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

Jenny A. Fisher
Harvard University

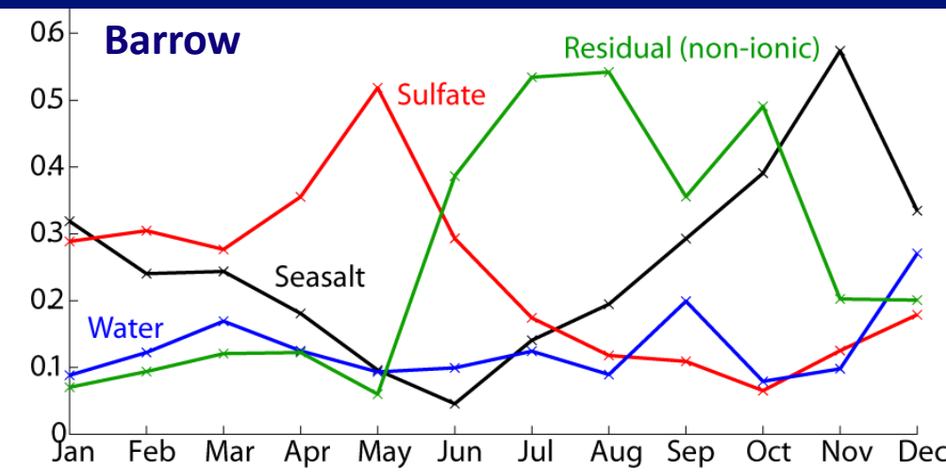
D.J. Jacob, Q. Wang, R. Bahreini, C.C. Carouge, M.J. Cubison, J.E. Dibb, T. Diehl, J.L. Jimenez, E.M. Leibensperger, M.B.J. Meinders, H.O.T. Pye, P.K. Quinn, S. Sharma, A. van Donkelaar, R.M. Yantosca

5th International GEOS-Chem Meeting
2 May 2011

With support from the NASA Tropospheric Chemistry Program

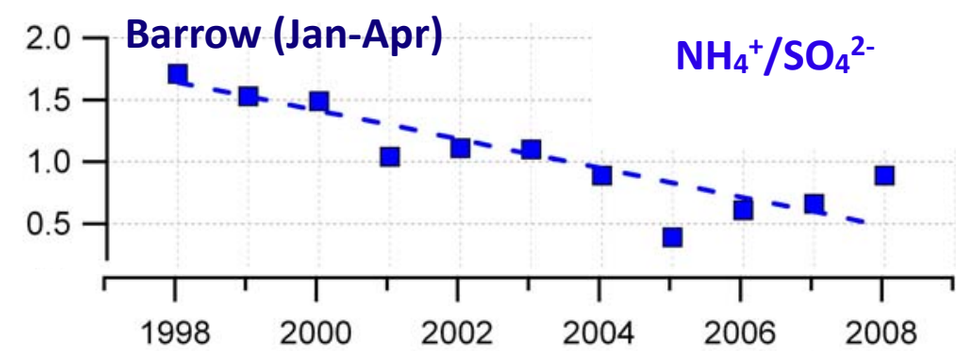
Motivation

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



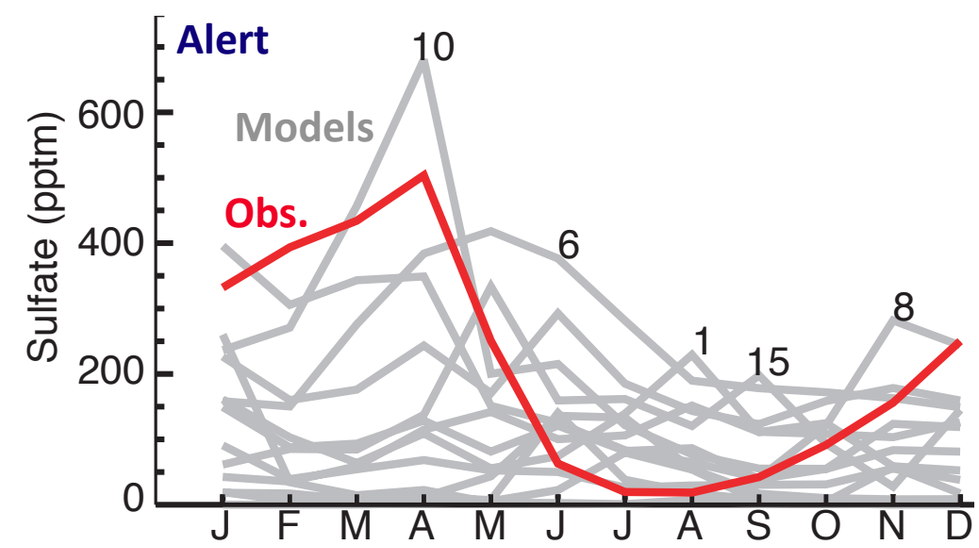
Quinn et al., 2002

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**.



Quinn et al., 2009

The **sources** of inorganic aerosol to the Arctic are **highly uncertain**, and observed concentrations **can't be reproduced** by models.

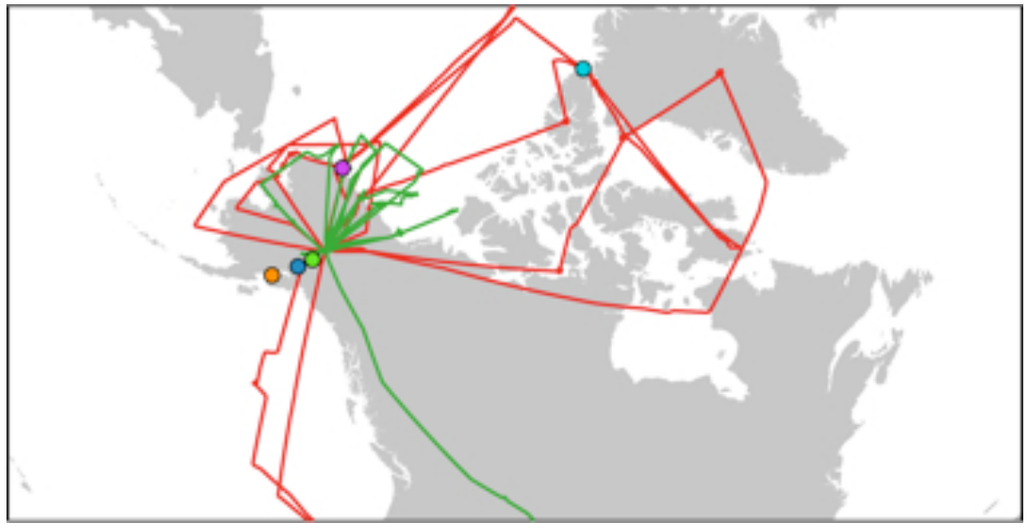


Shindell et al., 2008

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

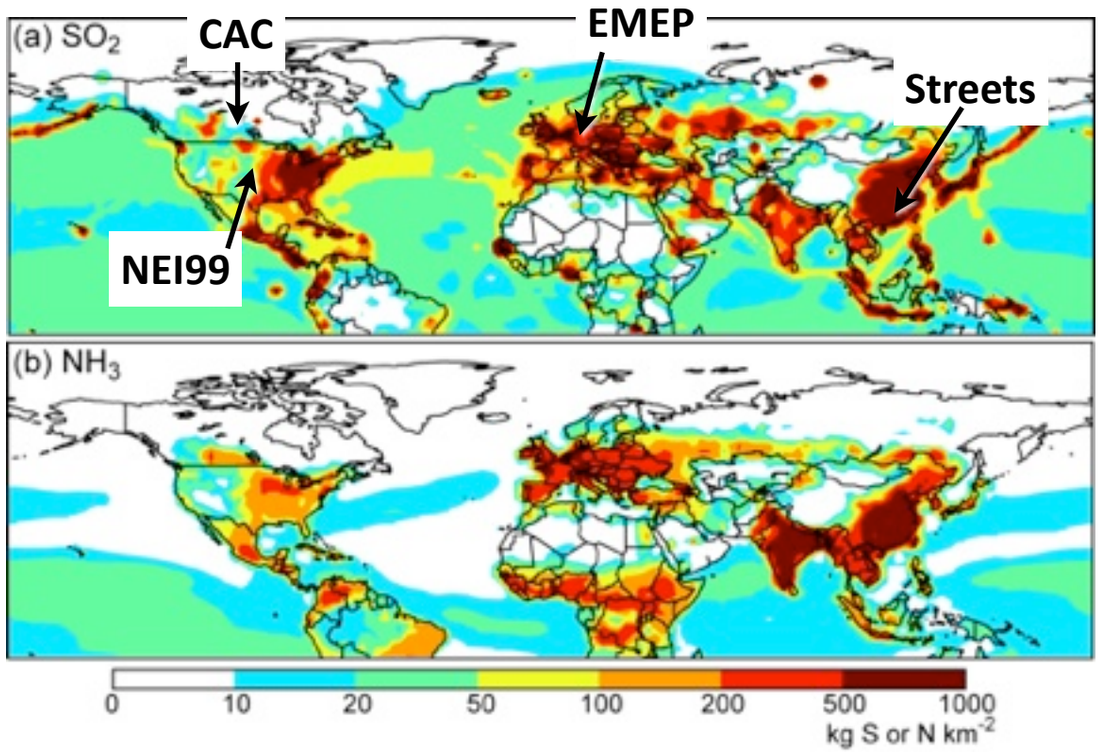


3. **GEOS-Chem v8-02-03** full chemistry, GEOS-5 meteorology, 2°x2.5° horizontal resolution

SO_2 emissions:
-EDGAR anthropogenic with regional overwrites (map)
-FLAMBE biomass burning with corrections (Fisher et al. 2010)
-AEROCOM volcanism

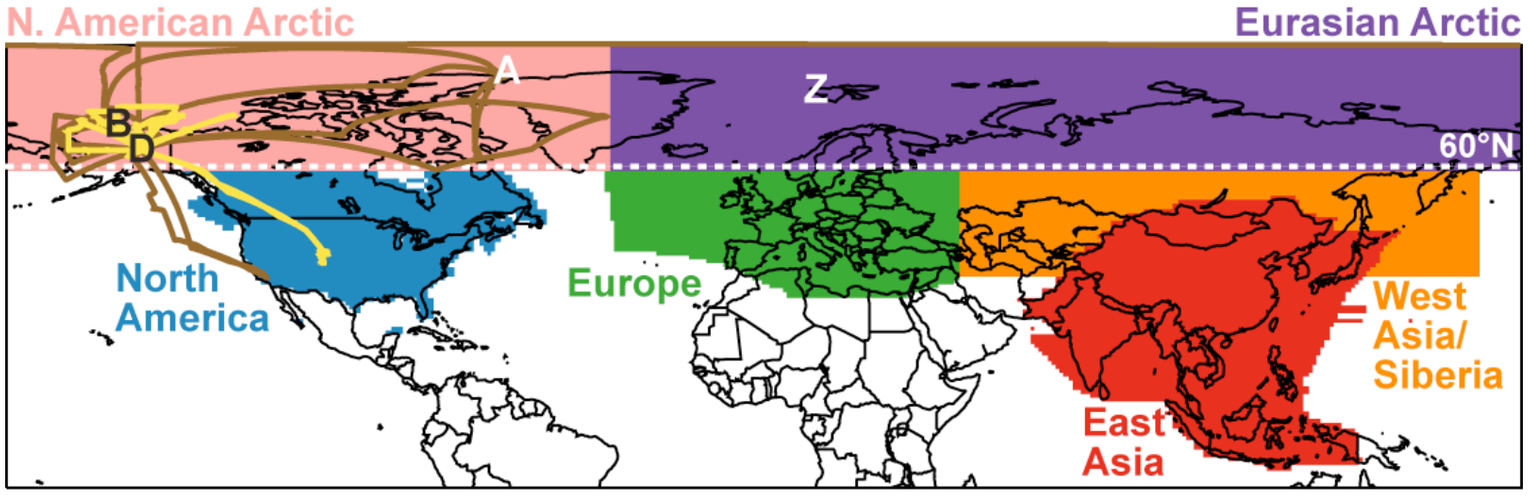
NH_3 emissions:
-Bouwman et al., 1997 anthropogenic (with overwrites) & natural
-FLAMBE biomass burning with corrections (Fisher et al. 2010)

Additions to standard v8-02-03 model
-Improved cold temp. wet scavenging (Q. Wang)
-Imposed aerosol dry deposition velocity over ice
-Imposed NH_3 seasonality over Asia



A diversity of sources contributed to aerosol in the North American Arctic during ARCTAS (April 2008)

Fossil fuel regions

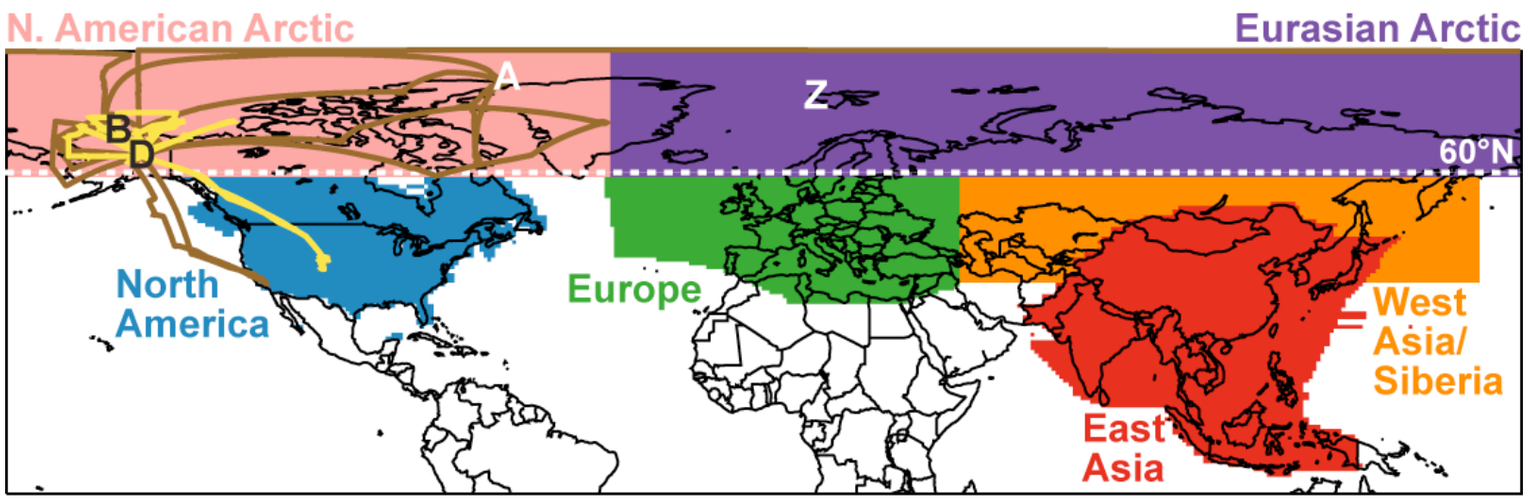


Other sources:

Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH_3

A diversity of sources contributed to aerosol in the North American Arctic during ARCTAS (April 2008)

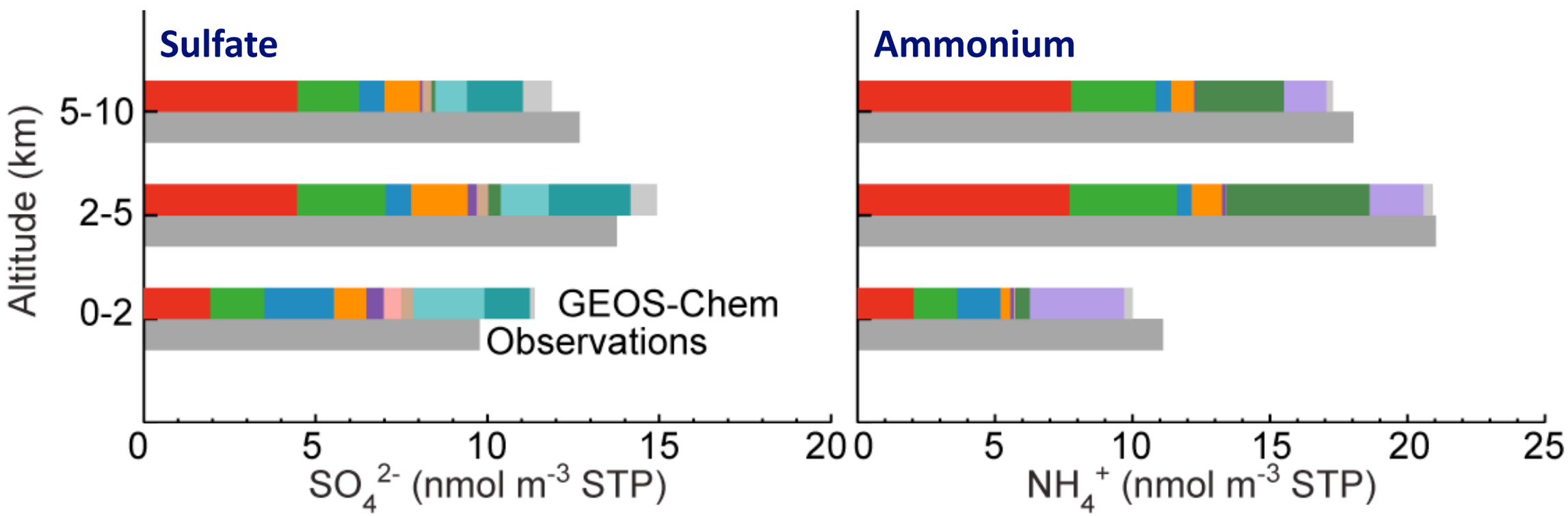
Fossil fuel regions



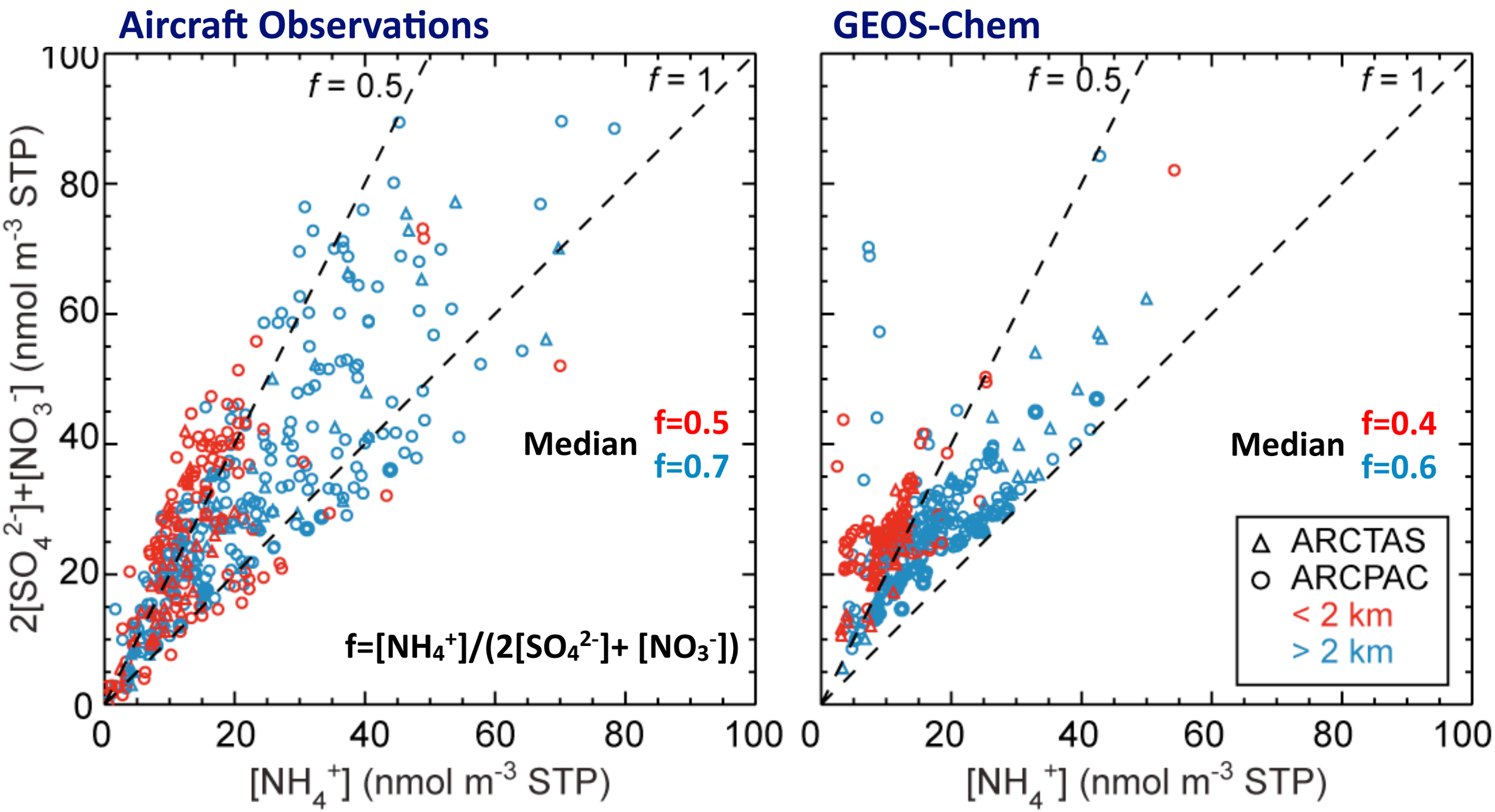
Other sources:

Ships, Biomass burning, DMS oxidation, volcanic emission, natural NH_3

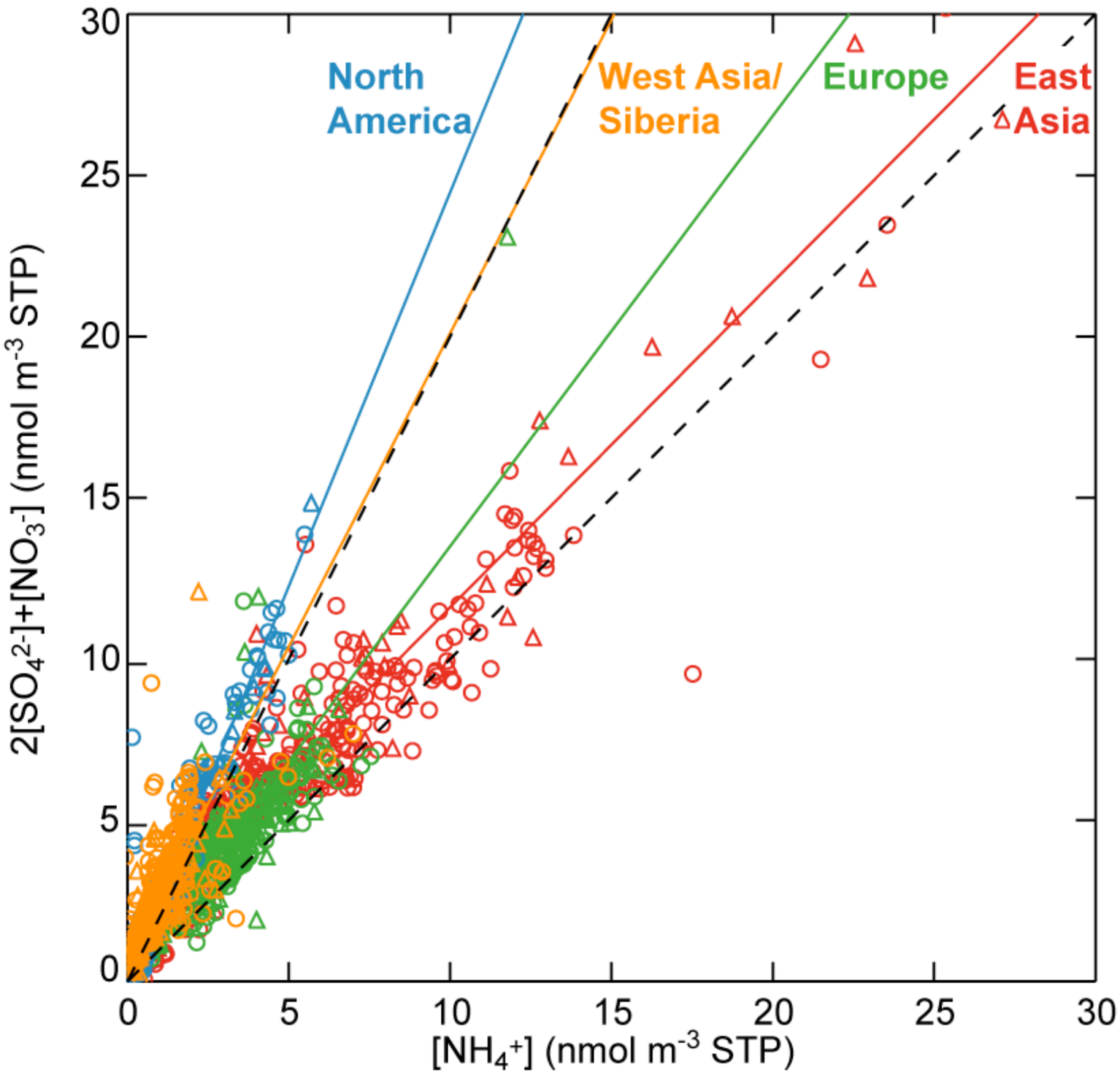
- E. Asia and Europe are major sources of sulfate and ammonium at all altitudes
- Below 2 km, E. Asian, European, and N. American sources have comparable influences on sulfate
- Natural sulfate sources (DMS, volcanic) are important at all altitudes
- Russian fires are an important source of ammonium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



Arctic spring aerosol during ARCTAS/ARCPAC ranged from very acidic to fully neutralized



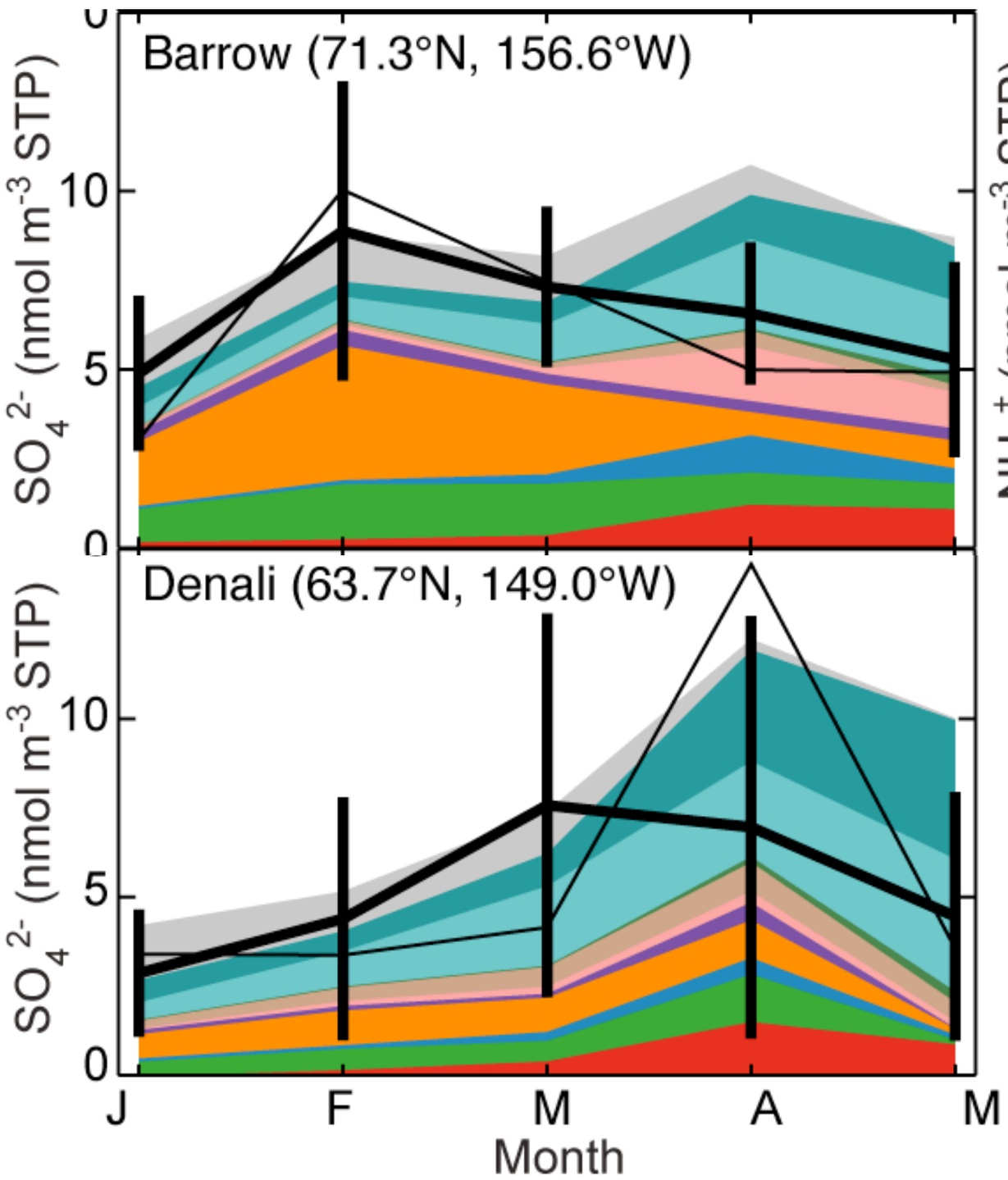
Different source regions show different neutralization signatures in the Arctic



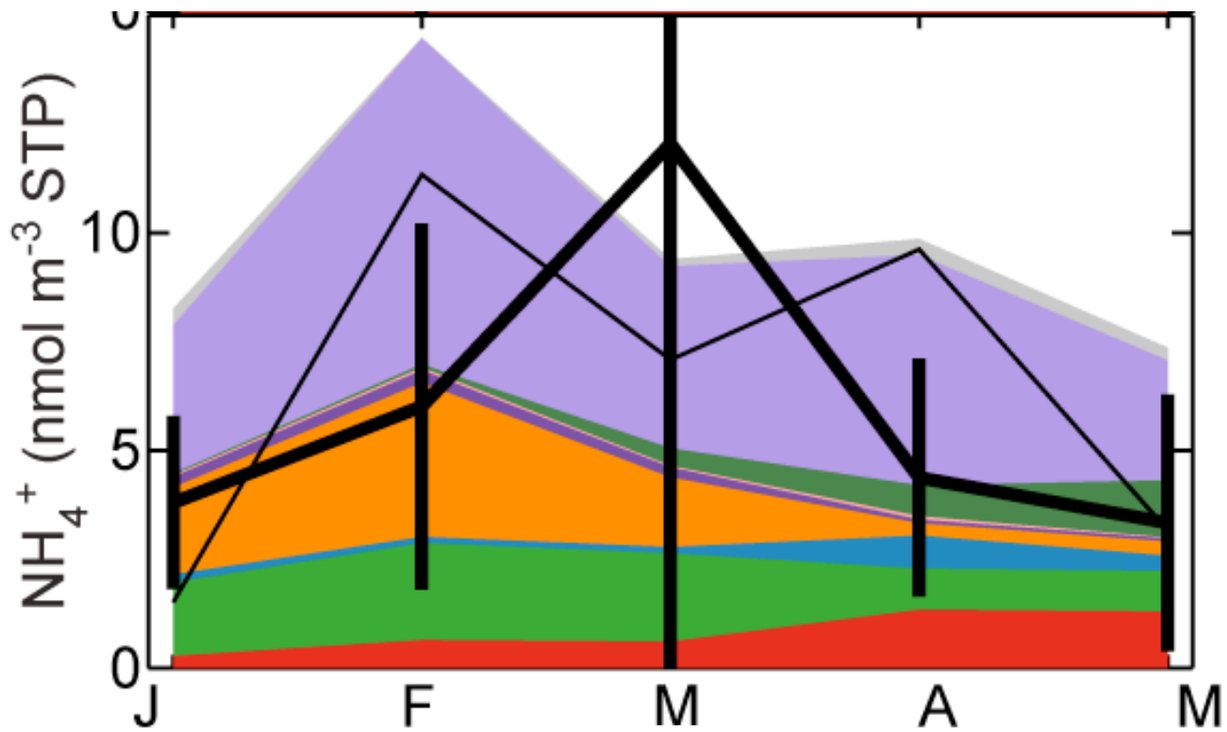
- Aerosol from Europe and East Asia is mostly neutralized
- Aerosol from West Asia / Siberia and North America is significantly more acidic
- Free tropospheric aerosol will likely become more neutralized as SO_X emission controls are implemented in East Asia
- However, opposite trend is occurring in the boundary layer, at least at Barrow...

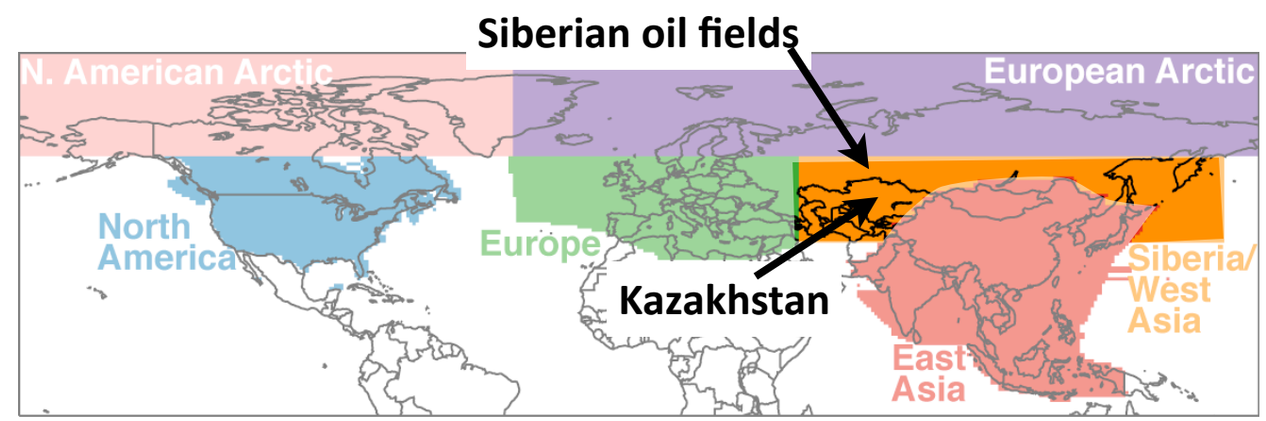
Surface data highlight importance of West Asian/Siberian emissions

Sulfate



Ammonium





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