:

atmospheric transport from mid-latitudes

Observations show more mercury in the Arctic than at mid-latitudes; hence the Arctic atmosphere is more likely a net exporter than importer of mercury in the summer.
Hypothesis 2: re-emission from snowpack and ice

Snowpack source reflects balance between re-emission and meltwater formation; mercury does not remain in snowpack late enough to sustain the summer peak.
Hypothesis 3: chemical kinetics in the Arctic surface ocean

Ocean evasion reflects redox chemistry AND total Hg pool. Even with enhanced reduction, the summer peak cannot be captured because total ocean Hg pool is depleted due to seasonal loss from subsurface exchange.
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Hypothesis 4: a large source to the Arctic Ocean from circumpolar rivers

Water discharge seasonality

All (>100 km$^3$ a$^{-1}$)

Russian only
Hypothesis 4: a large source to the Arctic Ocean from circumpolar rivers

With river source included, GEOS-Chem can reproduce the summer peak.

Large seasonal river source replenishes total Hg pool, drives spring-summer evasion from the surface ocean.
Proposed budget of mercury in the Arctic surface ocean

Atmosphere

- snowpack emission: 32
- deposition to snow/ice: 62
- atmospheric transport

Snow / Ice

- runoff to land

Surface Ocean

- meltwater: 20

Subsurface Waters

- river delivery: 160

All fluxes in Mg a⁻¹
Proposed budget of mercury in the Arctic surface ocean

- Atmosphere
  - Snowpack emission: 32 Mg a⁻¹
  - Deposition to snow/ice: 62 Mg a⁻¹
  - Atmospheric transport: 100 Mg a⁻¹

- Snow/Ice
  - Runoff to land
  - Evasion: 100 Mg a⁻¹

- Surface Ocean
  - Meltwater: 20 Mg a⁻¹

- Subsurface Waters

- Rivers
  - River delivery: 160 Mg a⁻¹

All fluxes in Mg a⁻¹
Conclusions and take-home messages

1. Mid-tropospheric pollution transport to the Arctic is evident in aircraft, satellite, and surface data. By using these data together with model simulations, we can quantify the source contributions, understand the drivers of variability, and assess the implications of Arctic pollution.
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2. Pollution in the Arctic originates from a diversity of sources - especially Europe, East Asia, and West Asia - with relative importance dependent on species, altitude, and season.
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4. Each anthropogenic aerosol source result in different contribution to aerosol composition and aerosol acidity.

5. The summer peak in atmospheric mercury cannot be explained in terms of conventional ideas but can be reproduced assuming a major source from rivers. This suggests the Arctic Ocean may be a net annual source to the atmosphere.
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