Sources and Removal of Springtime Arctic Aerosol

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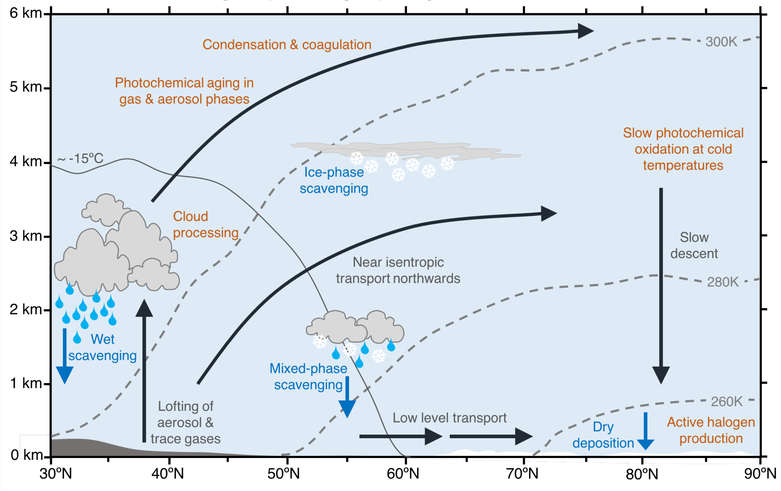
Motivation



Most chemical transport models cannot replicate observations of Arctic aerosol, particularly the vertical profile. This has important implications for estimates of aerosol impacts on climate[1].

We lack a predictive understanding of pollution transport to Arctic regions largely due to poor understanding of removal mechanisms and aerosol chemical and physical processing during transport[1].

- How does the vertical profile of aerosol chemical components vary under chronic Arctic Haze[2] conditions?
- How do the transport mechanisms of Arctic aerosol impact its properties?
- How is aerosol efficiently removed in Arctic air masses?

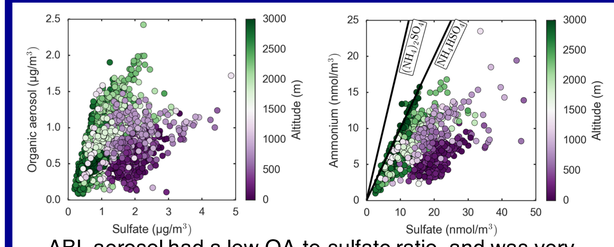
[1]Arnold, S. et al., Elementa, 2016
[2]Brock, C. et al., ACP, 11, 2011

Conclusions

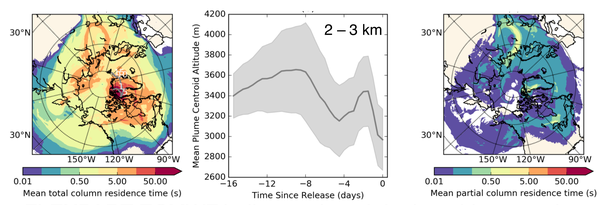
Under conditions consistent with chronic Arctic Haze[2] we observe evidence for:

- Sources of partially neutralized aerosol with higher organic and black carbon content in the mid-troposphere
- Contributions to acidic sulfate in the lower troposphere from slow gas-phase SO_2 oxidation in the Arctic
- Depletion of aerosol relative to CO in the mid-to-upper troposphere through wet scavenging over Greenland and in the lower troposphere likely through dry deposition to the surface

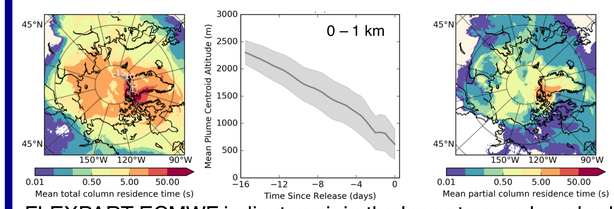
Aerosol Sources and Transport Pathways



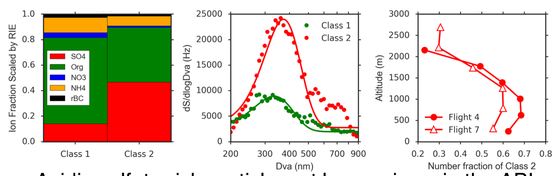
ABL aerosol had a low OA-to-sulfate ratio, and was very acidic. Near 3 km OA-to-sulfate ratios approach 1, and neutralization was consistent with ammonium bisulfate.



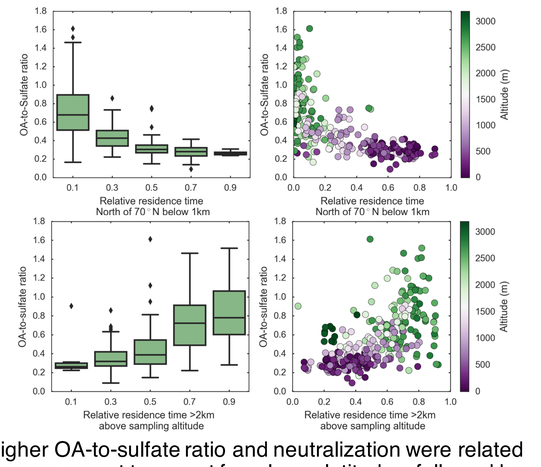
FLEXPART-ECMWF indicates that air in the mid-troposphere had a shorter residence time in the Arctic and likely originated from Northern Eurasia and North America. Asian sources may also contribute on a time-scale longer than 10 –15 days[3].



FLEXPART-ECMWF indicates air in the lower troposphere had a long residence time in the Arctic, and a history of descent from the mid – troposphere.



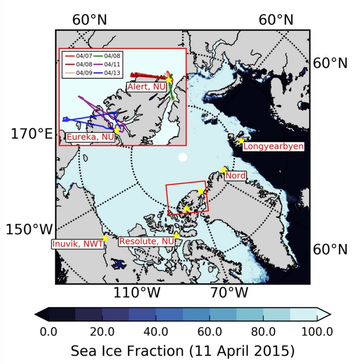
Acidic sulfate-rich particles, at larger sizes, in the ABL suggest that slow gas phase oxidation of transported SO_2 contributes to sulfate mass in ABL aerosol.



Higher OA-to-sulfate ratio and neutralization were related to more recent transport from lower latitudes, followed by descent from the mid-to-upper troposphere.

[3]Xu J. et al., ACPD, 2017

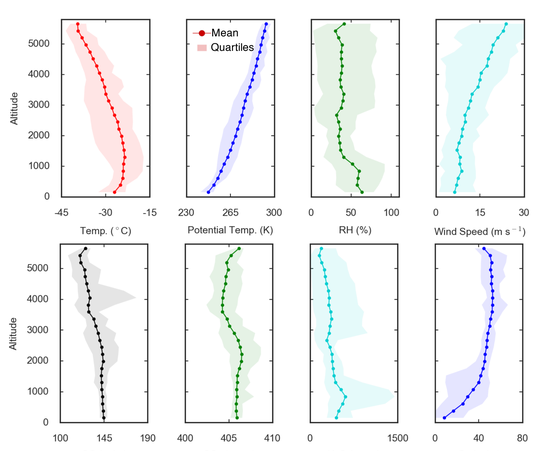
Campaign Overview

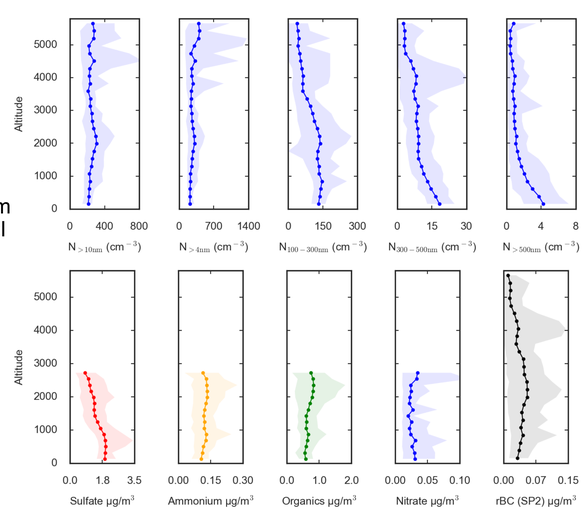


10 science flights from 4 – 21 April, 2015

6 flights in High Arctic Canada, based in Alert and Eureka, Nunavut

Measurements from 40 m to 6 km included aerosol size, number and non-refractory composition, trace gases and meteorological parameters.





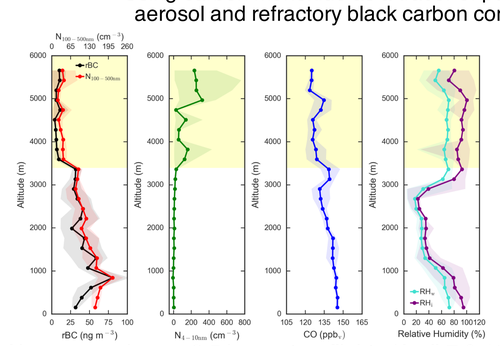
In High Arctic spring, we observed atmospheric conditions consistent with "chronic Arctic Haze[2]," with CO < 170 ppb_v, and Arctic Boundary Layer (ABL) heights, defined by the vertical extent of ozone depletion, up to ~1km.

Aerosol organic aerosol (OA)-to-sulfate ratio, refractory black carbon (rBC)-to-sulfate ratio and neutralization increased with altitude. Particle size distributions indicated a source of larger accumulation mode particles ($N_{300\text{-}500}$ and $N_{>500}$) at the surface, which likely contain components of sea salt.

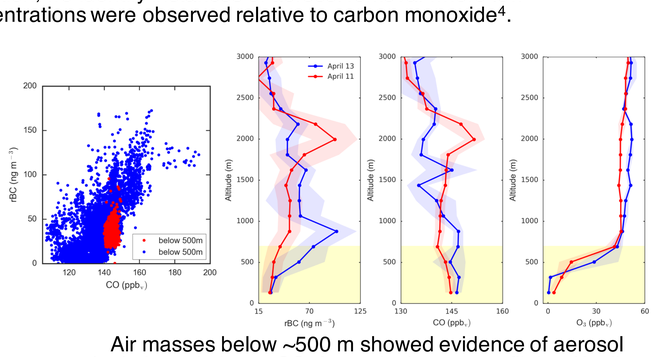
[2]Brock, C. et al., ACP, 11, 2011

Aerosol Depletion in Arctic Air Masses

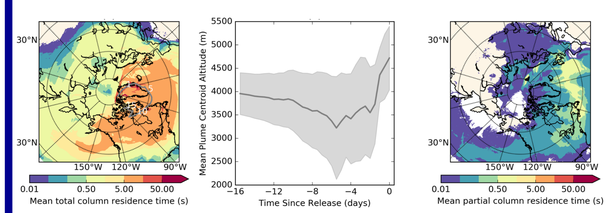
Using carbon monoxide as a tracer of pollution, we identify instances of aerosol removal where low aerosol and refractory black carbon concentrations were observed relative to carbon monoxide[4].



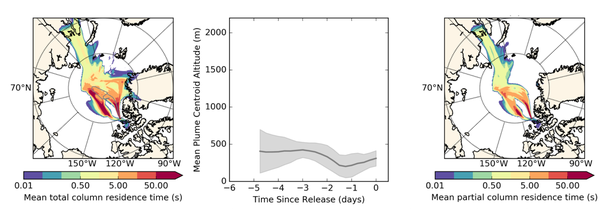
Air masses above ~3.5 km on two flights (April 7 & 13) had low aerosol concentrations, CO > 120 ppb_v and were at ice saturation, suggesting removal had occurred.



Air masses below ~500 m showed evidence of aerosol depletion relative to CO. In particular, on two flights (April 11 & 13) where aerosol and rBC concentrations decreased toward lower altitudes and O_3 fully was depleted.



Aerosol-depleted air masses at ice saturation had risen during transport over Greenland within the past 4 – 5 days, and likely experienced scavenging. Enhanced concentrations of small particles accompanied this efficient clean out.



These air masses had spent up to 4 days at low altitude over sea ice, and were more depleted in aerosol and O_3 compared to ABL air masses that descended from aloft, suggesting dry deposition to the surface occurred.

[4]Garrett, T. et al., GRL, 38, 2011

Acknowledgements

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