Arctic Aerosols: Sources and Composition

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Aerosol terminology

- **Aerosol** – a dispersion of solid and liquid particles suspended in gas (air)

*Note: “aerosol” is defined as the dispersion of both particles and gas, but in common practice it is used to refer to the particles only!*

- **Primary aerosol** – atmospheric particles that are emitted or injected directly into the atmosphere.

- **Secondary aerosol** – atmospheric particles that are created by low-volatility gas phase molecules
  - **Condensation** onto existing particles
  - **Nucleation** of new particles

*Primary and secondary may be natural or anthropogenic*
Aerosols: solid or liquid particles in the atmosphere

Primary Emissions

Direct emission of particles

Nucleation of small particles

Aerosol Processing

Gaseous Precursor

Condensation of gases, heterogeneous chemistry

Droplet Activation
### Uncertainties Due to Aerosols

<table>
<thead>
<tr>
<th>Emitted compound</th>
<th>Resulting atmospheric drivers</th>
<th>Radiative forcing by emissions and drivers</th>
<th>Level of confidence</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO₂</td>
<td>CO₂</td>
<td>1.68 [1.33 to 2.03]</td>
<td>VH</td>
</tr>
<tr>
<td>CH₄</td>
<td>O₂, H₂O, O₃, CH₄</td>
<td>0.97 [0.74 to 1.20]</td>
<td>H</td>
</tr>
<tr>
<td>Halo-carbons</td>
<td>O₂, CFCs, HCFCs</td>
<td>0.18 [0.01 to 0.35]</td>
<td>H</td>
</tr>
<tr>
<td>N₂O</td>
<td>N₂O</td>
<td>0.17 [0.13 to 0.21]</td>
<td>VH</td>
</tr>
<tr>
<td>Antropogenic</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO</td>
<td>CO₂, CH₄, O₃</td>
<td>0.23 [0.16 to 0.30]</td>
<td>M</td>
</tr>
<tr>
<td>NMVOC</td>
<td>CO₂, CH₄, O₃</td>
<td>0.10 [0.05 to 0.15]</td>
<td>M</td>
</tr>
<tr>
<td>NO₂</td>
<td>Nitrate, CH₄, O₃</td>
<td>-0.15 [-0.34 to 0.03]</td>
<td>M</td>
</tr>
<tr>
<td>Aerosols and precursors</td>
<td>Mineral dust, Sulphate, Nitrate, Organic carbon, Black carbon</td>
<td>-0.27 [-0.77 to 0.23]</td>
<td>H</td>
</tr>
<tr>
<td></td>
<td>Cloud adjustments due to aerosols</td>
<td>-0.55 [-1.33 to -0.06]</td>
<td>L</td>
</tr>
<tr>
<td></td>
<td>Albedo change due to land use</td>
<td>-0.15 [-0.25 to -0.05]</td>
<td>M</td>
</tr>
<tr>
<td>Natural</td>
<td>Changes in solar irradiance</td>
<td>0.05 [0.00 to 0.10]</td>
<td>M</td>
</tr>
</tbody>
</table>

#### Total anthropogenic RF relative to 1750

- **2011**: 2.29 [1.13 to 3.33] (H)
- **1980**: 1.25 [0.64 to 1.86] (H)
- **1950**: 0.57 [0.29 to 0.85] (M)

IPCC AR5, 2013
How much aerosol is there?

- Total mass concentration is between 1-100 μg m\(^{-3}\) (Arctic: < 1 μg m\(^{-3}\))
  - Total mass of air ~1 kg m\(^{-3}\)
  - ~1-100 ppb\(_{\text{mass}}\)
- Total number concentration is between 100-100,000 # cm\(^{-3}\) (Arctic: ~ 1-200 cm\(^{-3}\))
Size distributions

Number distribution
\[ n_n(\log D_p) = \frac{dN}{d \log D_p} \]

Surface area distribution
\[ n_s(\log D_p) = \frac{dS}{d \log D_p} \]

Volume distribution
\[ n_v(\log D_p) = \frac{dV}{d \log D_p} \]

Seinfeld and Pandis, *Atmospheric Chemistry and Physics*
Atmospheric aerosol sources

[Image of a diagram showing various sources of aerosols, including direct and indirect radiative forcing, nucleation, oxidation, and various natural and anthropogenic sources such as hydrocarbons, SO\textsubscript{2}, NO\textsubscript{x}, NH\textsubscript{3}, POA, sea-salt, dimethylsulfide, and phytoplankton.]

https://www.bnl.gov/today/body_pics/2016/03/aerosol-sources-hr.jpg
Global Aerosol Source Contributions

- **Mineral**: 2000 Tg/yr
- **Sea Salt**: 3000 Tg/yr
- **Volcanic Dust**: 3000 Tg/yr
- **Biological debris**: 50 Tg/yr
- **Natural Secondary**: 275 Tg/yr
- **Anth Primary**: 105 Tg/yr
- **Anth Secondary**: 360 Tg/yr

Units: Tg/yr
Seinfeld and Pandis, Atmospheric Chemistry and Physics
Aerosols in the Arctic

Transport

Sources

Radiation
Seasonality of Arctic Aerosol Mass

Quinn 2007  Tellus B, 59, 99
Arctic Haze

Winter Transport to Arctic

Inefficient ice-cloud scavenging

Ascent over polar dome

Anthropogenic Arctic emissions

Deposition on snow

Ice-ocean feedbacks

Arnold et al. 2016 *Elementa*, 4, 104

How will transport change in the future?
Seasonal Transport into the Arctic

Winter
• northern Eurasia influence
• no solar radiation
• stable inversions
• minimal deposition
• shorter transport time

Summer
• less continental influence
• solar radiation
• convection
• large rate of wet deposition
• longer transport time
Average Aerosol Composition at Alert: 2006

Units: µg/m³

Black Carbon
0 – 0.15 µg/m³

Organics???

<table>
<thead>
<tr>
<th>Element</th>
<th>Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO₄</td>
<td>0.3653</td>
</tr>
<tr>
<td>Cl</td>
<td>0.1743</td>
</tr>
<tr>
<td>Na</td>
<td>0.1340</td>
</tr>
<tr>
<td>NH₄</td>
<td>0.0462</td>
</tr>
<tr>
<td>NO₃</td>
<td>0.0572</td>
</tr>
<tr>
<td>Fe</td>
<td>0.0398</td>
</tr>
<tr>
<td>Mn</td>
<td>0.0008</td>
</tr>
<tr>
<td>Pb</td>
<td>7E-6</td>
</tr>
<tr>
<td>Br</td>
<td>0.0023</td>
</tr>
</tbody>
</table>


Sharma et al. 2004 JGR, 109, D15
Sources of Arctic Aerosol: 1980-1995

- Photochemical sulphate
- Anthropogenic sulphate
- Mixed photochemical sulphate / sea salt
- Biogenic sulphur (MSA)
- Sea salt
- Smelter
- Soil
- Nitrate, bromine, iodine
- **Biomass burning**

Future Sources:
- Shipping
- Oil extraction

Siros and Barrie *1999 JGR-Atmosphere*, 104, 11599
Marine Sulphur Chemistry as a Source of Arctic Aerosol

Phytoplankton

Charlson et al. 1987 Nature, 326, 655
Biogenic sulphate

Transported

Local

Vertical bars represent the middle 90 percentile

Month

Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

$f_b$
Nucleation in Marine Environments

- too many existing particles for nucleation to occur in boundary layer

- driven by sulphuric acid

- nucleation in free troposphere
  - colder, cleaner, greater solar radiation
Nucleation and Growth
Nucleation / Growth at Whistler Mountain
“Banana Plot”

Unpublished data
Ambient Nucleation Rates in Boreal Forest

**Activation**
A: $0.4-3.0 \times 10^{-6} /s$

**Kinetic**
K: $0.2-1.4 \times 10^{-12} \text{cm}^3/s$

H$_2$SO$_4$ is main component

Sihto et al., 2006 *Atmos Chem Phys*
Organics Contribute to Nucleation

Grey dots:
Ambient data from Hyytiala

Organics formed by
TMB oxidation

As do:
NH$_3$
Amines
Biogenic VOCs

Metzger et al. 2010 Proc Nat Acad Sci USA
Schematic of Particle Formation

I
Small clusters and molecules
- No direct connection to NPF
- Very slow growth

II
Critical size for clustering
- Sulfuric acid and amines
- Stabilizing organic compounds
- Slowly growing (<1 nm/h)
- Determines $J_{1.5}$

III
Growing clusters
- Organics start to dominate
- Rapidly growing (~2 nm/h)
- Nano-Köhler
- Determines $J_{3}$

Key processes:
- Gas-phase reactions, cluster formation/evaporation
- Cluster stabilization
- Activation of clusters for enhanced growth

Kulmala, 2013 Science
Nucleation in Marine Environments

• too many existing particles for nucleation to occur in boundary layer

• driven by sulphuric acid

• nucleation in free troposphere
  – colder, cleaner, greater solar radiation
Particle Growth in Canadian High Arctic

Alert (82.5N, 62.3W)

Eureka (80N, 86.5W)

Ridge Lab of the Polar Environment Atmospheric Research Laboratory (610 m asl)

Dr. Neil Trivett Global Atmosphere Watch Observatory (210 m asl)
Regional conditions favourable for nucleation and growth

Chang, Hayes et al. *in prep*
Organics are oxygenated but not from biomass burning

Chang, Hayes et al. *in prep*
NETCARE: Marine Emissions and Organics Contribute to Aerosol Growth

Willis et al. 2016 Atmos Chem Phys, 16, 7663
NETCARE: Marine Emissions and Organics Contribute to Aerosol Growth

Willis et al. 2016 Atmos Chem Phys, 16, 7663
Other NETCARE Results

• E. Mungall – volatile organics originating from ocean surface

• J. Burkart, D. Collins – particle growth events in Canadian Arctic
Summer Arctic Aerosol Composition and Sources
Arctic Summer Cloud and Ocean Study
Aug 1 – Sep 9, 2008

map courtesy of I. Brooks
Aerosol Chemical Composition Measured by AMS

MSA fragmentation from Langley et al. 2010 Atmos. Chem. Phys.
Chang et al. 2011 *Atmos. Chem. Phys.*, 11, 10619
Composition of Factors

Chang et al. 2011 Atmos. Chem. Phys., 11, 10619
Summary

• winter aerosol is transported

• summer aerosol is more local
  – organic component is important
  – nucleation and growth observed and linked to marine sources
  – How will this change in future climate?
  – How will industrial activities affect aerosol processes?