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Photoacoustic Observations of Aerosol Optical Properties Aloft Alaska: Links to Chemical Composition

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ARM

ASP

Earth & Environmental Sciences

ASR Cloud/Aerosol Meeting Oct 1 2009, Boulder CO

Outline

- Motivation

  Models under-predict Arctic warming & ice melting

  Arctic aerosol forcing is large and episodic but uncertain
- ISDAC Observational Strategy

  State-of-art in situ measurements of optical properties (photoacoustic), chemistry/size distributions (SPLAT).
- Key Objective

  Prognostic models for aerosol optical properties from size distributions and chemical composition (mixing state, coating)
- Fresh Results to Guide Modelers

  Quantify spring forcing by Arctic haze over Alaska

  Improve Arctic aerosol-cloud-radiation treatments.

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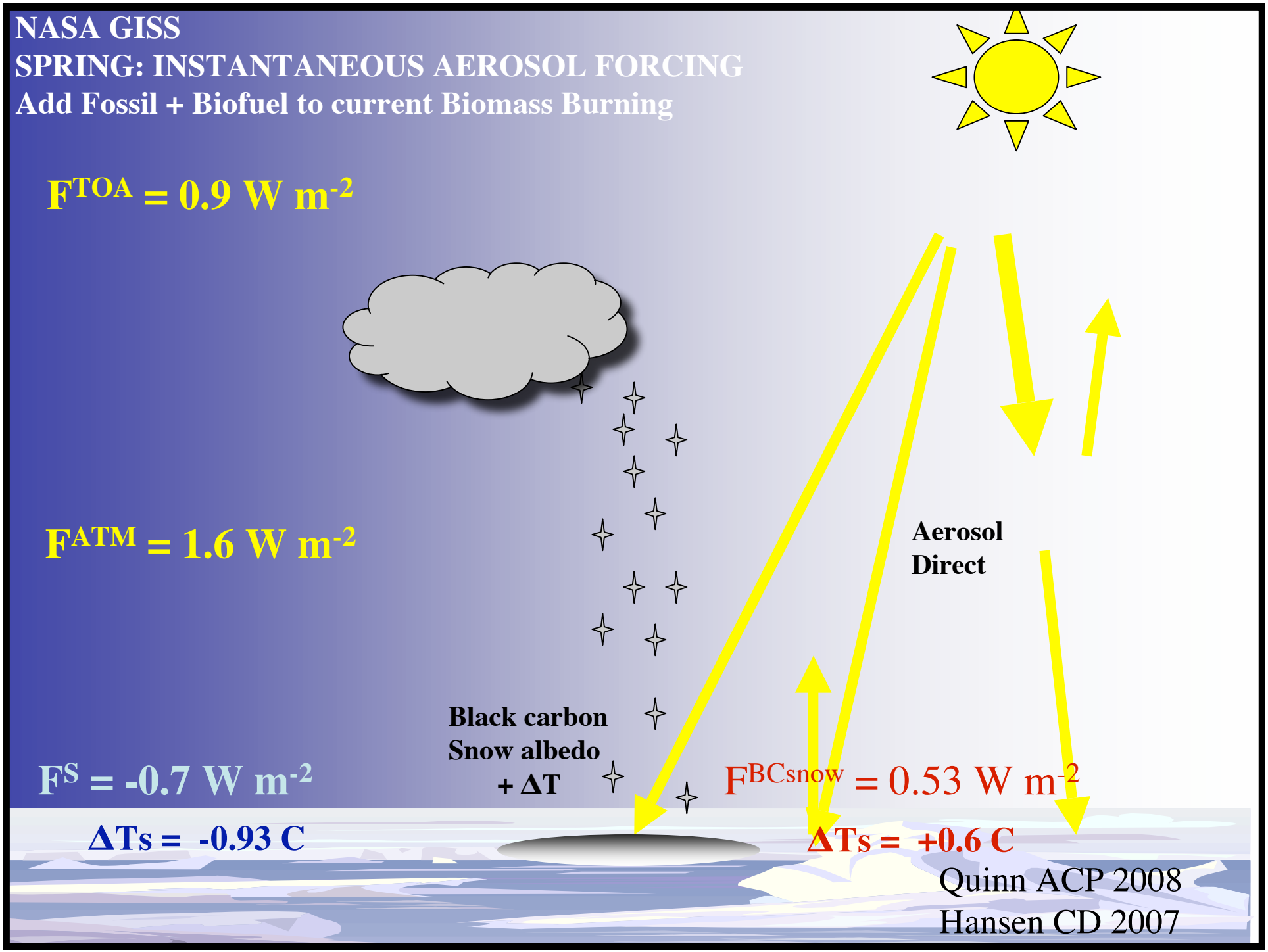
NASA GISS
SPRING: INSTANTANEOUS AEROSOL FORCING
Add Fossil + Biofuel to current Biomass Burning
F^TOA = 0.9 W m^-2
F^ATM = 1.6 W m^-2
Aerosol
Direct
Black carbon
Snow albedo
+ ΔT
F^S = -0.7 W m^-2
F^BCsnow = 0.53 W m^-2
ΔTs = -0.93 C
ΔTs = +0.6 C
Quinn ACP 2008
Hansen CD 2007

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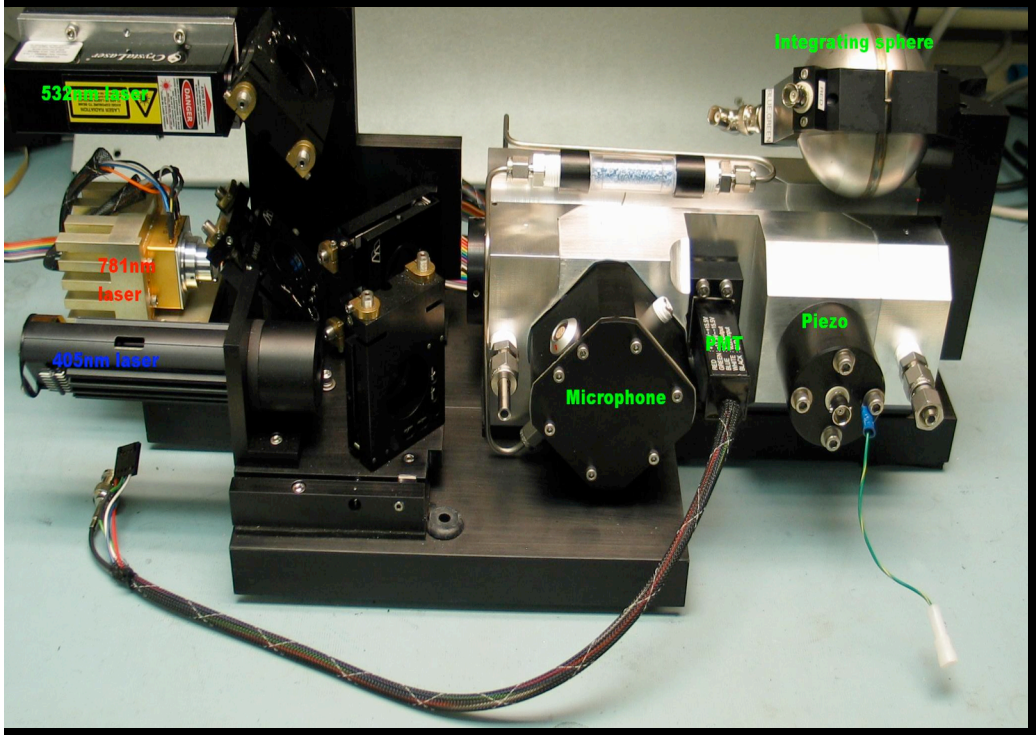
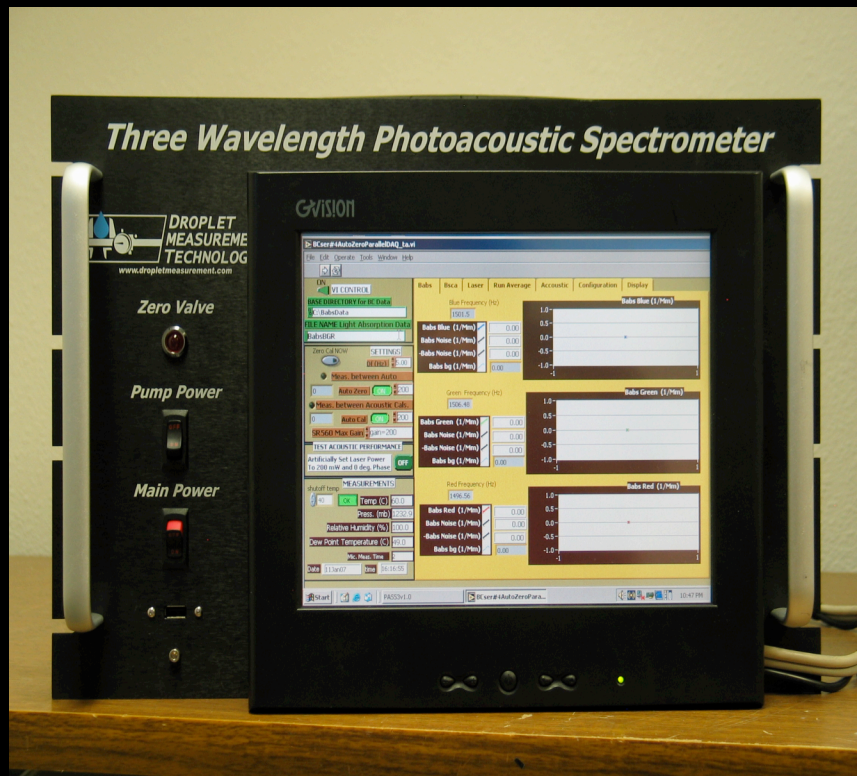
3-Laser Photoacoustic (DMT, LANL) Absorption and Scattering 405, 532, 781nm

- Measures absorption, scattering and single scatter albedo (=scattering/extinction)
- Direct (matrix free) absorption, simultaneous scattering, precise SSA (dry) meas't
- Can discriminate small from big, as well as soot, dust and sulfate.
- How dark (warming) or light (cooling) are aerosols?
- What aerosols are good cloud/ice nuclei (sulfate, soot, dust, organics)?
- Do cloud processes darken aerosols (CHAPS) as they deposit on snow (ISDAC)?





Satellite Retrievals → Global Assimilation → Regional Prediction → Validation

RAQMS Realtime Air Quality Modeling System

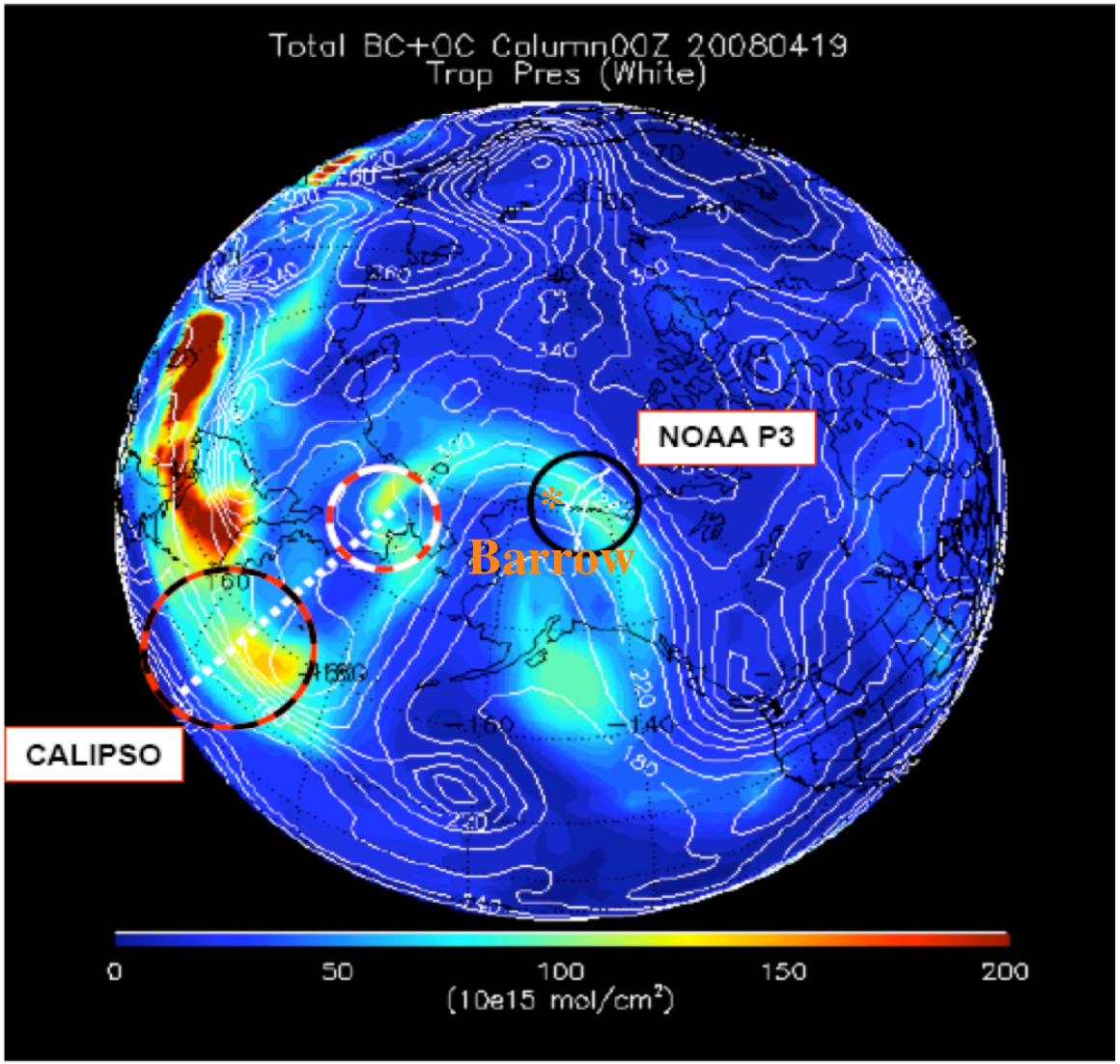
April 19, 2008 BC+OC

RAQMS total column BC+OC (x1015 mol/cm2) analysis at 00Z on April 19th, 2008.

The tropopause pressure is contoured.

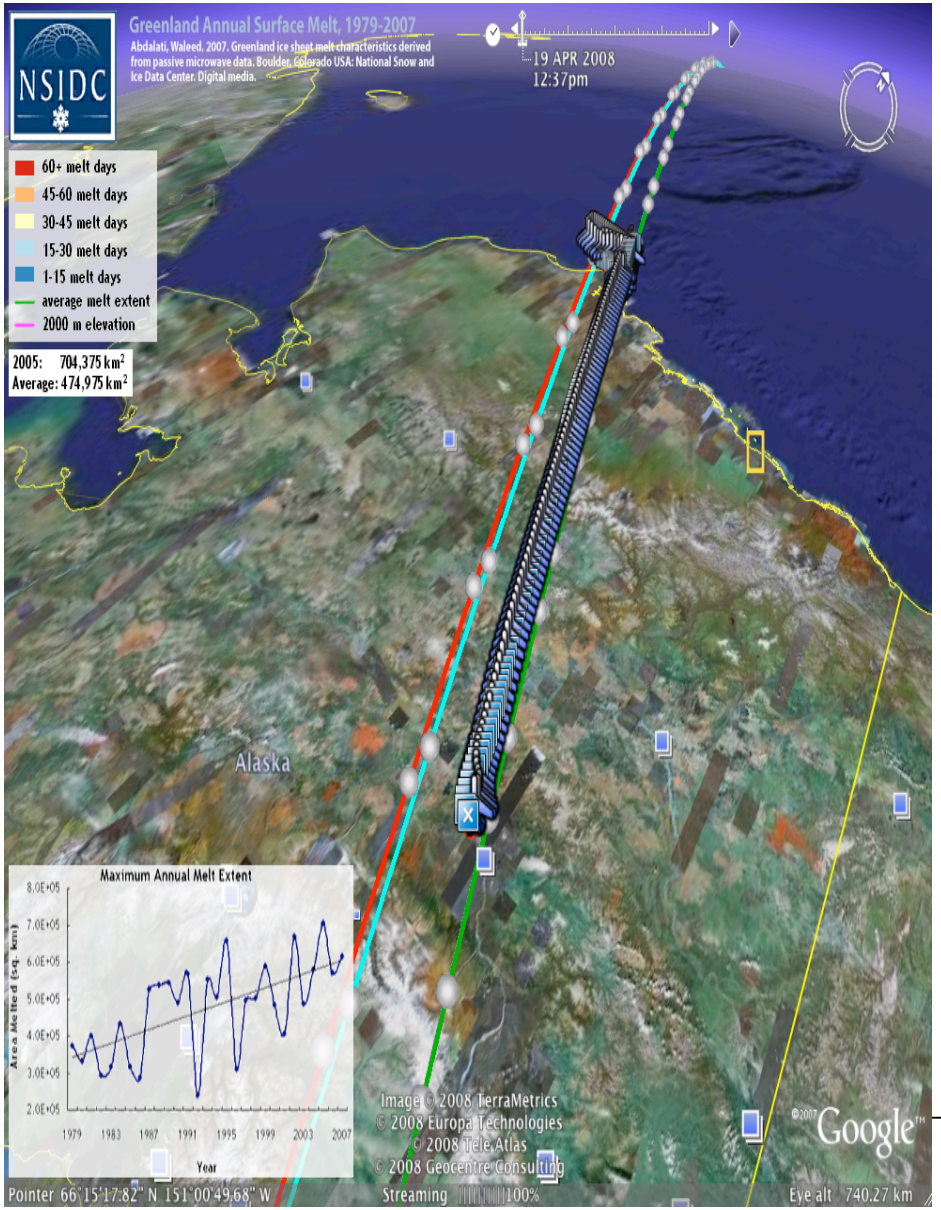
The location of the 15:17:11Z April 18th, 2008 CALIPSO Track is shown as a bold dotted line.

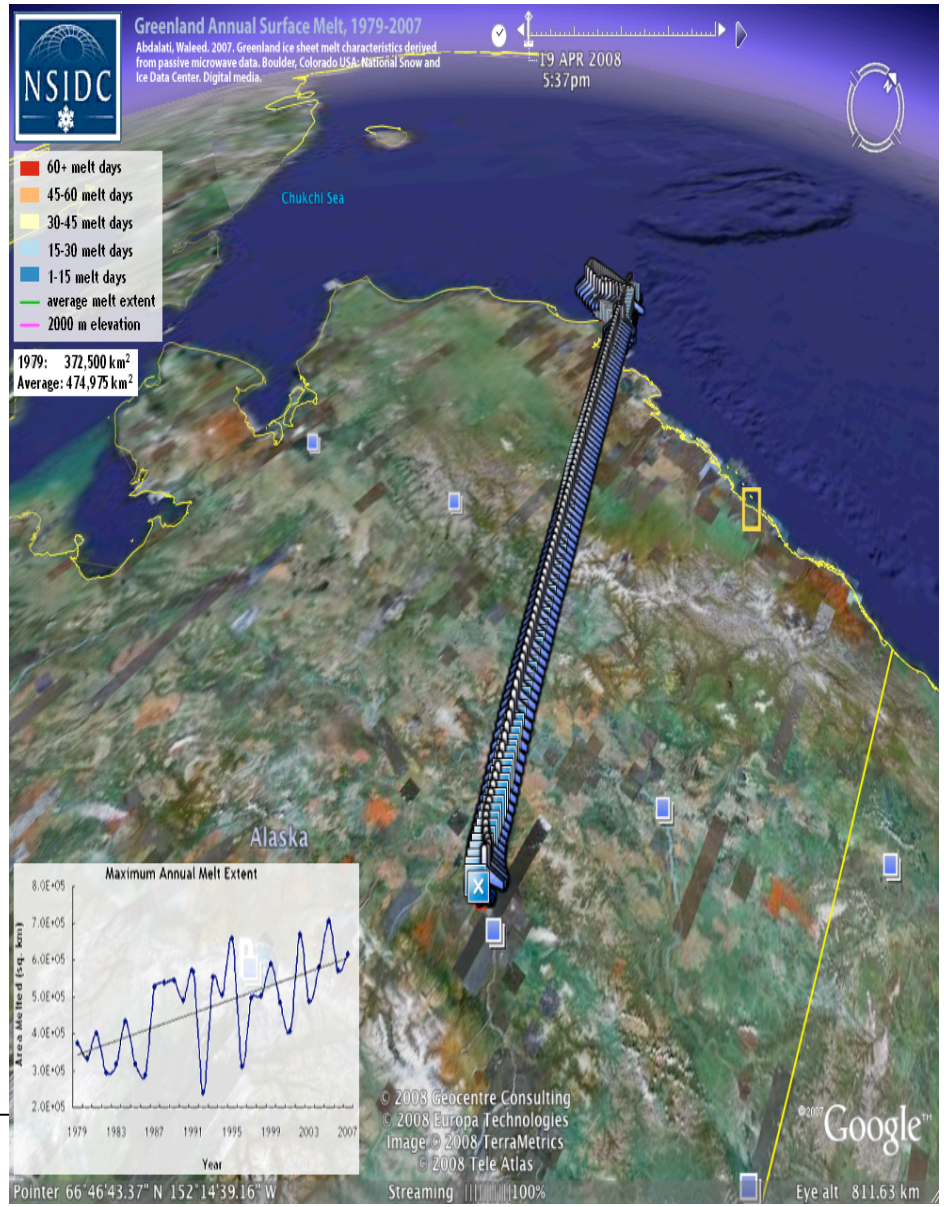
The flight track of the NOAA P3, which sampled the predicted biomass burning plume is also shown off the Northern coast of Alaska.



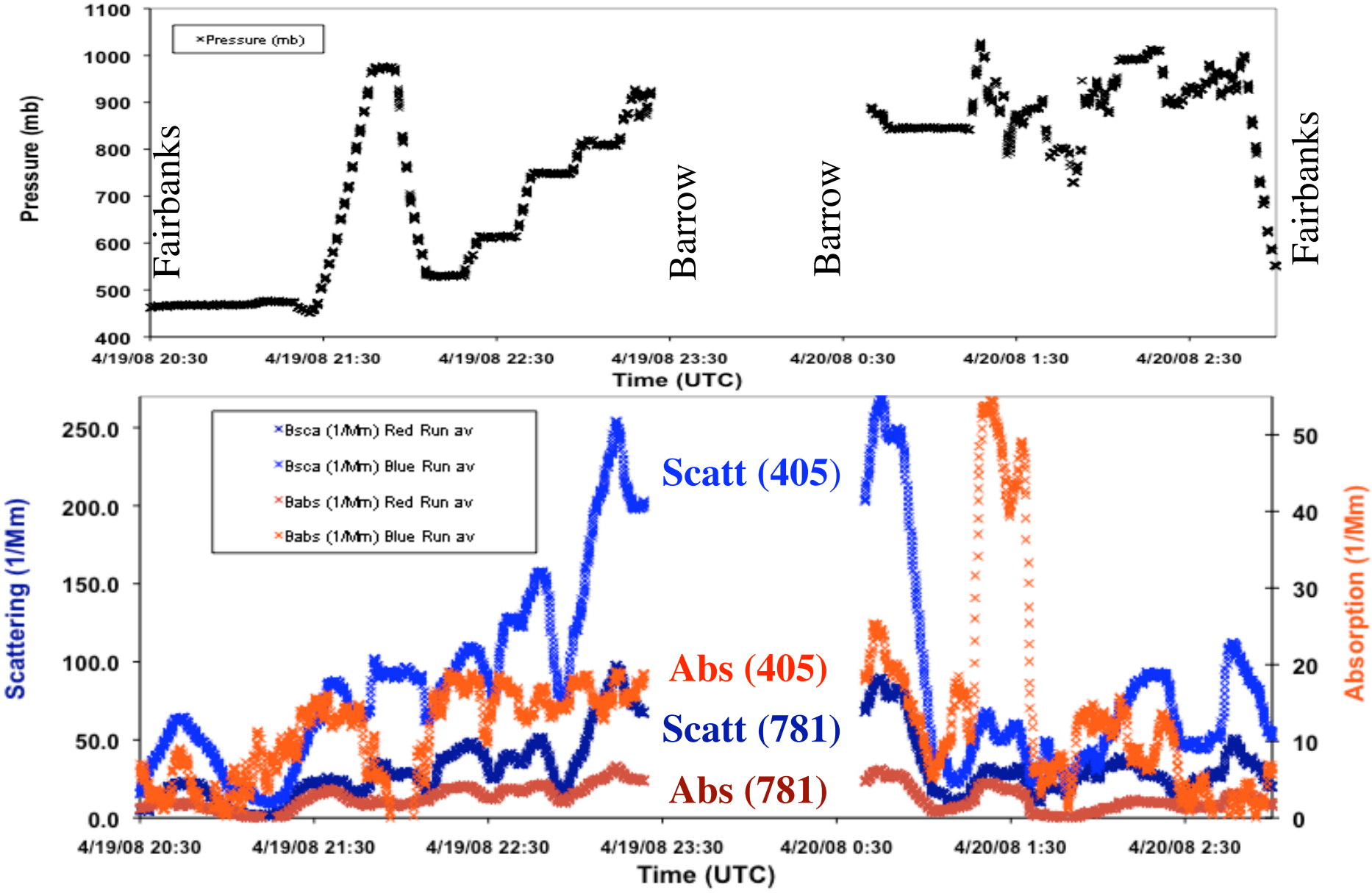
Brad Pierce, NOAA

C-580 Flight: 19th April 2009, 11.30am – 8.30pm





Flight Track Optical Properties: Time Series



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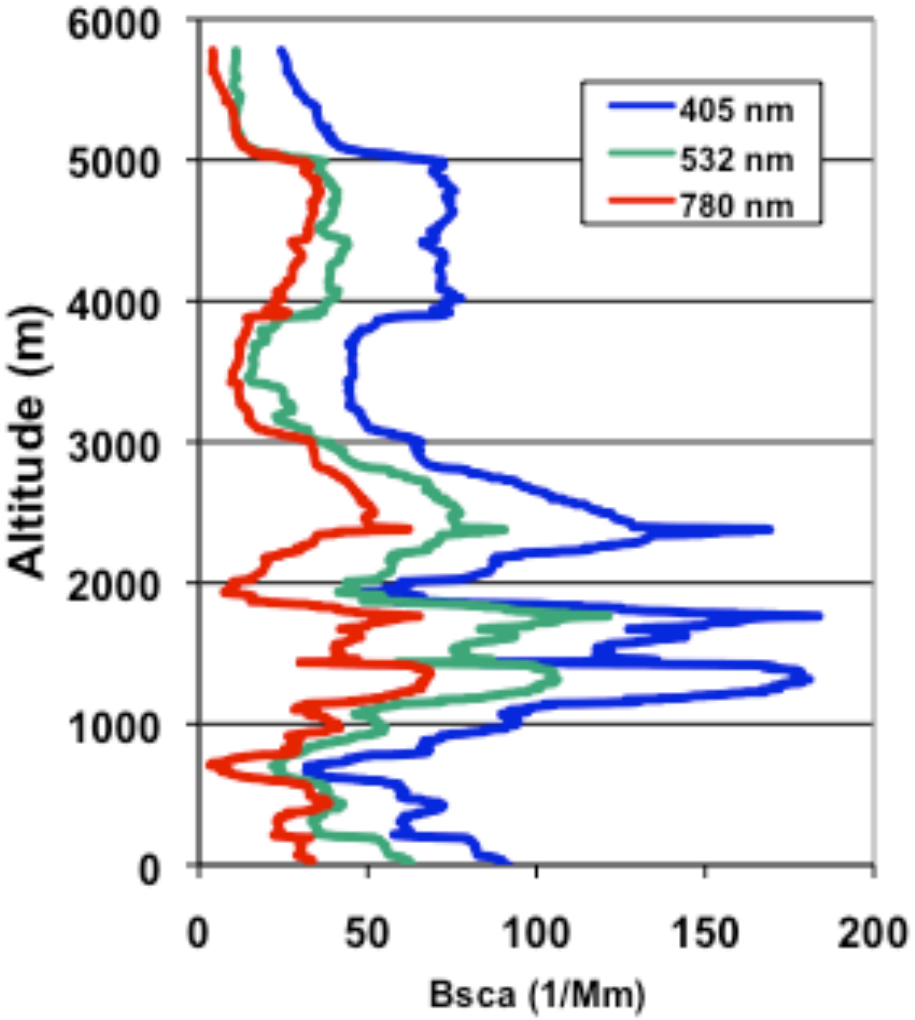
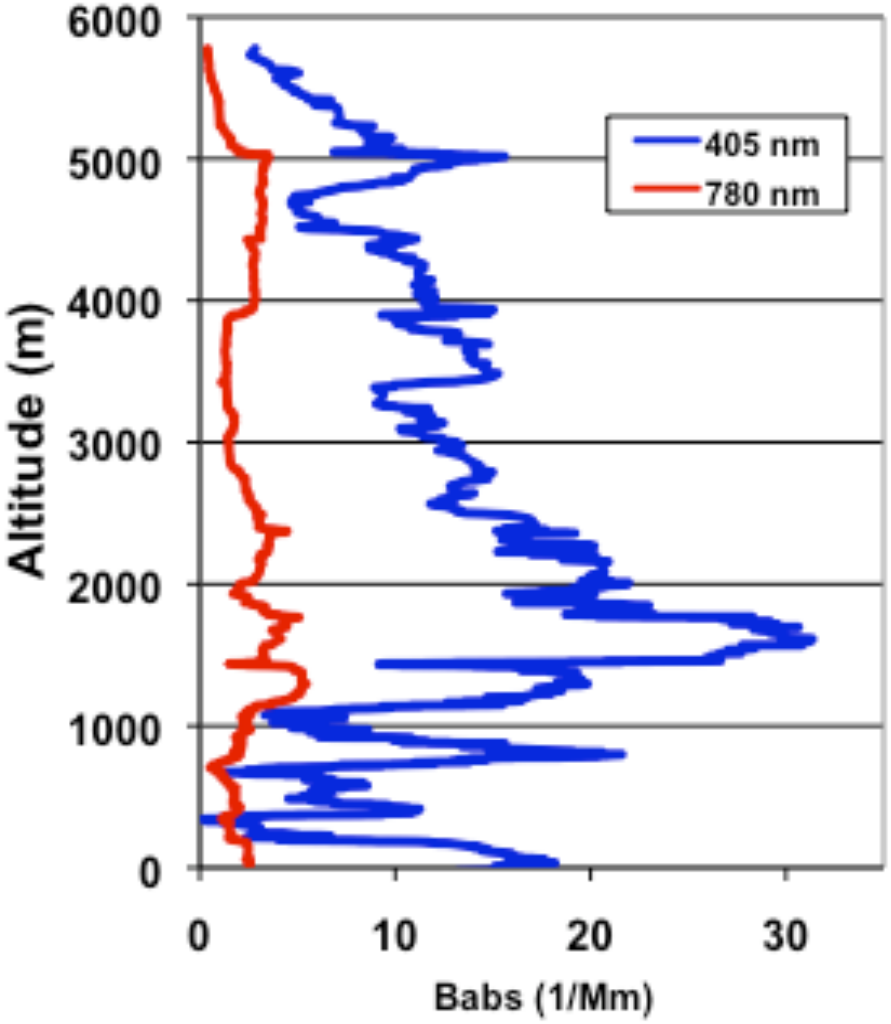
Pollution Layers: Soot, OC, Dust, Sulfate

Scattering Mm^{-1}



Absorption Mm^{-1}



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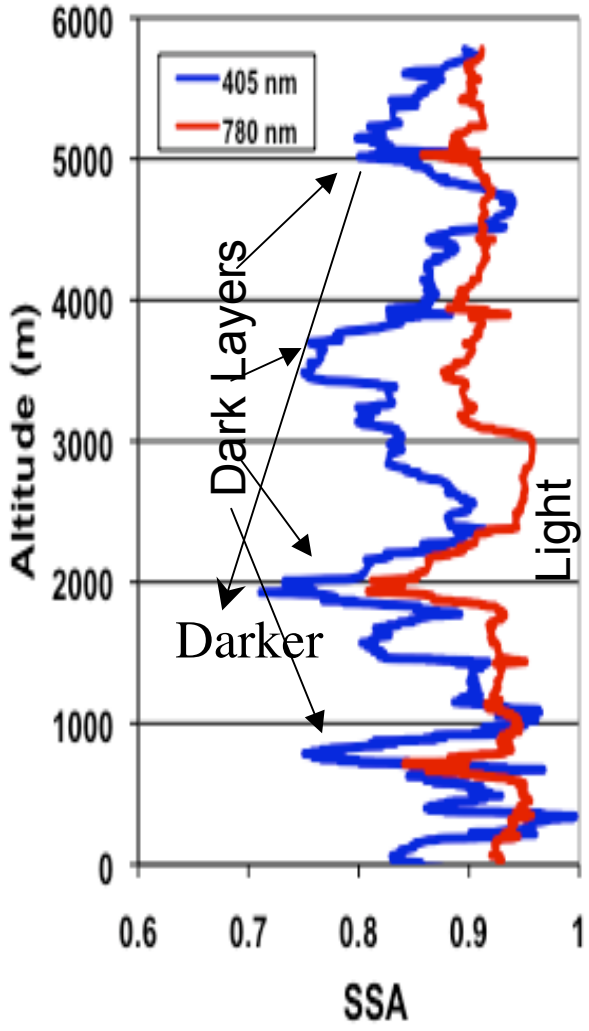
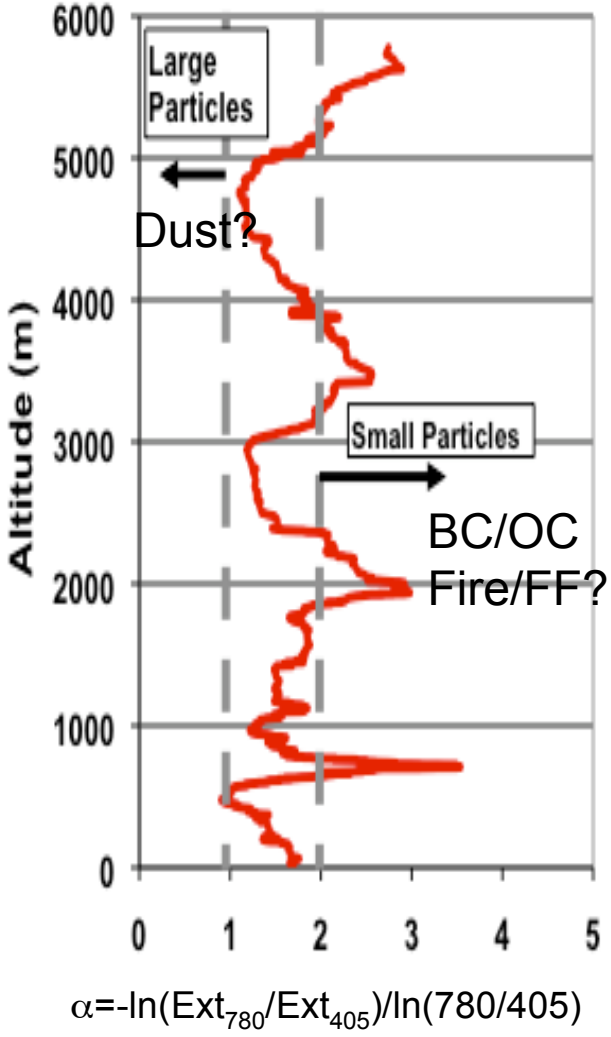
Pollution Layers: Soot, OC, Dust, Sulfate

Single Scatter Albedo

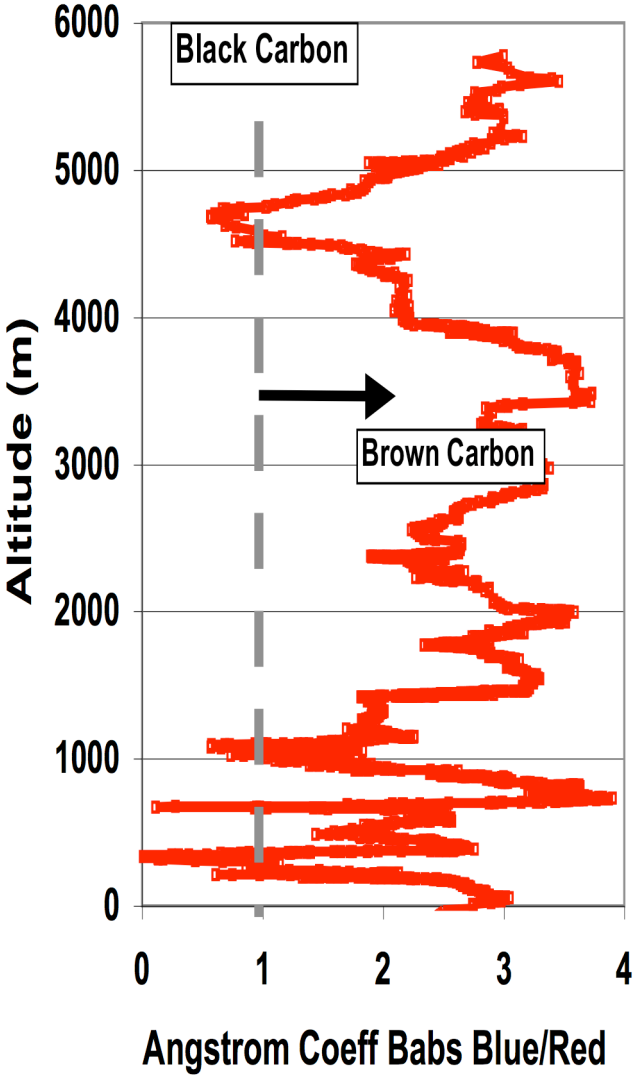


Angstrom Exp (405/780)

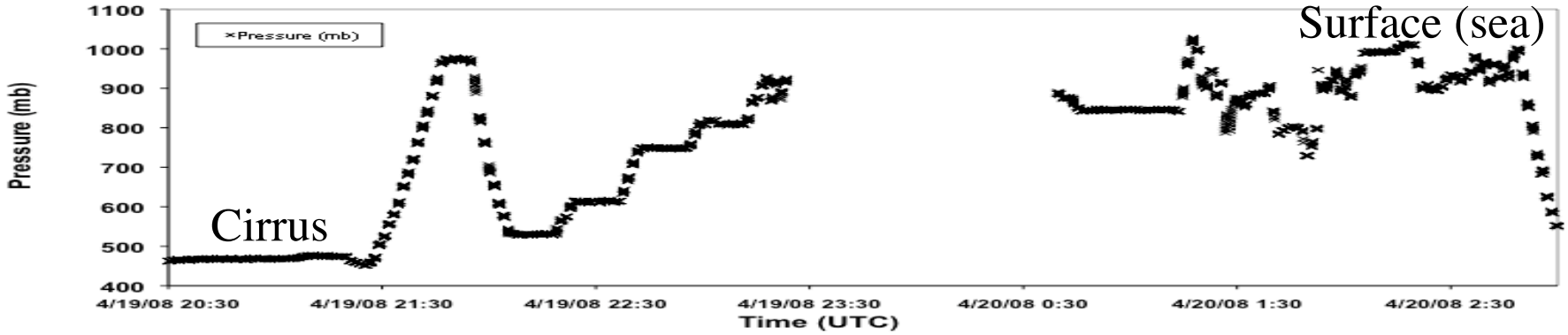
Extinction

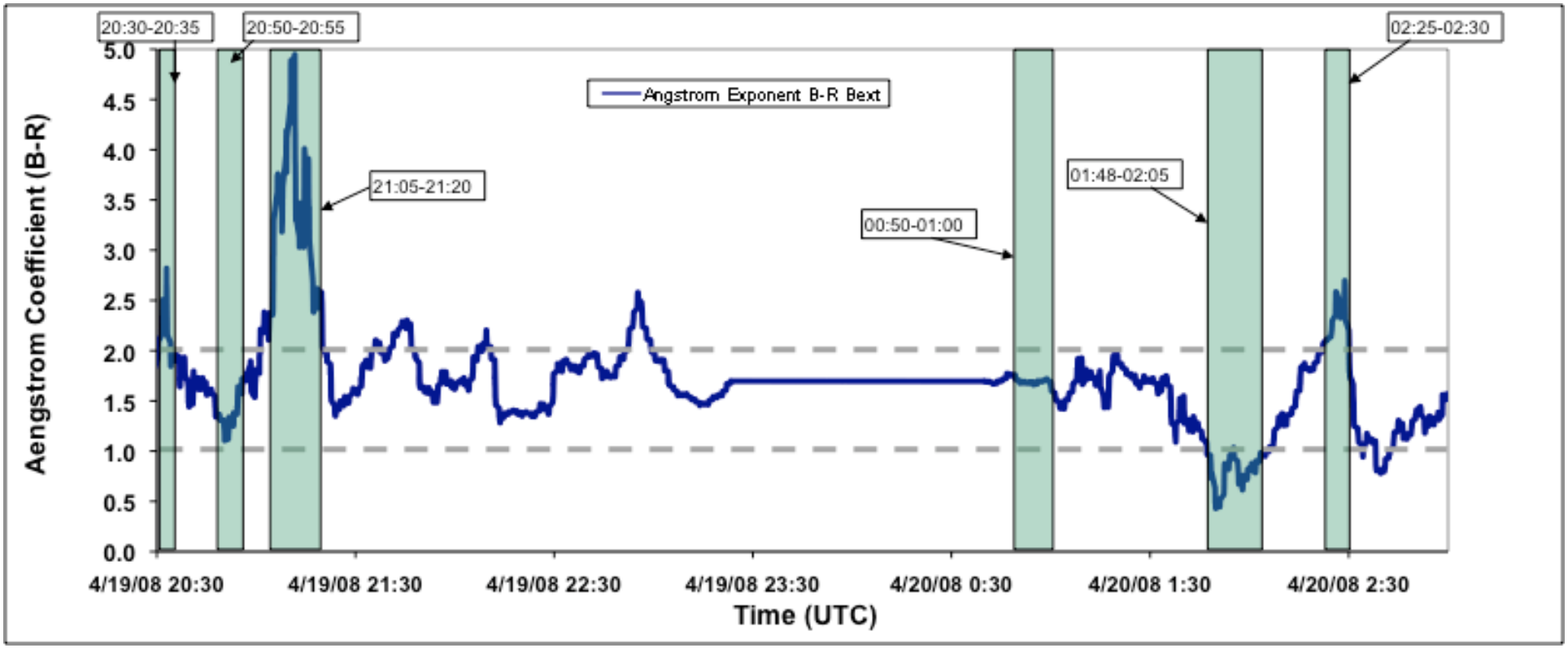


Absorption



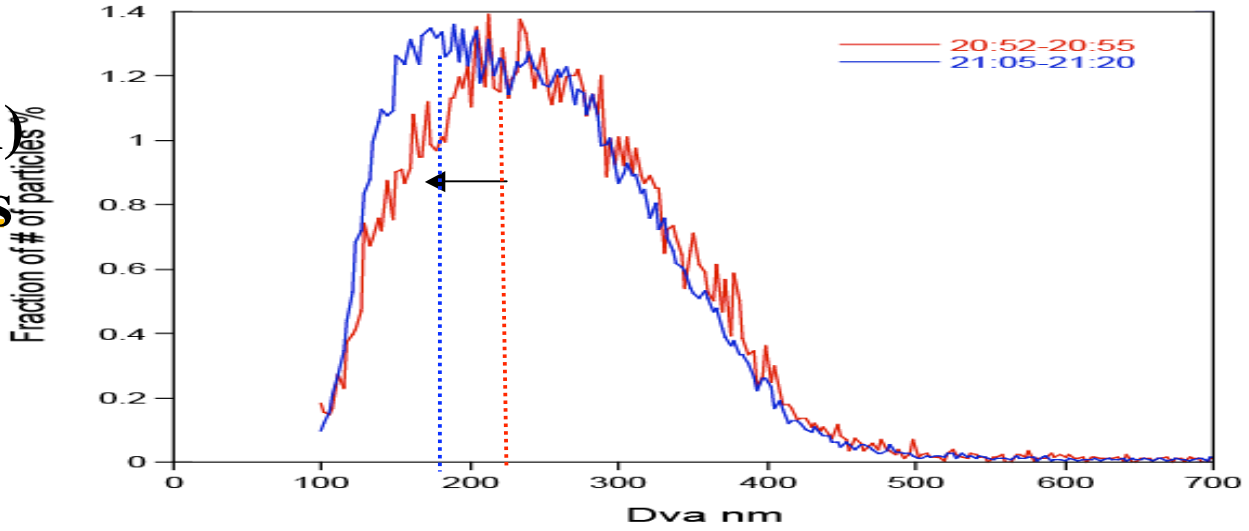
Large Angstrom Exponent Gradients Along Flight Track: April 19

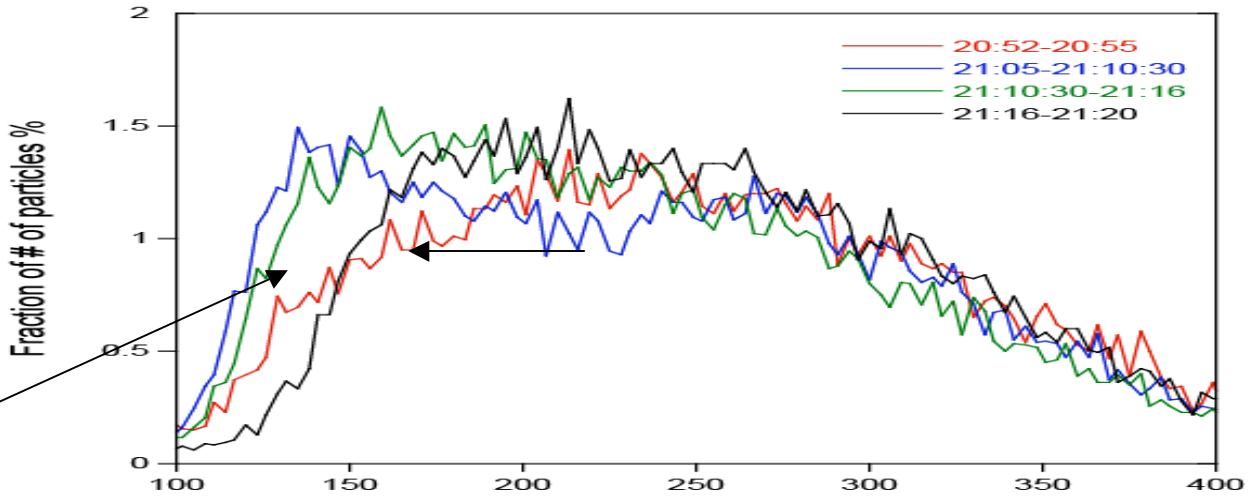


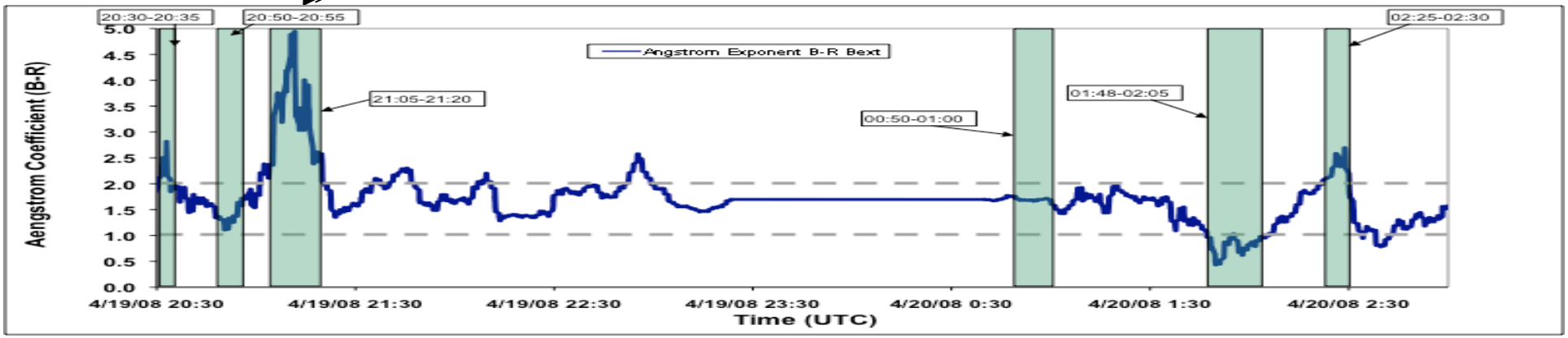


Gradients in Optical Properties (Angstrom) and Size Distributions

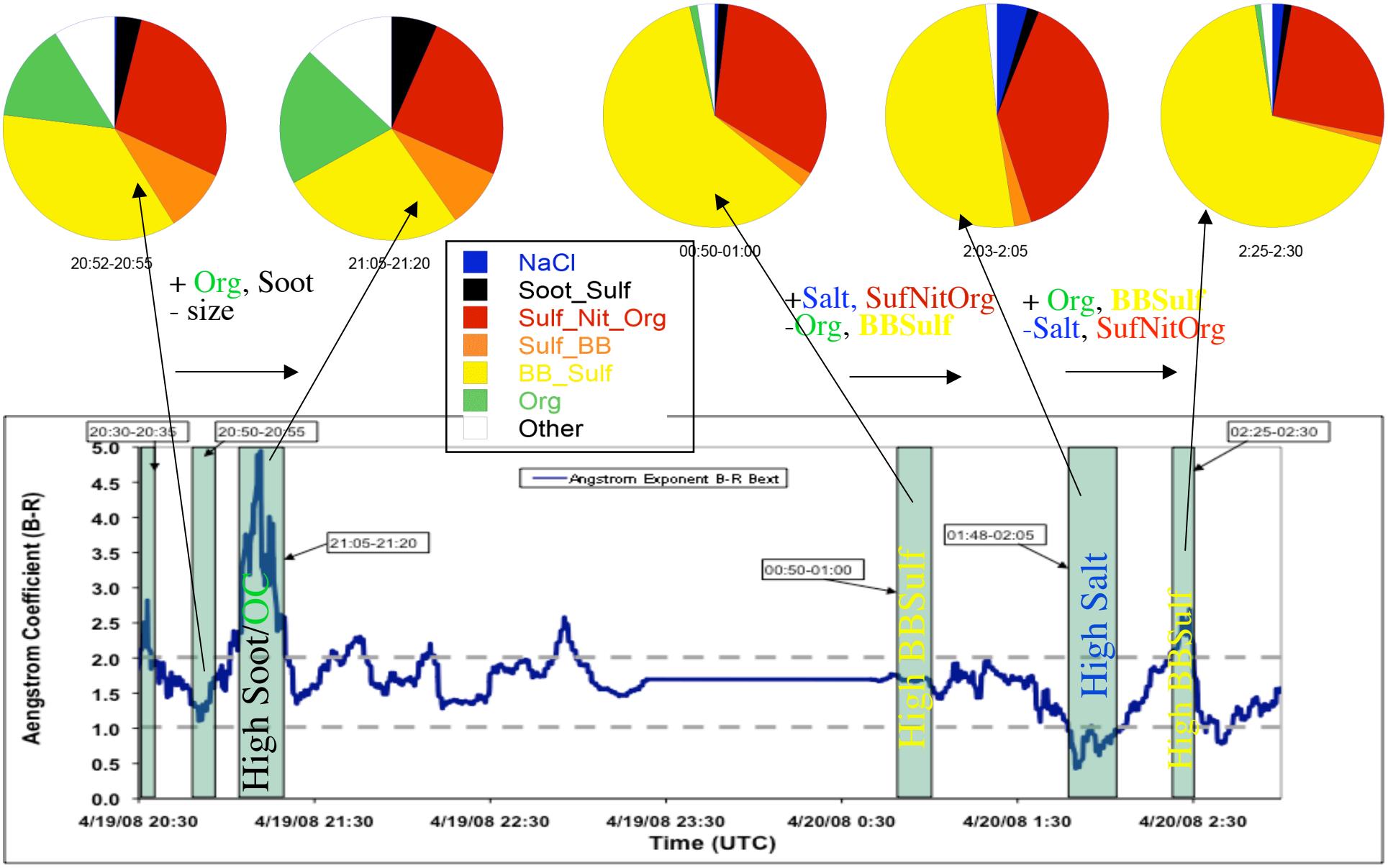
During 21:05-21:20 period when Angstrom exponent increased dramatically (and varied) there were more small particles and size distribution was changing quite rapidly with time





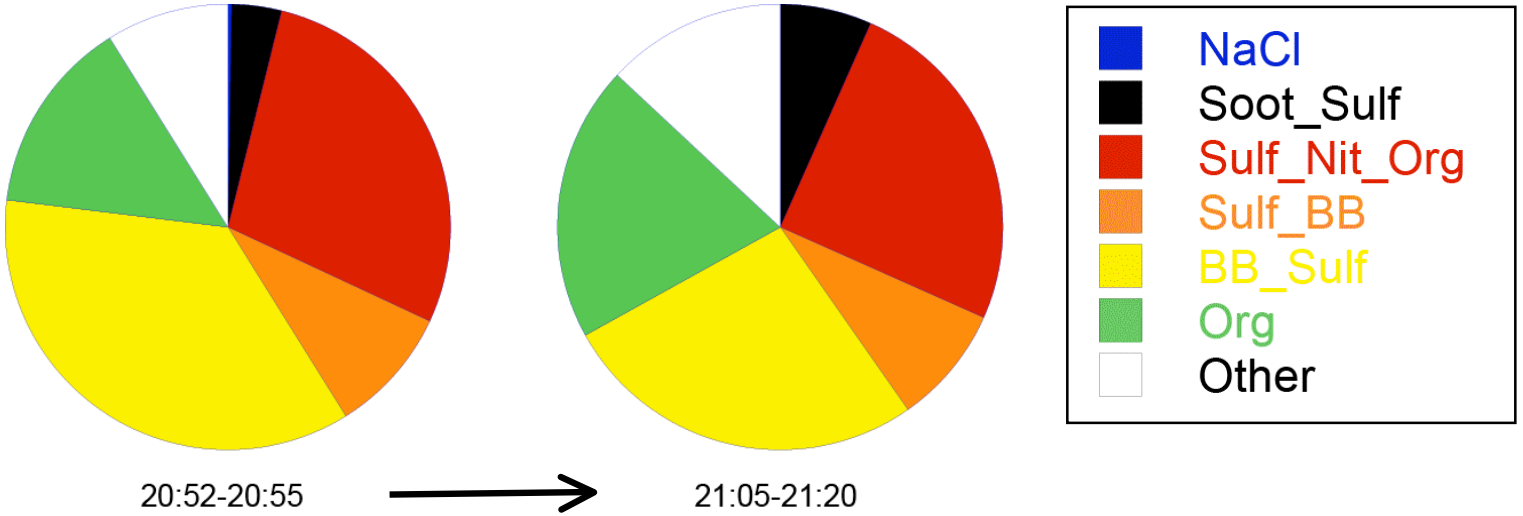


Link Optical Properties (Angstom Gradients) to Chemical Composition Changes



Organic/soot enhancement correlates with smaller particles that are darker in the blue (405 nm)

Soot (~80%) internally mixed with sulfate (20%)
BB internally mixed with sulfate and vice versa



- Increase in Soot & Organic Components (x2)
- Mean diameter decreases from 220nm to 175nm
- Angstrom exponent increases by 2.5
- 405 nm absorption increases x 4, scattering decreases x 1/4
- SSA (405nm) drops from 0.95 to 0.65

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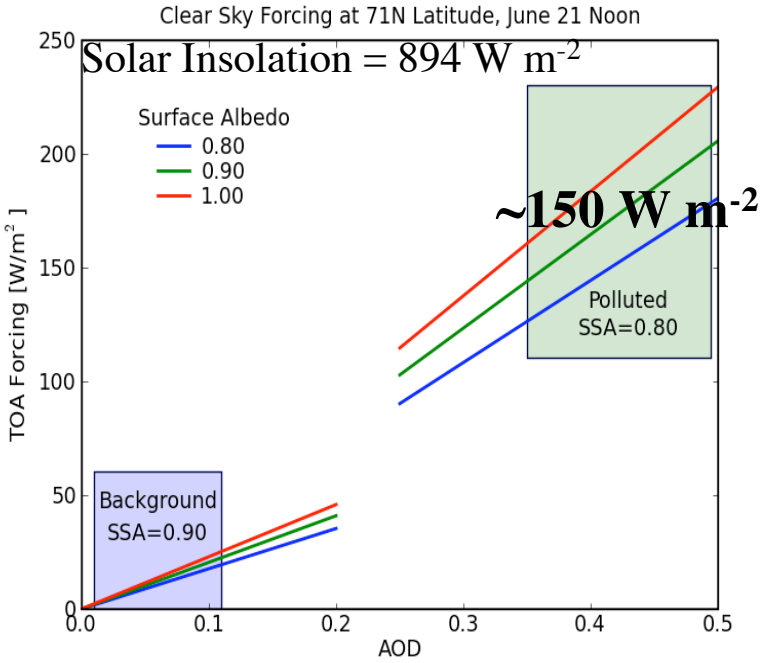
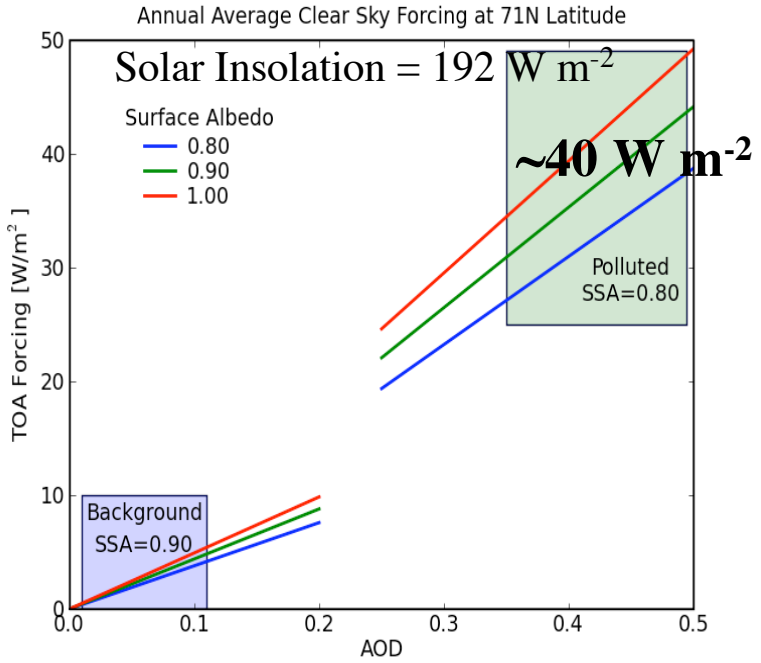
What is clear sky TOA radiative forcing of Arctic Pollution?

S = Solar constant
T = Transmission
N = Cloudiness ~ 1
g = Asymm Par. ~0.7 (AERONET)
w = Single Scatt. Albedo~0.8
a = Surface albedo
τ= Aerosol Optical Depth
• **Chylek GRL, 1995**

$$\Delta F = -\frac{S}{4} T^2 (1-N)\,\tau\,[(1-a)^2(1-g)\omega - 4a(1-\omega)]$$





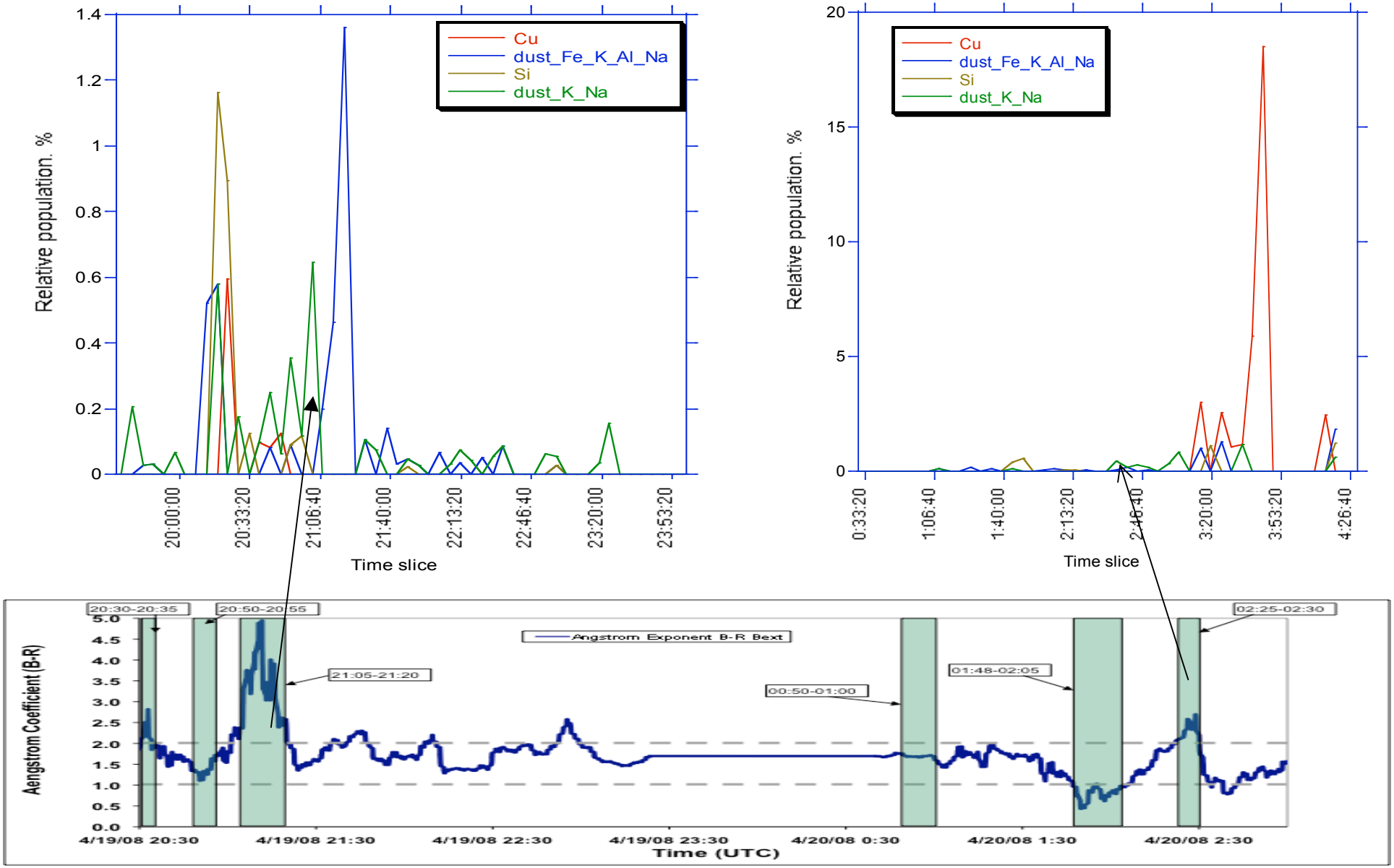
Perturbation ΔGISS^{Mod} is 0.92 Wm^{-2} (Spr.), 0.3 Wm^{-2} (Ann.) *Quinn 08, <1% of our observation*

Pollution (Fires, dust, industrial) events cause transient direct forcings that can last for 10-20 days and are orders of magnitude larger than the mean Arctic aerosol and GHG forcing and can be highly variable.

Conclusions

- Arctic haze optical properties consistent with layers of aged biomass-fires transported over long range.
- Internally mixed Soot-Sulf, Sulf-BB, Sulf-Nit-Org (SPLAT)
- Optical property gradients correlated to changes in chemical composition and size distributions: Need Closure Studies
- Absorption Angstrom exponent suggest “brown carbon”
- Our SSA imply large episodic forcing above Arctic in April that are not captured in current climate change projection models

SPLAT observed dust at high altitude but number concentrations were low (1-2%)



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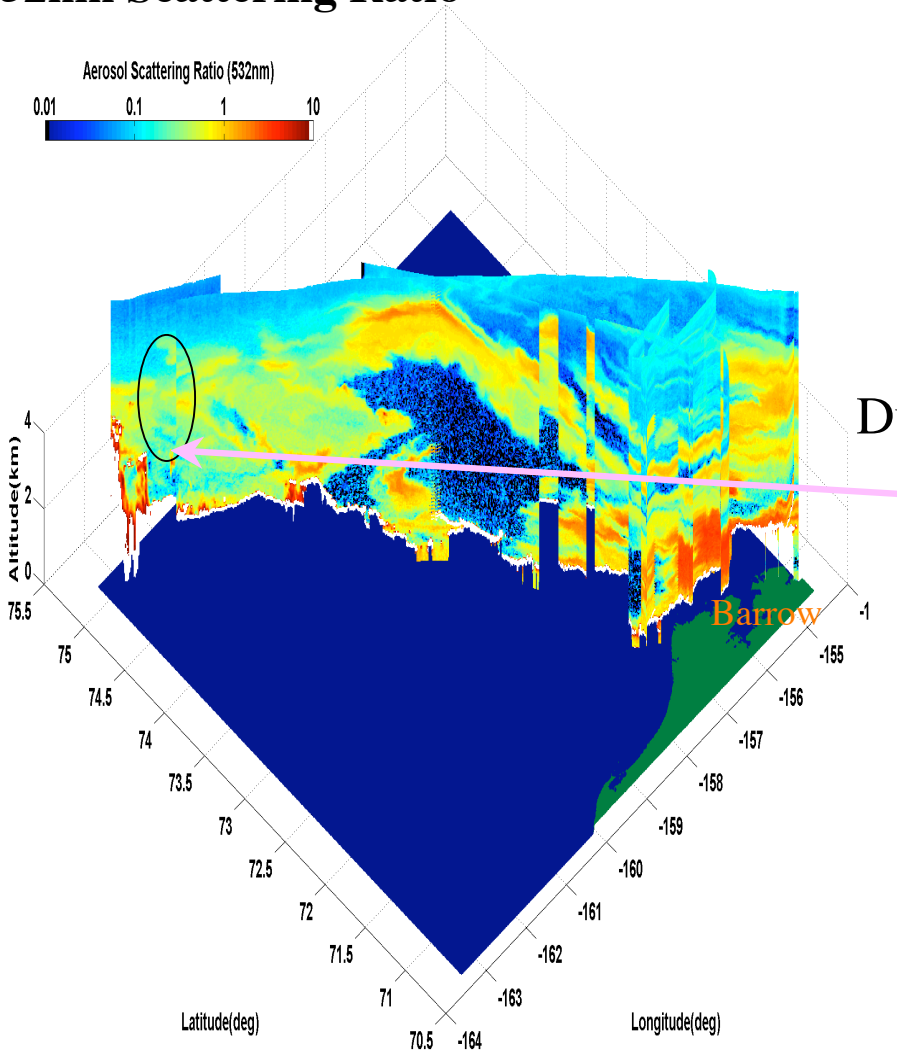
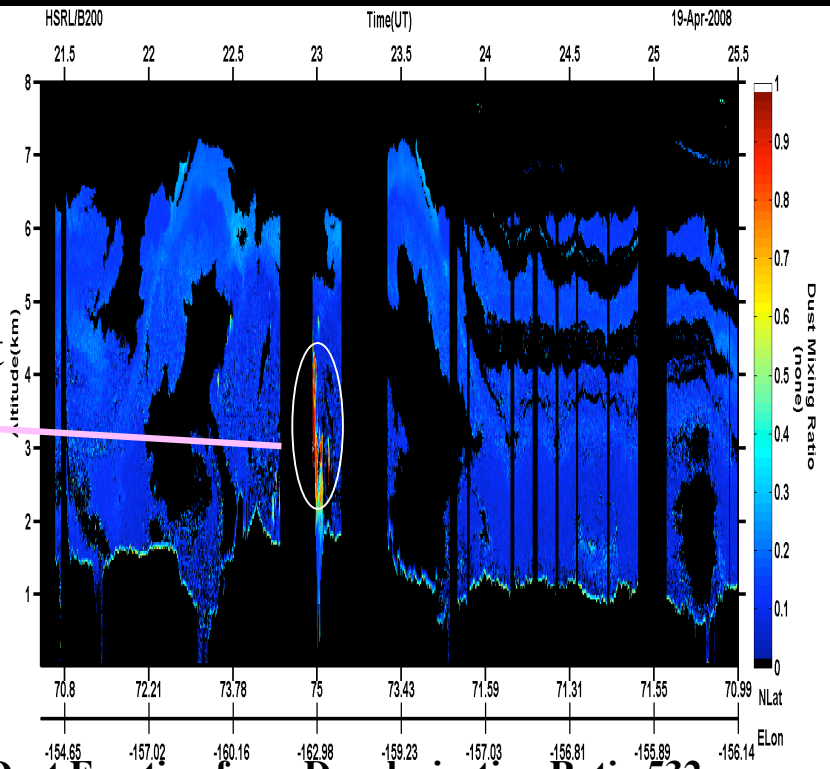
4/19 Pollution NASA HSRL/B-200 Lidar Profiles: Depolarization Derived Dust

532nm Scattering Ratio



Dust



Dust Fraction from Depolarization Ratio 532 nm

Scale: Ground (Barrow), in situ profile, LIDAR, satellite

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Ferrare et al

Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008

C. Warneke,[1,2] R. Bahreini,[1,2] J. Brioude,[1,2] C. A. Brock,[1] J. A. de Gouw,[1,2] D. W. Fahey,[1] K. D. Froyd,[1,2] J. S. Holloway,[1,2] A. Middlebrook,[1] L. Miller,[2,3] S. Montzka,[3] D. M. Murphy,[1] J. Peischl,[1,2] T. B. Ryerson,[1] J. P. Schwarz,[1,2] J. R. Spackman,[1,2] and P. Veres[1,2]

BB CO Plume Fires, Flexpart

MODIS fire

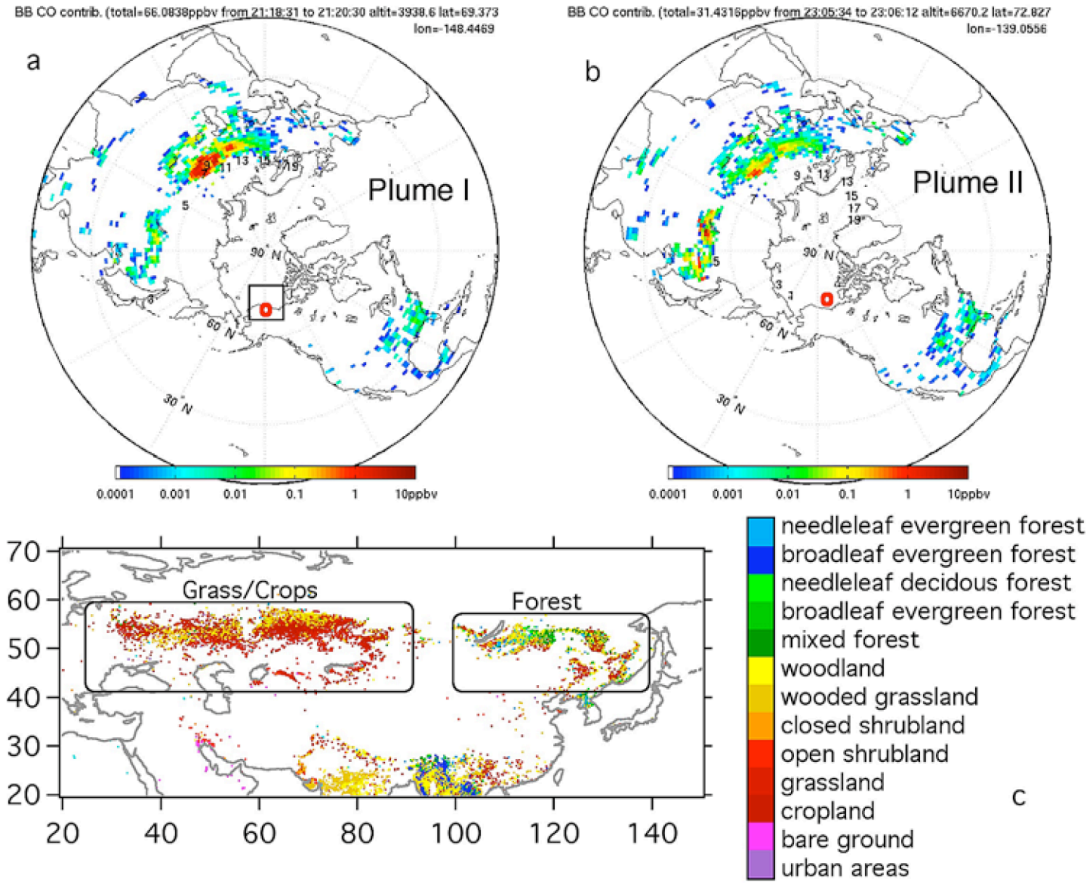

Figure 2. (a) and (b) FLEXPART BB CO source contribution for plume I and II in Figure 1 on a logarithı square in Figure 2a indicates the area where the ARCPAC flights took place and all the BB plumes were observe fire detection between April 1, 2008 and April 20, 2008. The color code indicates the dominant land cover c

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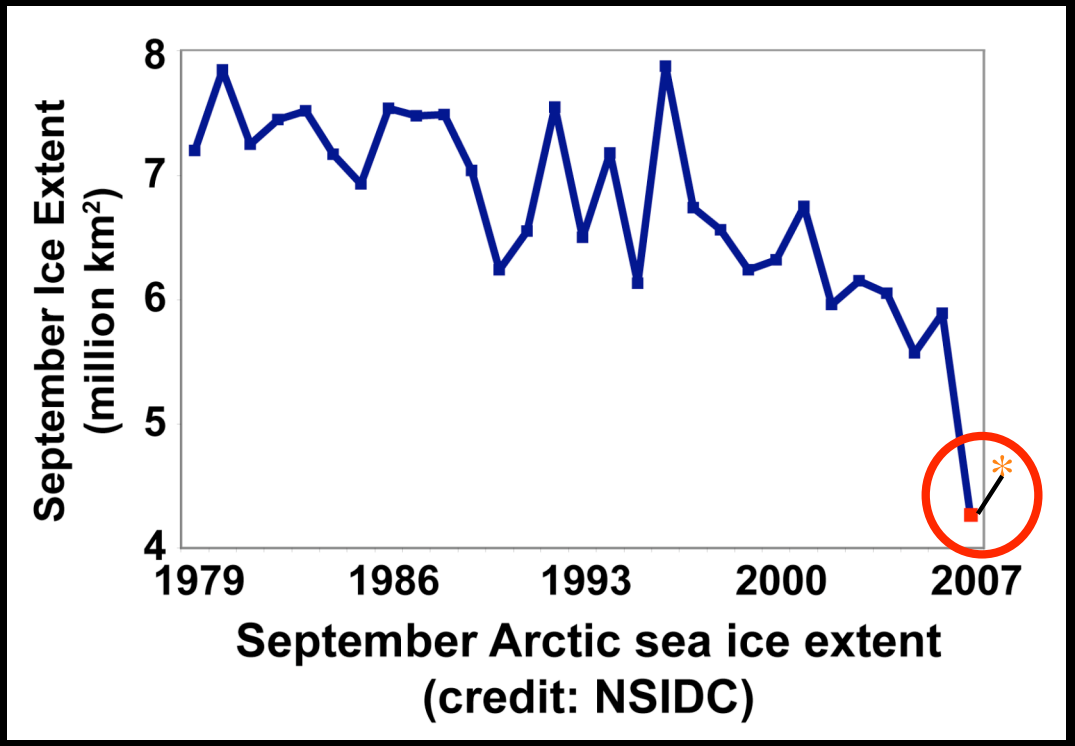
2007-08 Record Minima in Sept. Arctic Sea Ice Extent



Additional open ocean = Texas + Alaska

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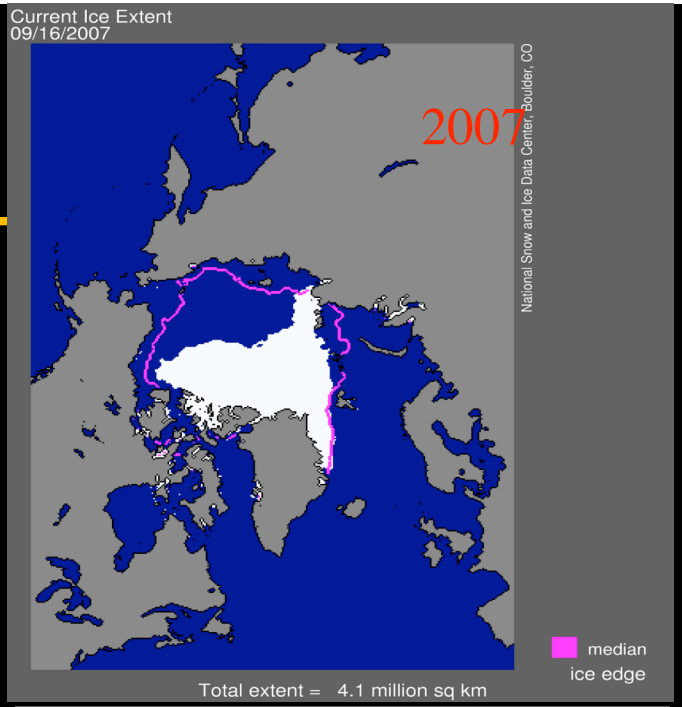

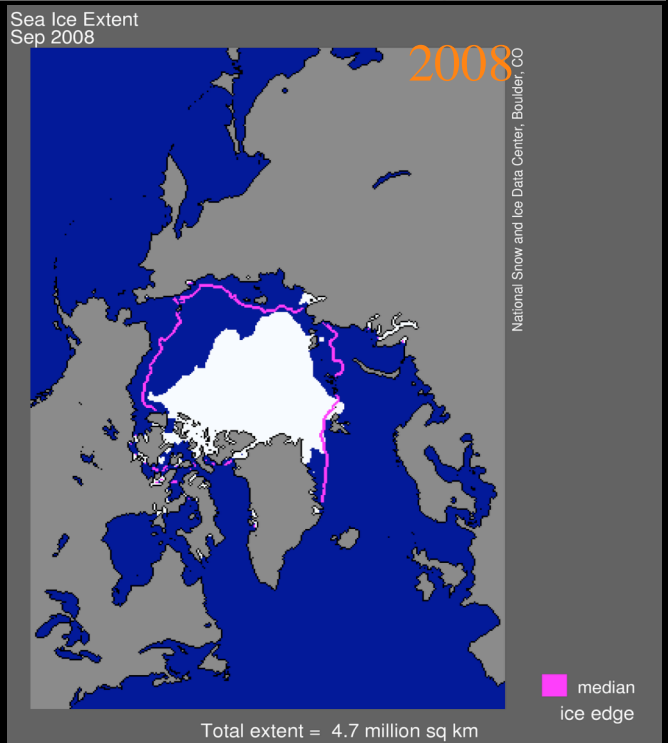

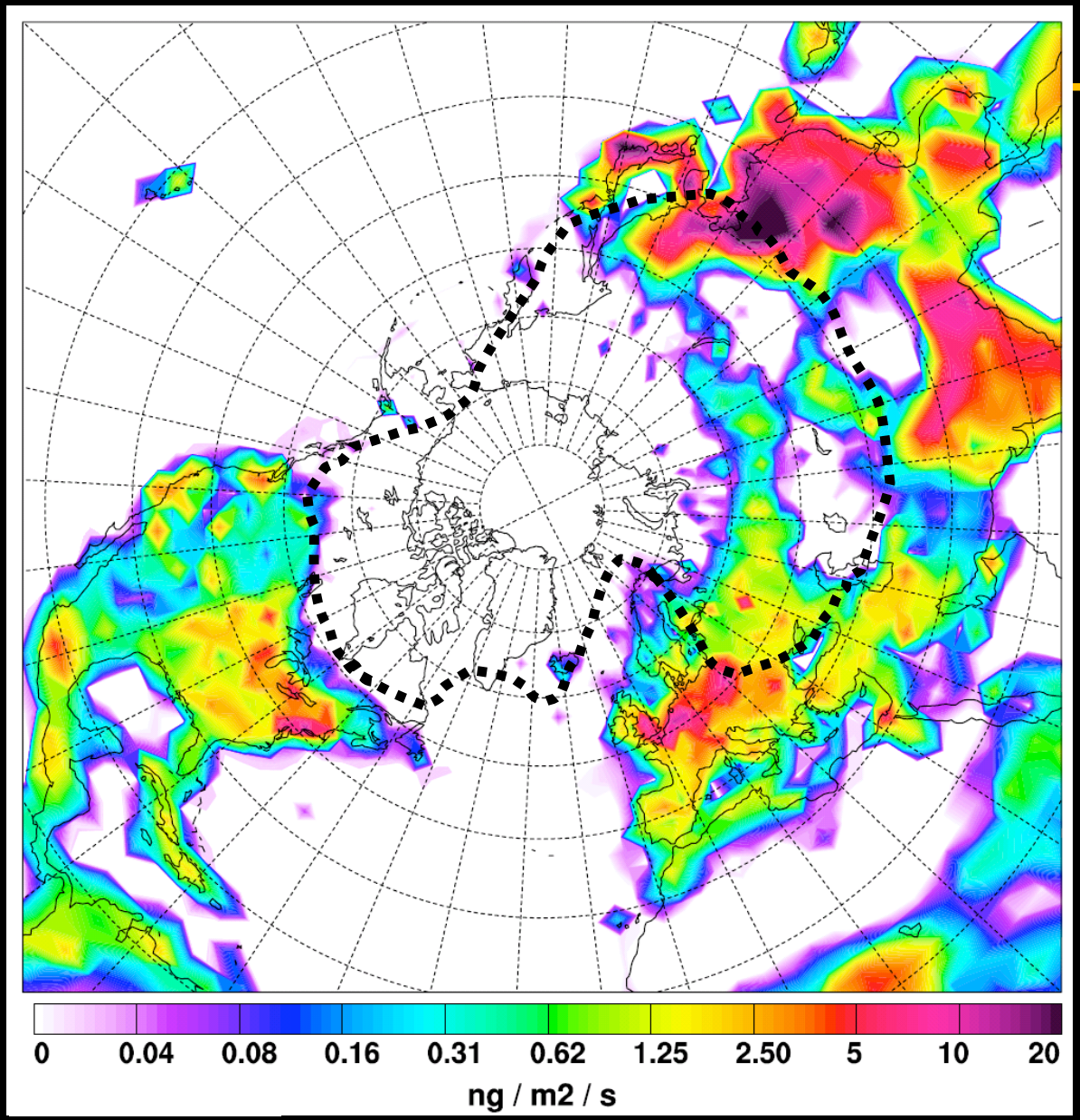

Anthropogenic sources of soot (industrial, biofuel, fires)

Sources in northern Europe and NE China are consistently within or near the mean position of the Arctic front.

Stohl et al., 2006

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Challenges for Arctic Assessments

- Model transport of long-range pollutants into Arctic
- Capture variability in Arctic dynamics of fires/dust
- Is fossil/biofuel energy the major human addition?
- Is Arctic oil-gas exploration affecting Siberian fires?
- How will NW passage shipping exacerbate pollution?
- Reliable baseline and pollution emissions scenarios.

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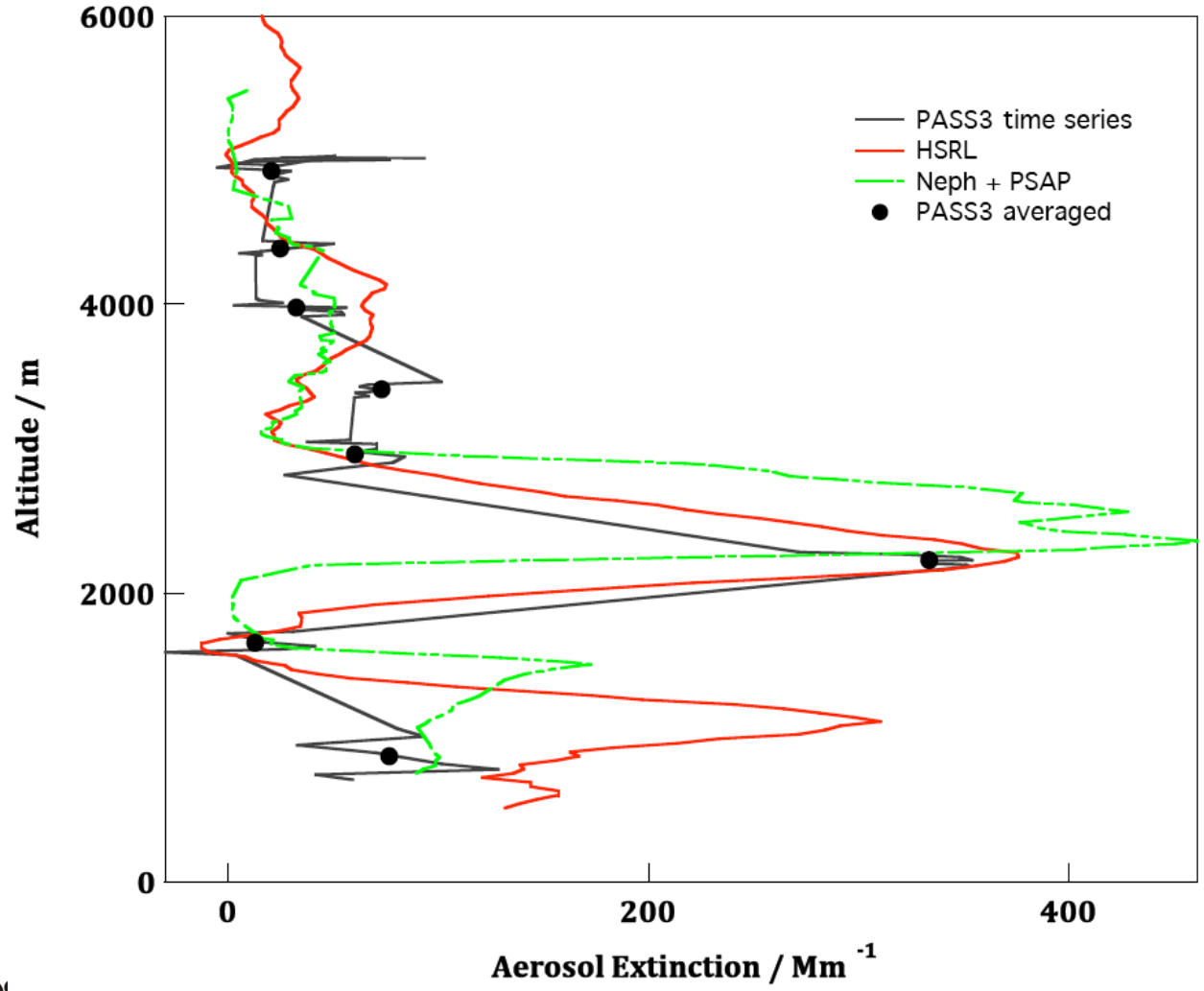

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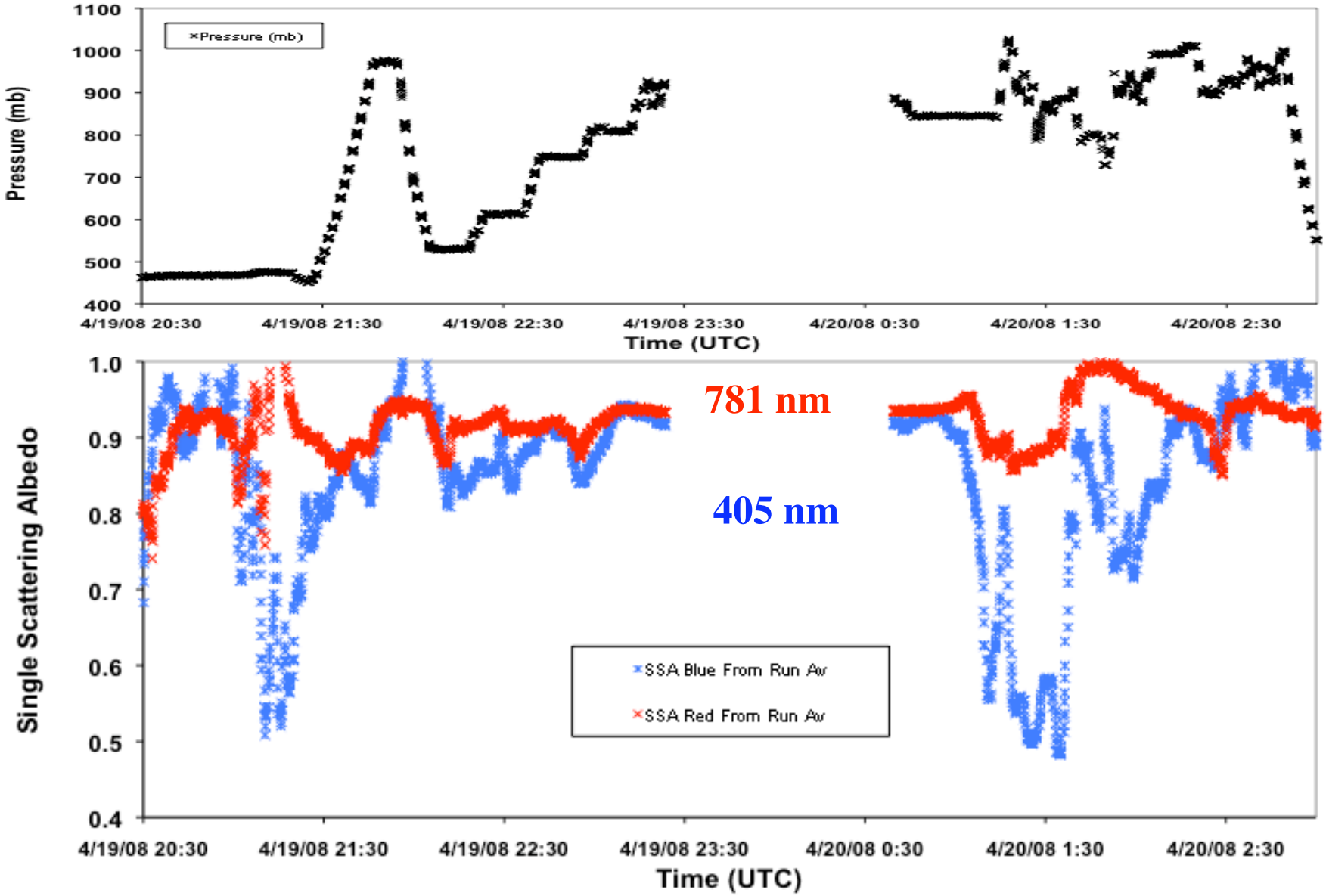
Flight Track Single Scatter Albedo: Time Series



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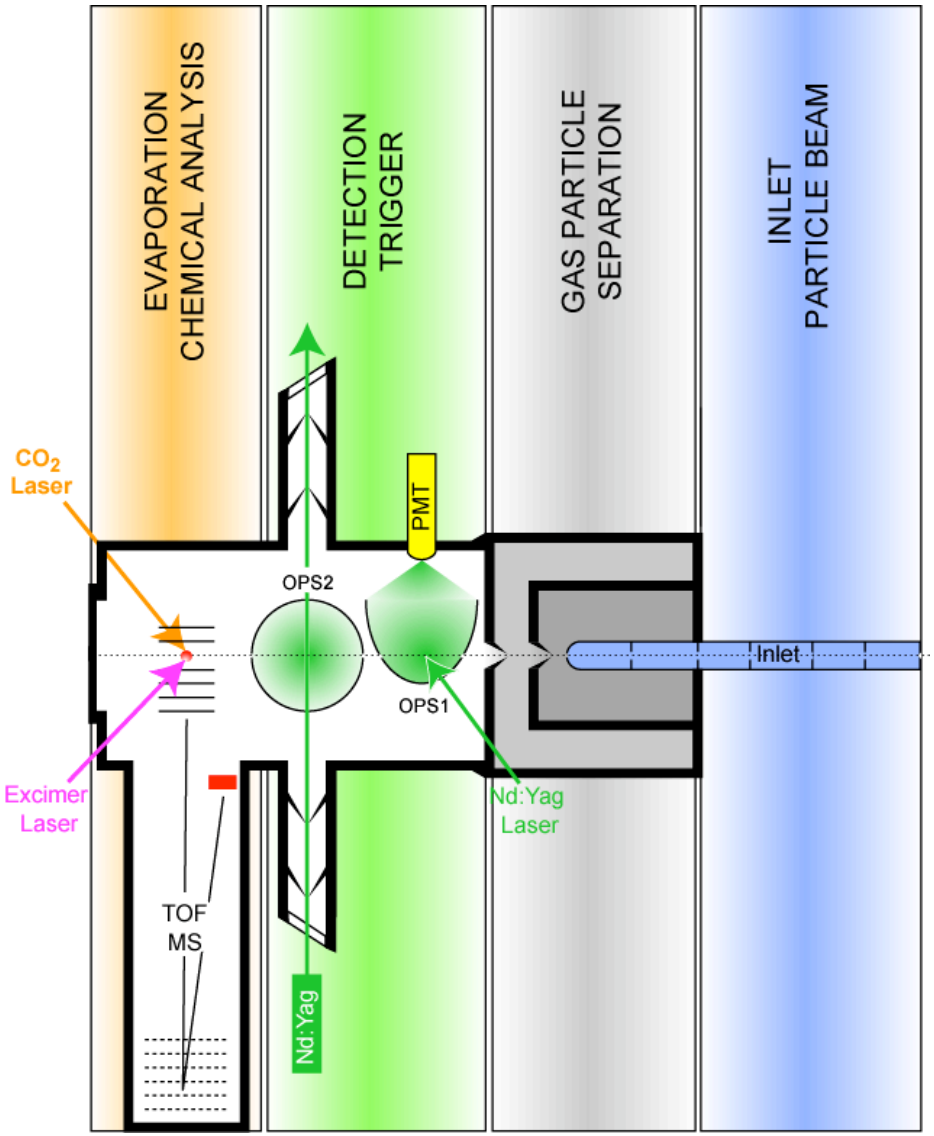
SPLAT II: An Ultra-Sensitive, High-Precision Single Particle Mass Spectrometer (PNNL)



- Provides in *Real-time* the size and internal composition of individual 50 nm to 3 µm particles
- High sensitivity: detects 1p/sec for an aerosol sample of 1p/cm^3 with d>125 nm
- High sensitivity to small particles: detects 40% of 100 nm particles
- Sampling rate: sizes up to 500 p/sec, 100 of which are also chemically characterized
- Measures refractory and non-refractory aerosol fractions in each particle
- Measures aerodynamic size with better than 1% accuracy

Large Pollution in Polar Regions

(ISDAC 19 April 2008)

Layer of Arctic Haze