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# Photoacoustic Observations of Aerosol Optical Properties Aloft Alaska: Quantifying Arctic Radiative Forcing

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ARM Meeting March 31 2009, Louisville KY

# Outline

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- **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).*

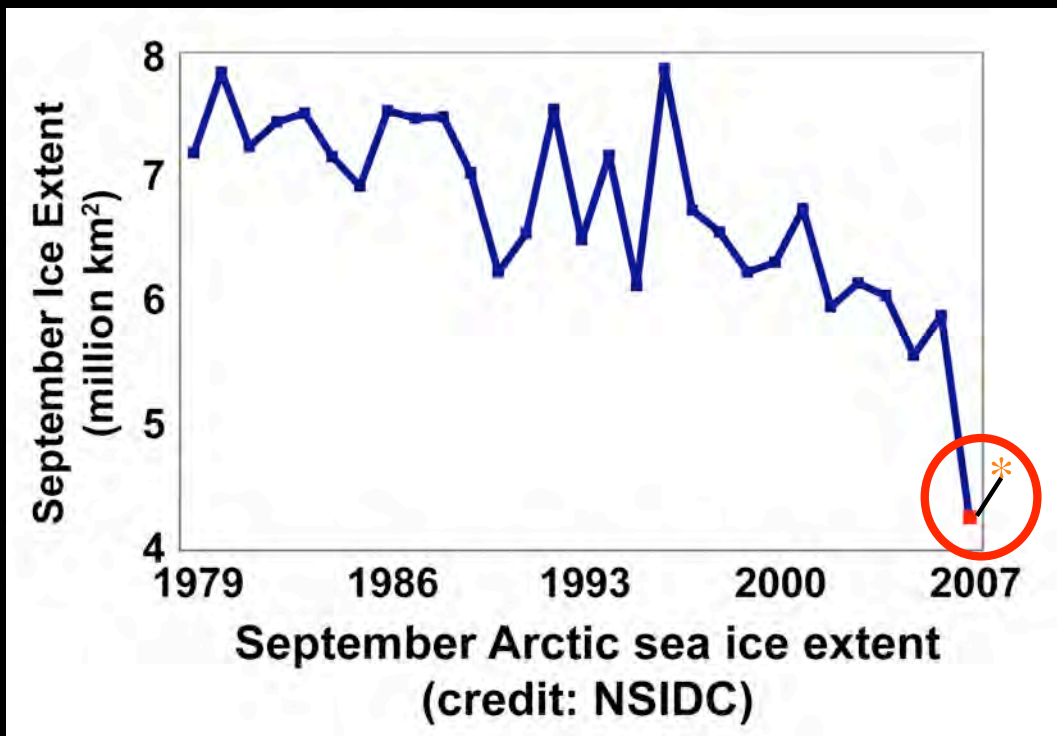
- **Key Objective**

*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.*

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



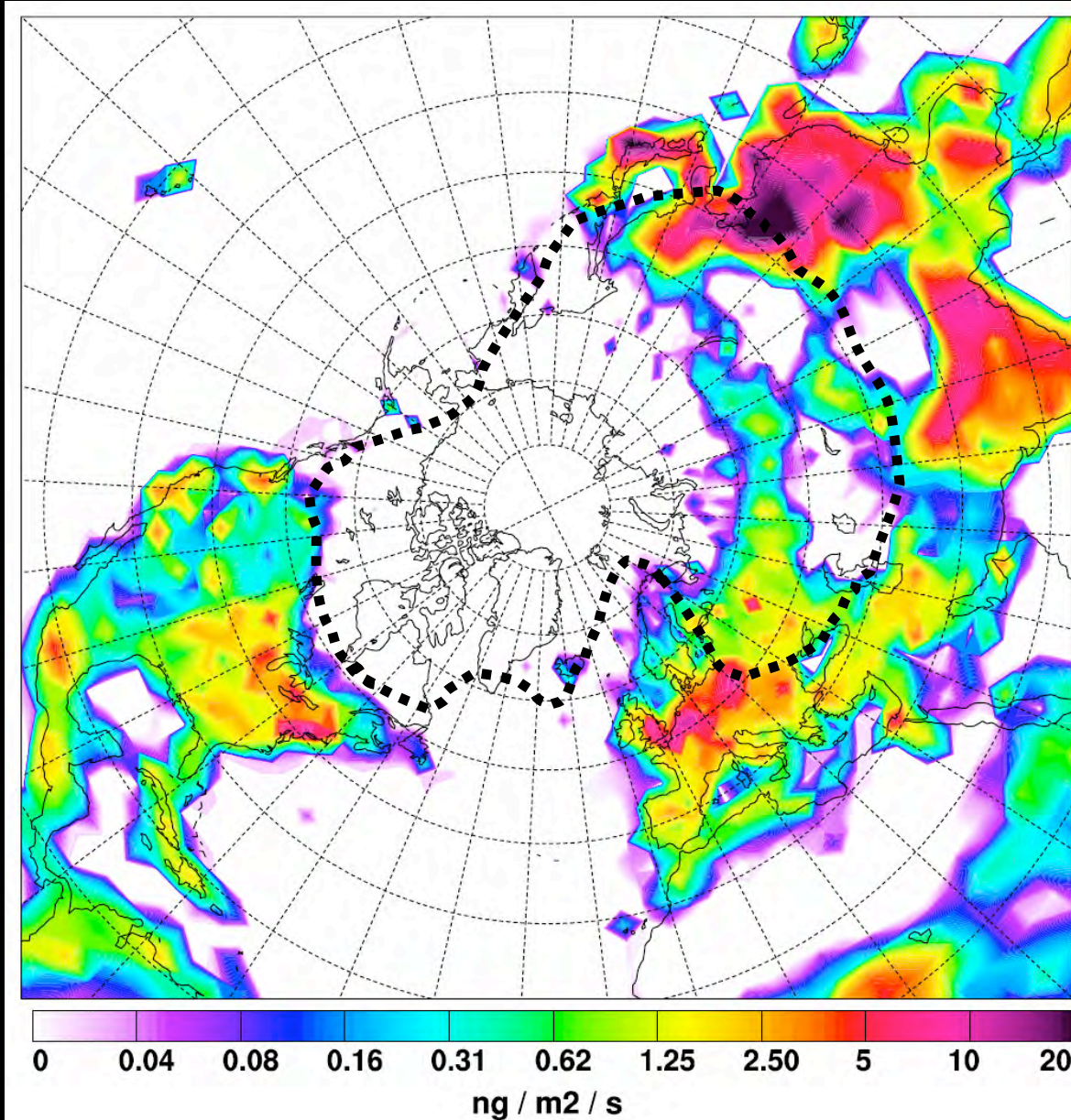
*Additional open ocean = Texas + Alaska*



## **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.**

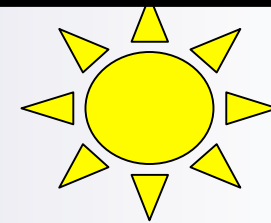
*Stohl et al., 2006*



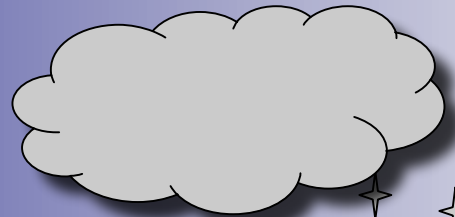
NASA GISS

# SPRING: INSTANTANEOUS AEROSOL FORCING

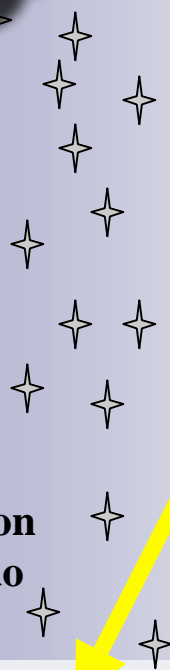
Add Fossil + Biofuel to current Biomass Burning



$$F^{TOA} = 0.9 \text{ W m}^{-2}$$



$$F^{ATM} = 1.6 \text{ W m}^{-2}$$



Aerosol Direct

$$F^S = -0.7 \text{ W m}^{-2}$$

Black carbon  
Snow albedo  
+  $\Delta T$

$$F^{BCsnow} = 0.53 \text{ W m}^{-2}$$

$$\Delta T_s = -0.93 \text{ C}$$

$$\Delta T_s = +0.6 \text{ C}$$

Quinn ACP 2008

Hansen CD 2007



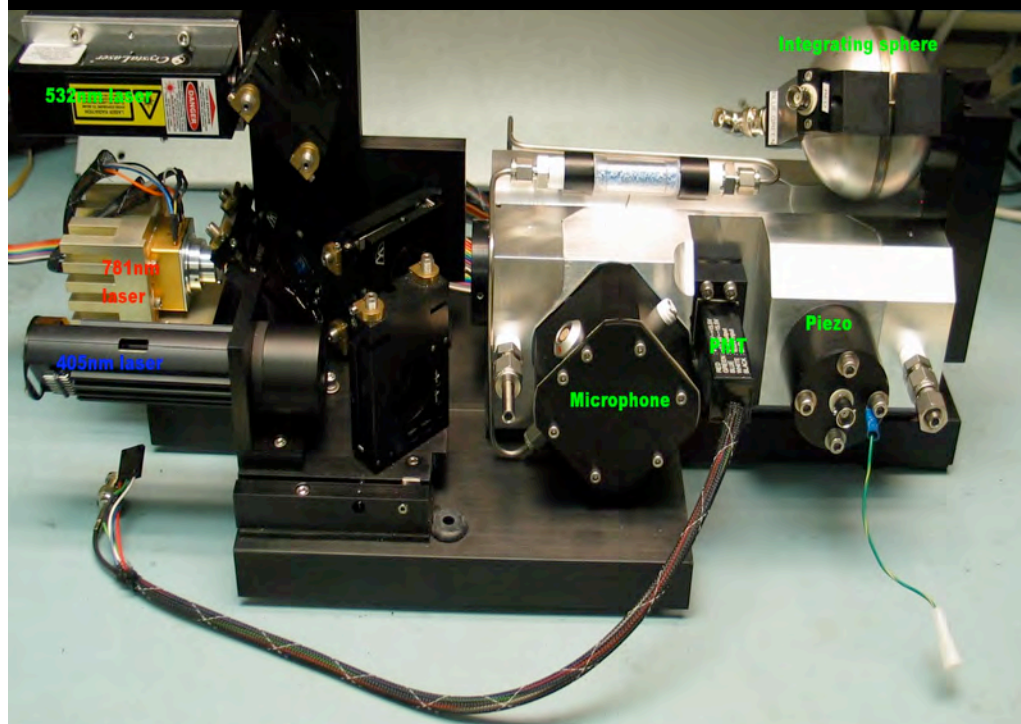
# Challenges for Arctic Assessments

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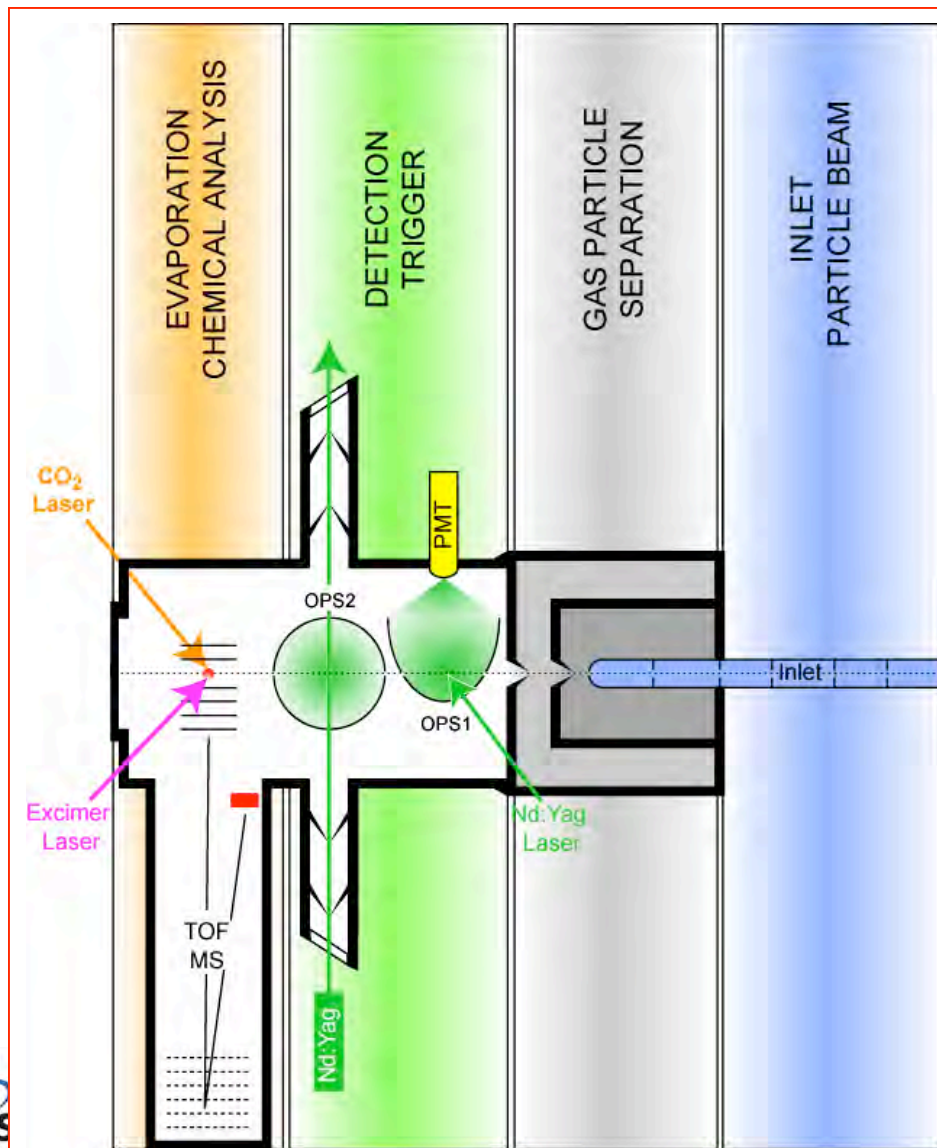
- Model transport of long-range pollutants into Arctic
- Capture variability in Arctic dynamics of fires/dust
- Is fossil/biofuel energy the major human addition?
- Is Arctic oil-gas exploration affecting Siberian fires?
- How will NW passage shipping exacerbate pollution?
- Reliable baseline and pollution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

# 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)?



# 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  $\mu\text{m}$  particles
- High sensitivity: detects 1p/sec for an aerosol sample of  $1\text{p}/\text{cm}^3$  with  $d > 125\text{ nm}$
- 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

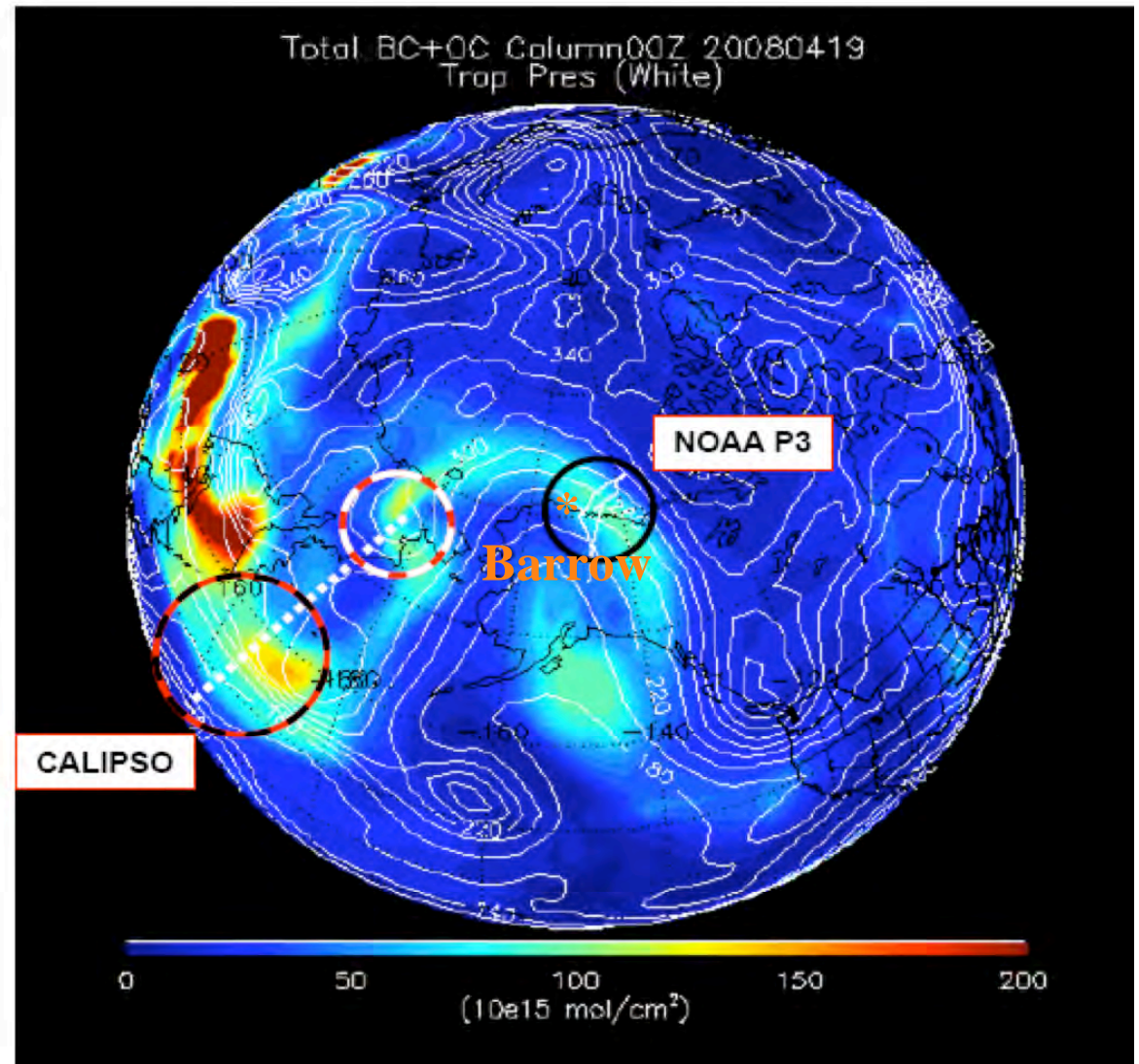


RAQMS total column BC+OC (x10<sup>15</sup> mol/cm<sup>2</sup>) analysis at 00Z on April 19th, 2008.

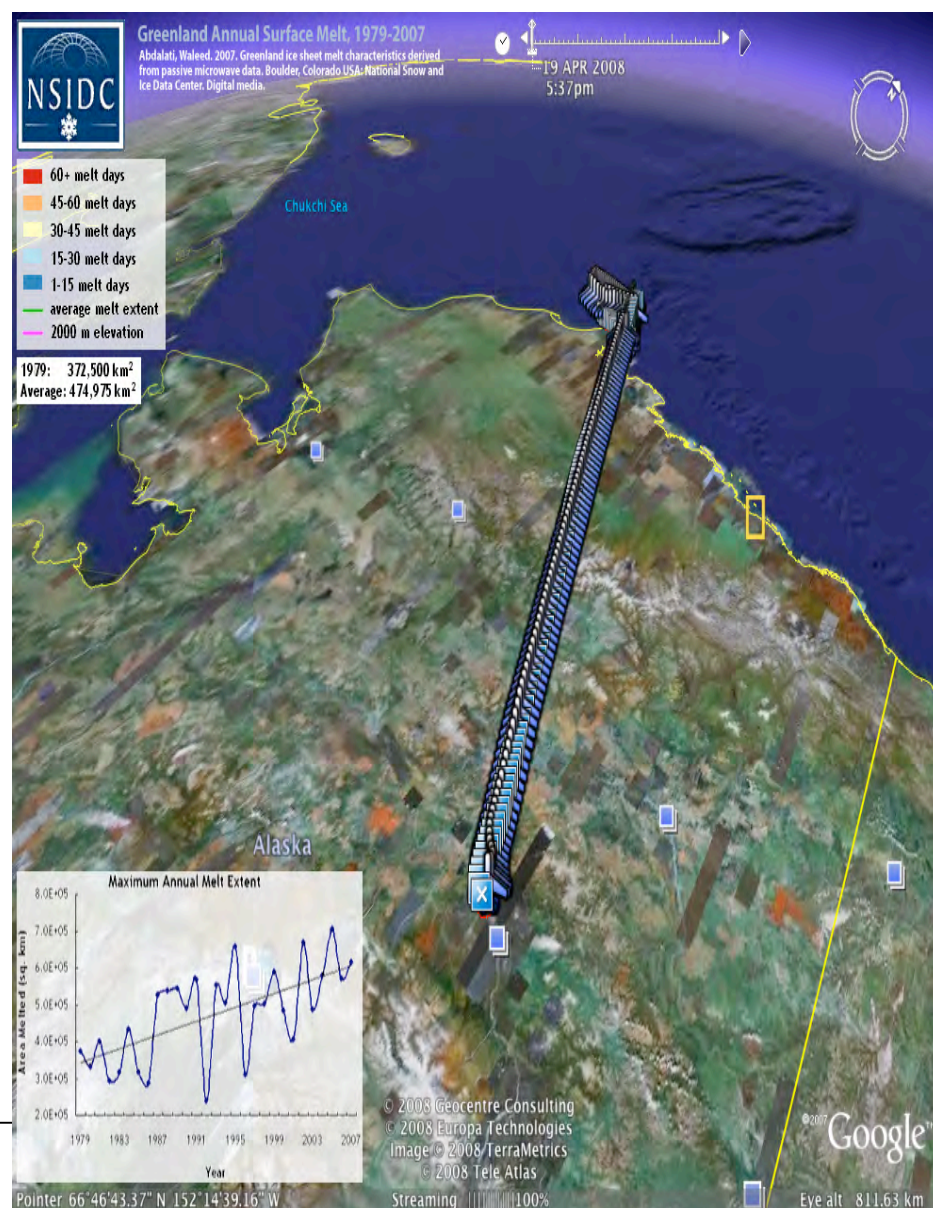
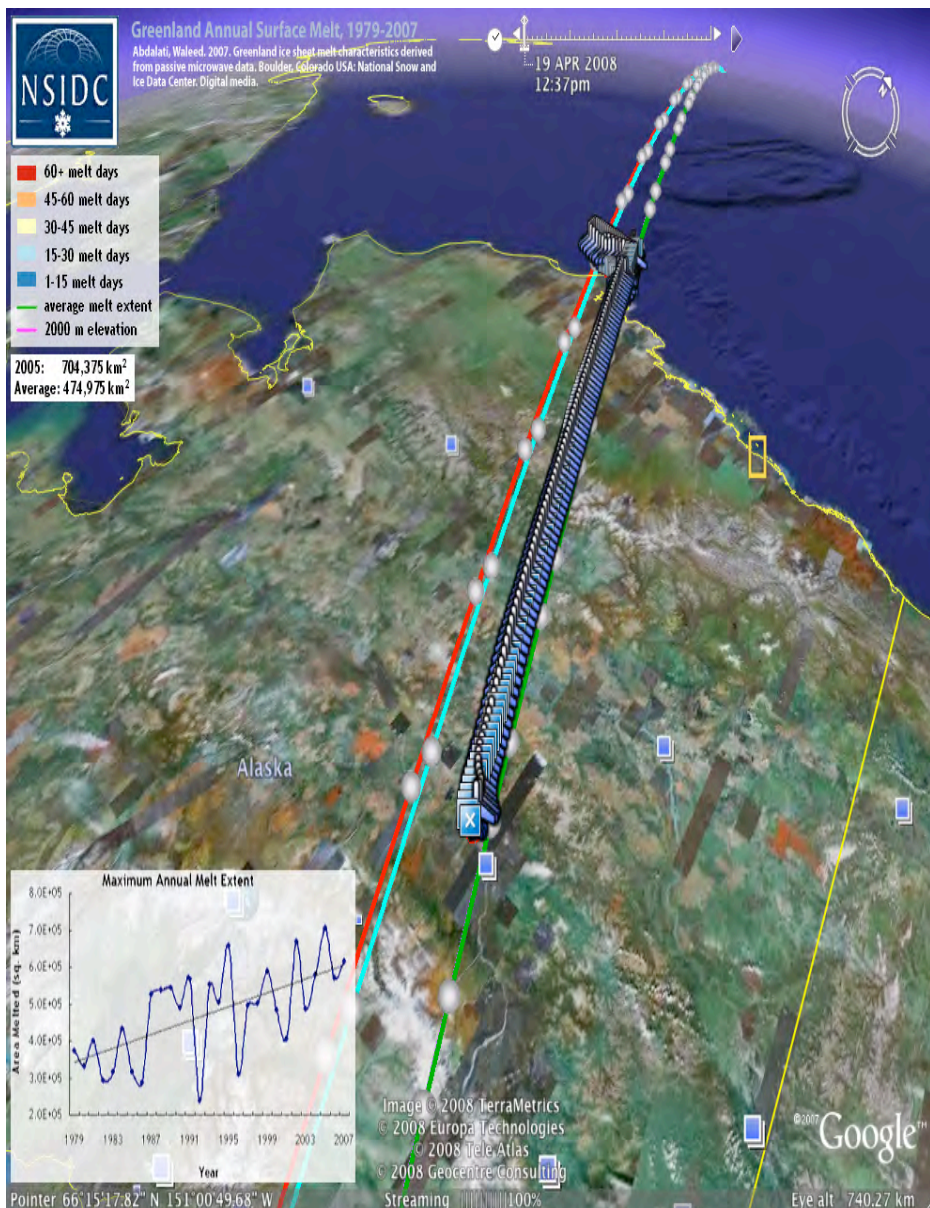
The tropopause pressure is contoured.

The location of the 15:17:11Z April 18<sup>th</sup>, 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.



# C-580 Flight: 19<sup>th</sup> April 2009, 11.30am – 8.30pm

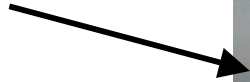


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# Large Pollution in Polar Regions

(ISDAC 19 April 2008)

Layer of  
Arctic Haze



Motivations

Aerosol effects

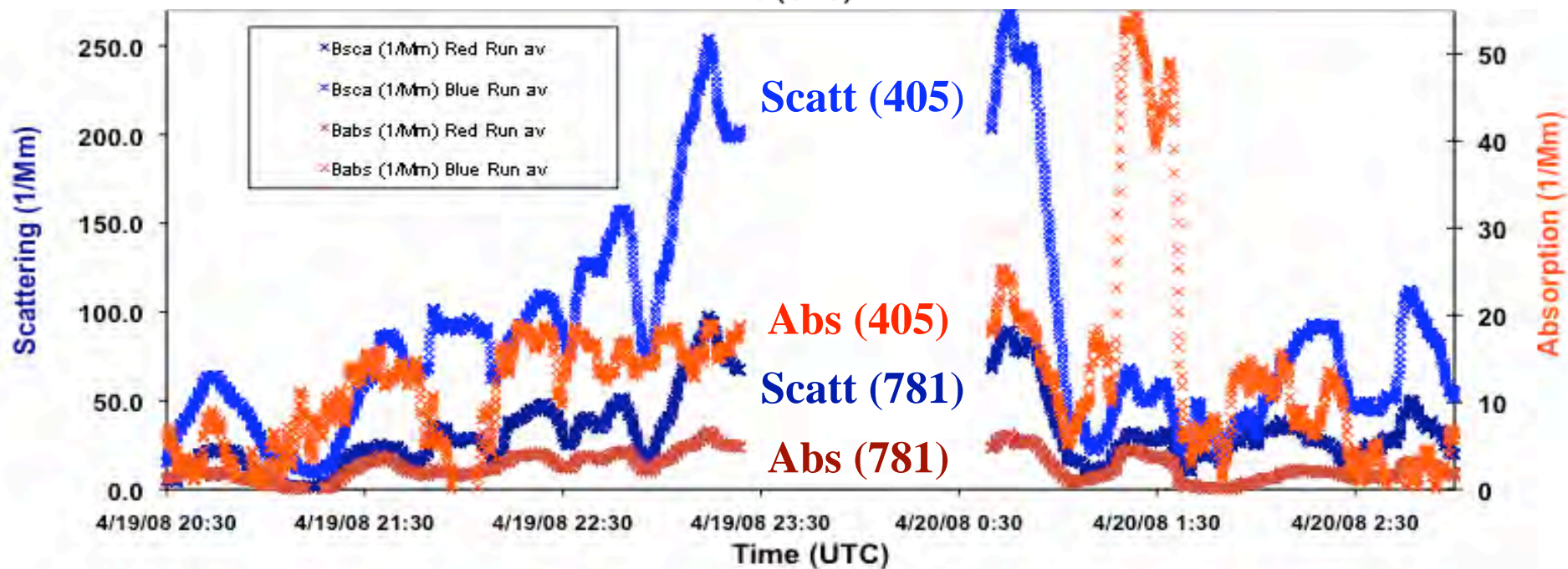
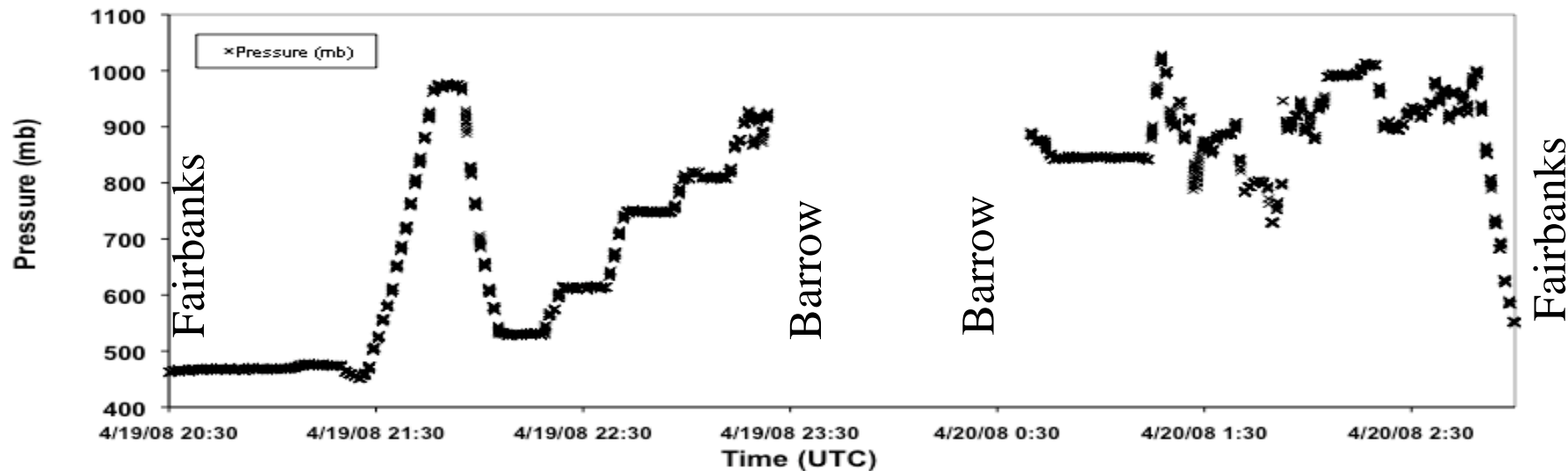
Optical properties

Mexico City

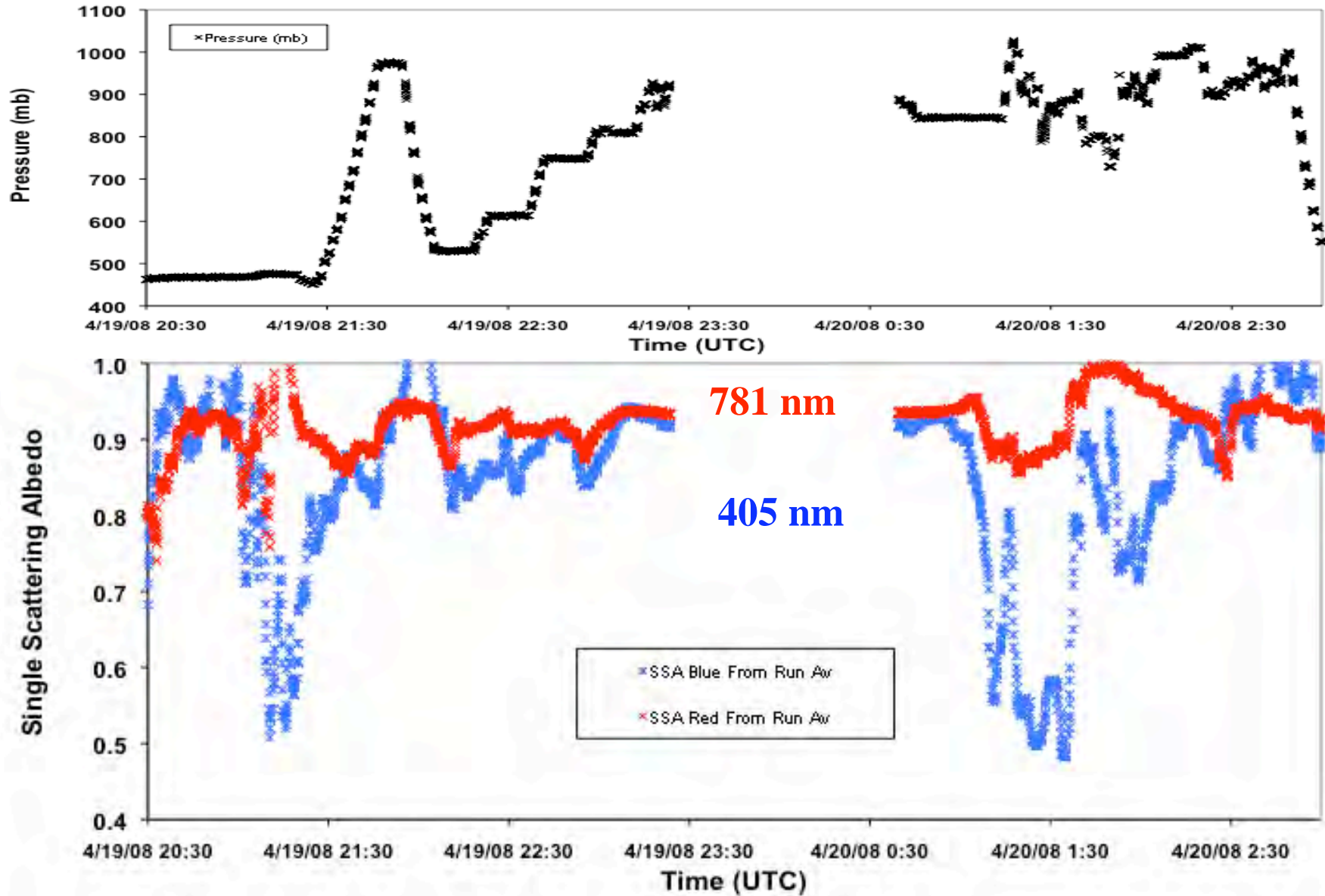
ISDAC

Future research

# Flight Track Optical Properties: Time Series

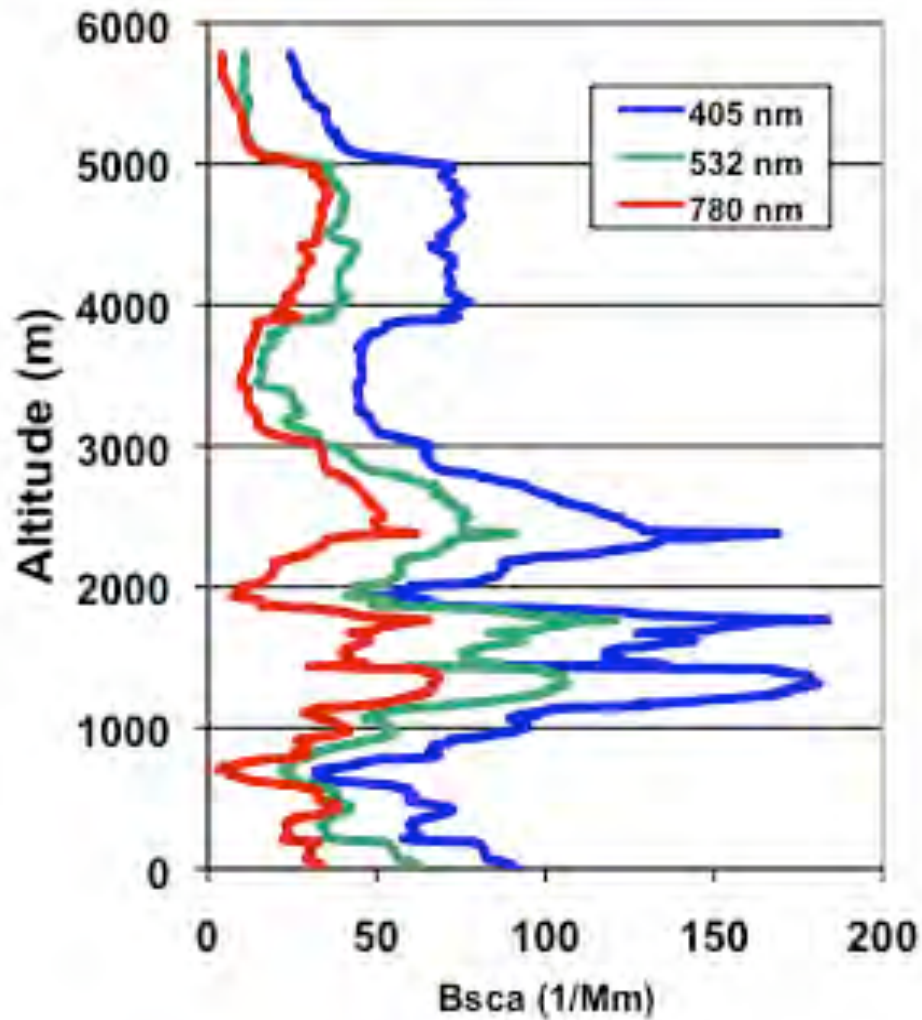


# Flight Track Single Scatter Albedo: Time Series

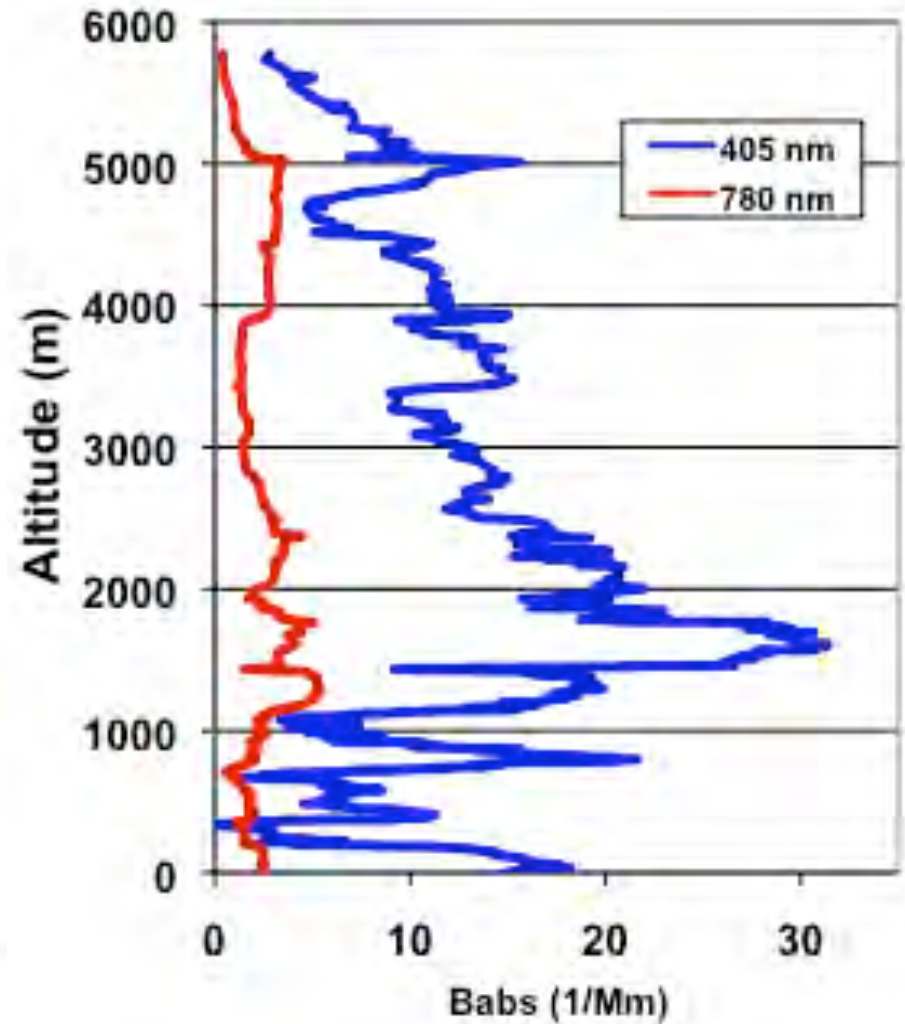


# Pollution Layers: Soot, OC, Dust, Sulfate

Scattering  $\text{Mm}^{-1}$



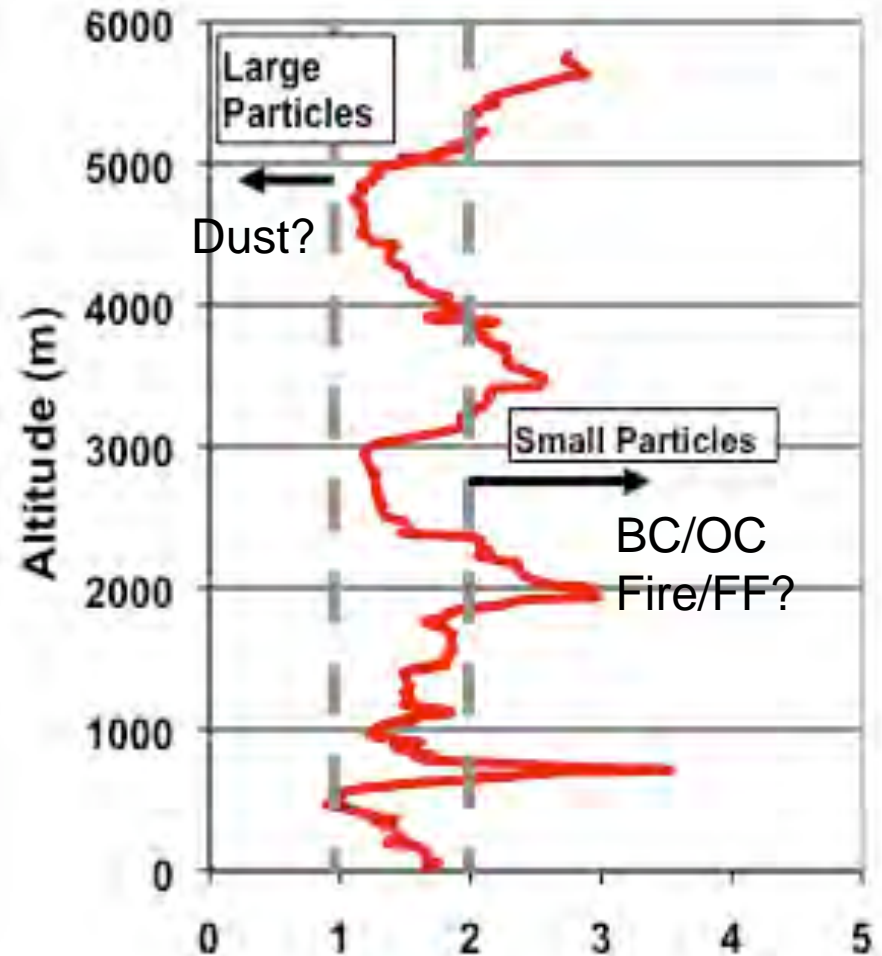
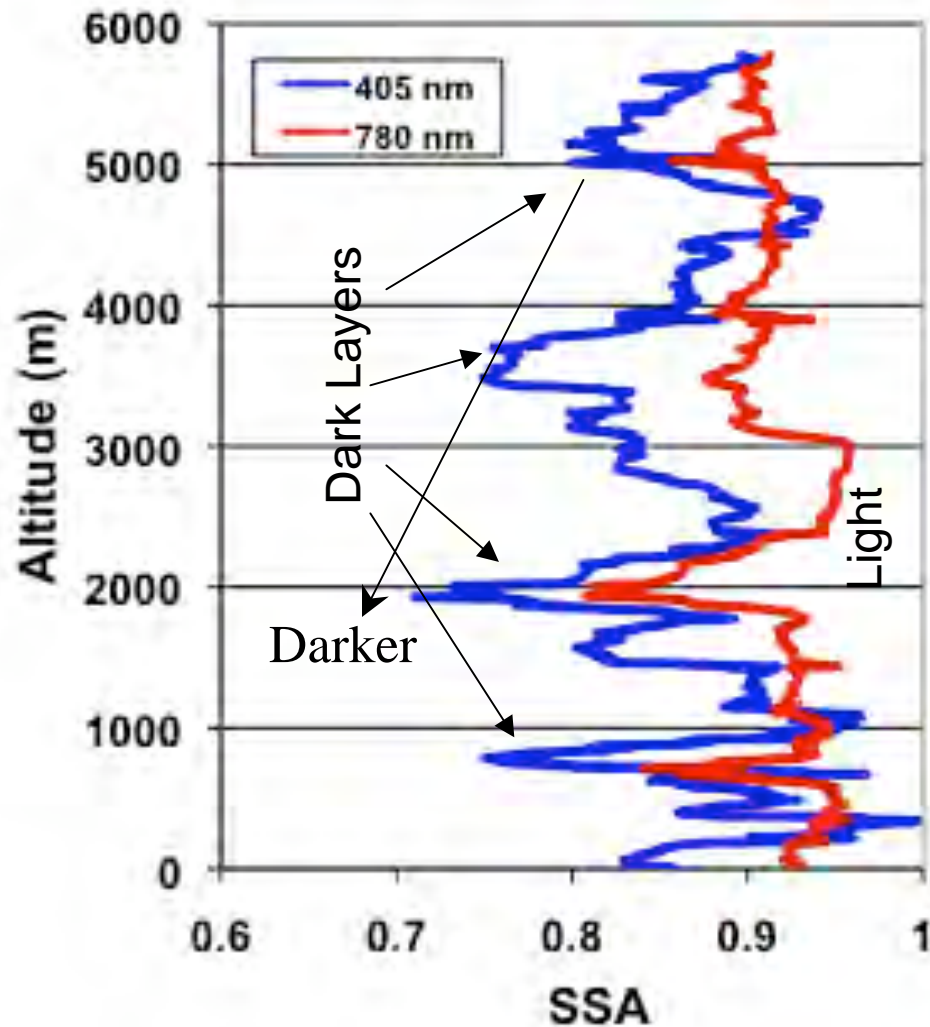
Absorption  $\text{Mm}^{-1}$



# Pollution Layers: Soot, OC, Dust, Sulfate

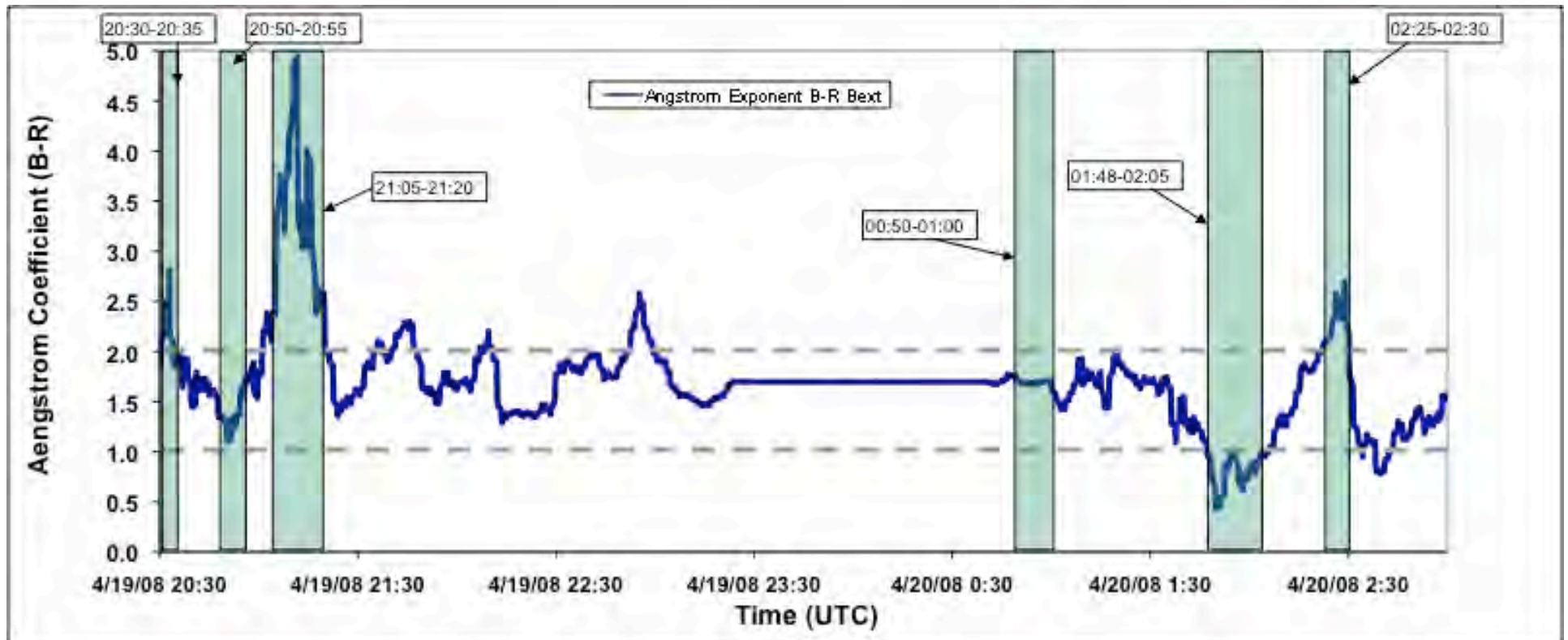
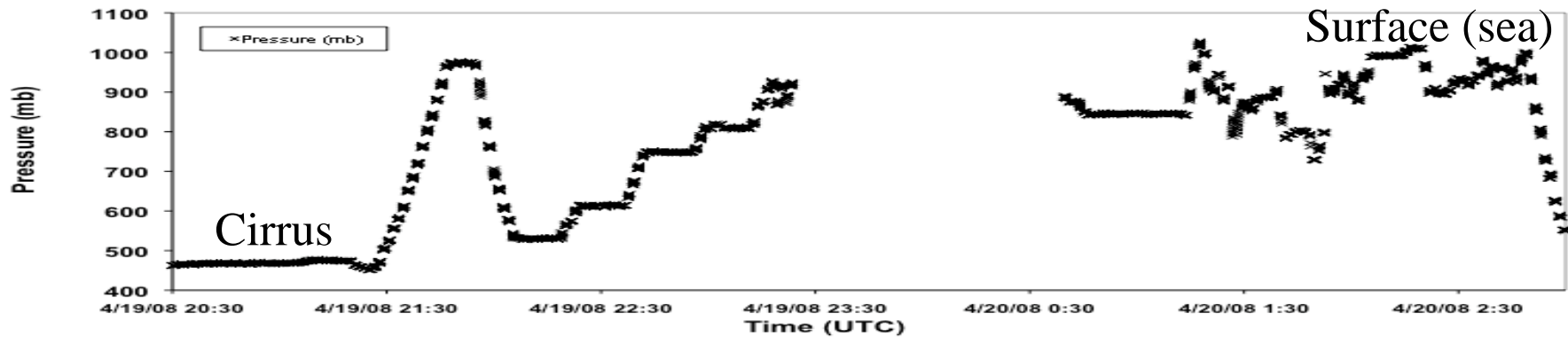
Single Scatter Albedo [S/(S+A)]

Angstrom Coeffn



$$\alpha = -\ln(\text{Ext}_{780} / \text{Ext}_{405}) / \ln(780 / 405)$$

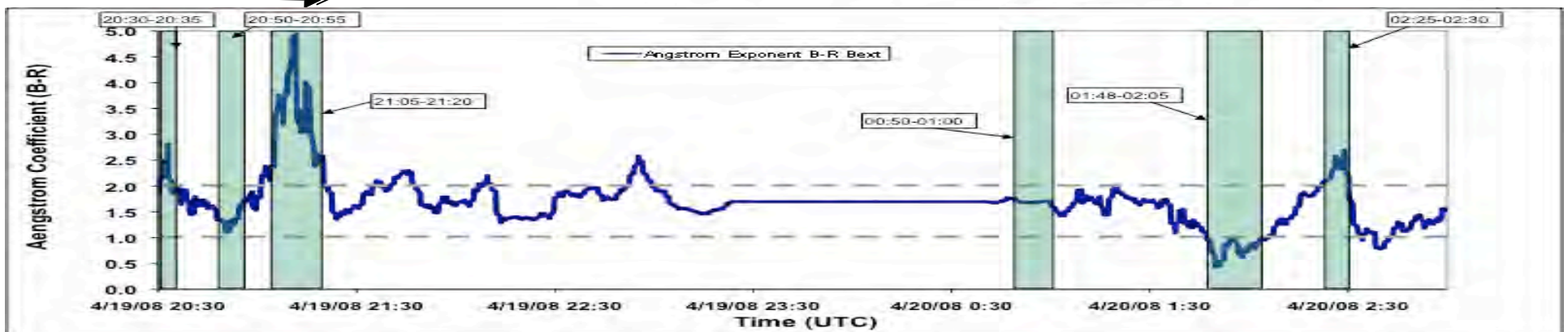
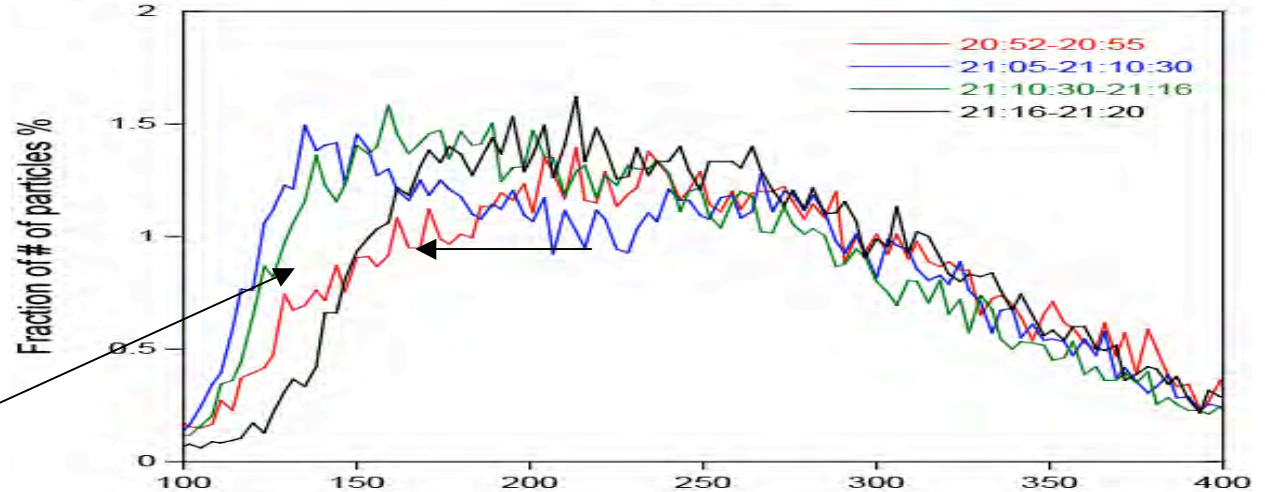
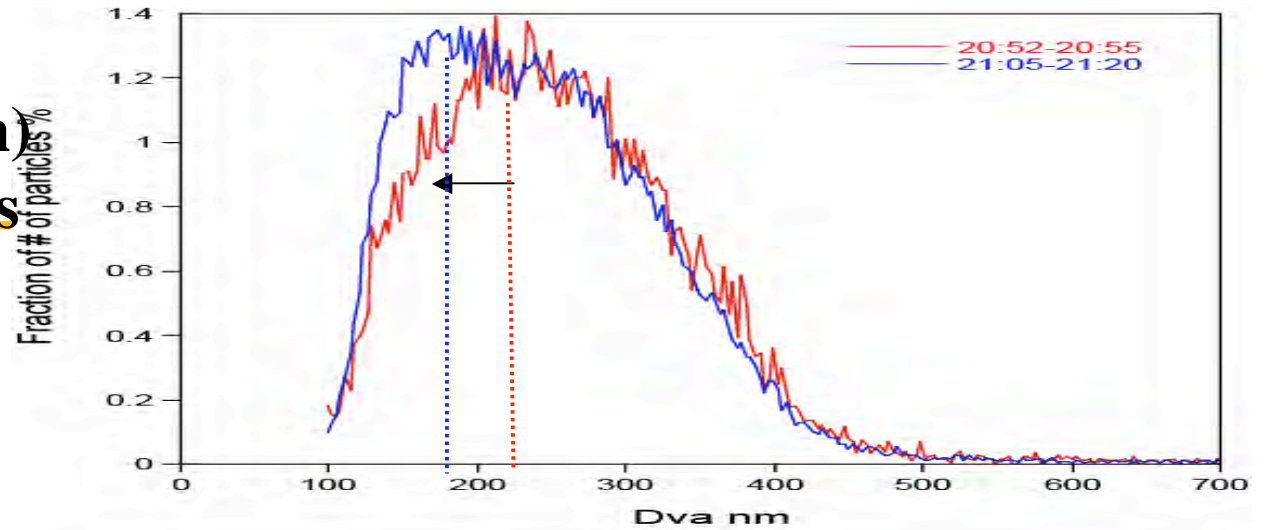
# Large Angstrom Exponent Gradients Along Flight Track: April 19



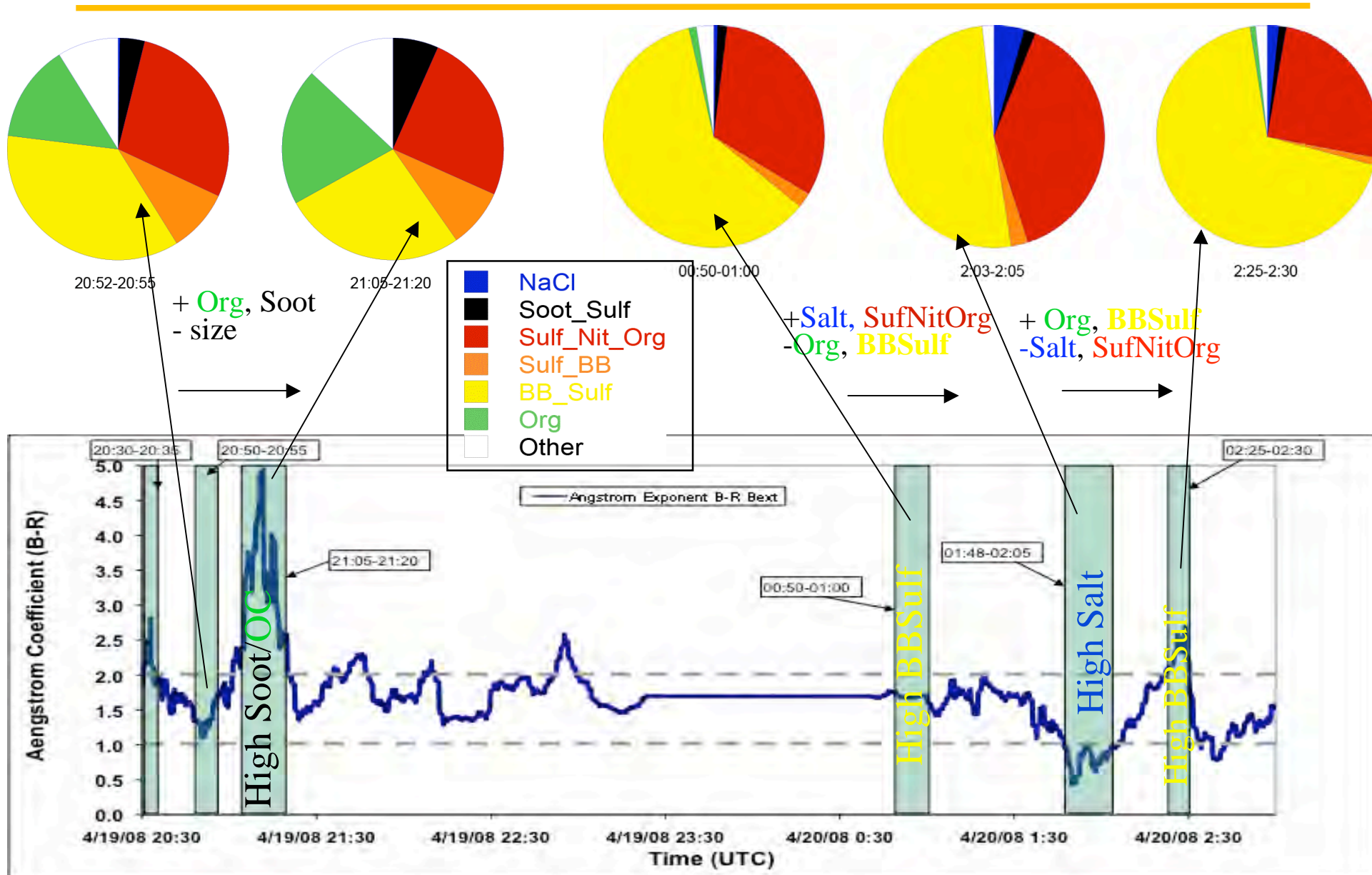


# Gradients in Optical Properties (Angstrom) and Size Distributions

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

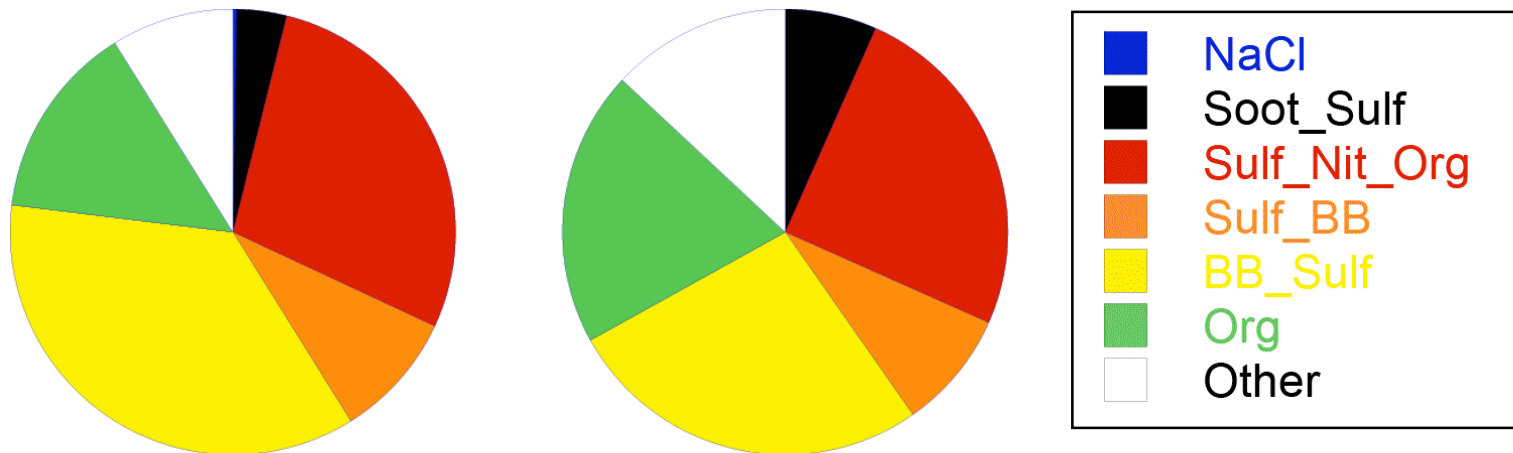


# Link Optical Properties (Angstrom Gradients) to Chemical Composition Changes



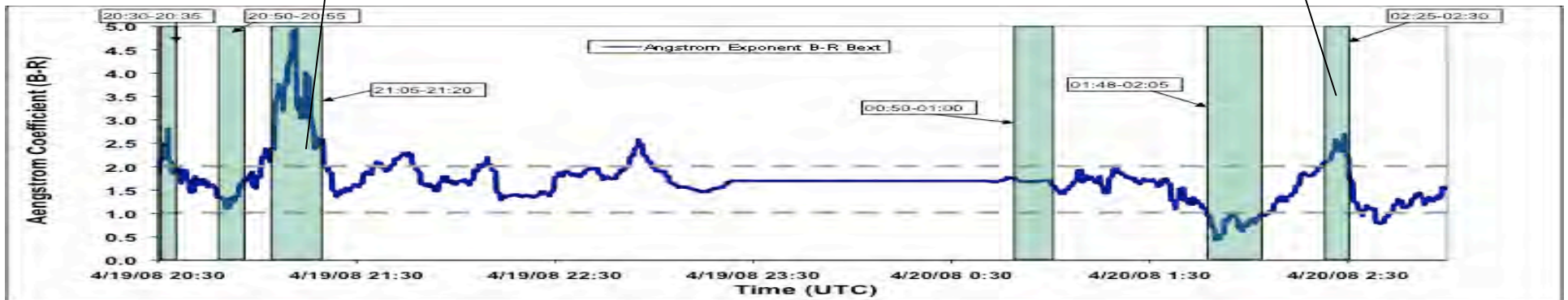
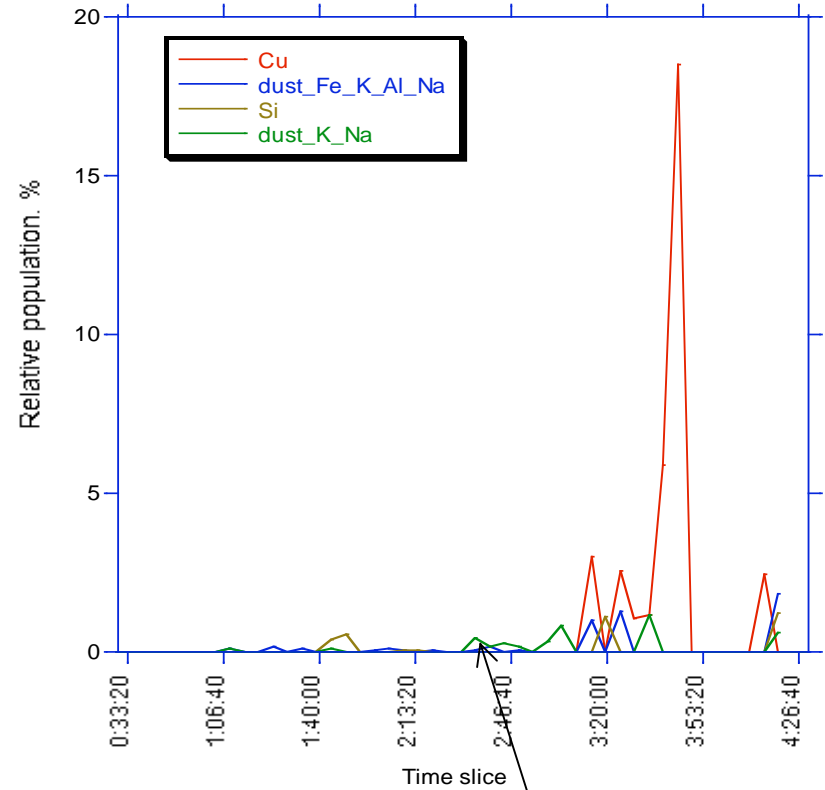
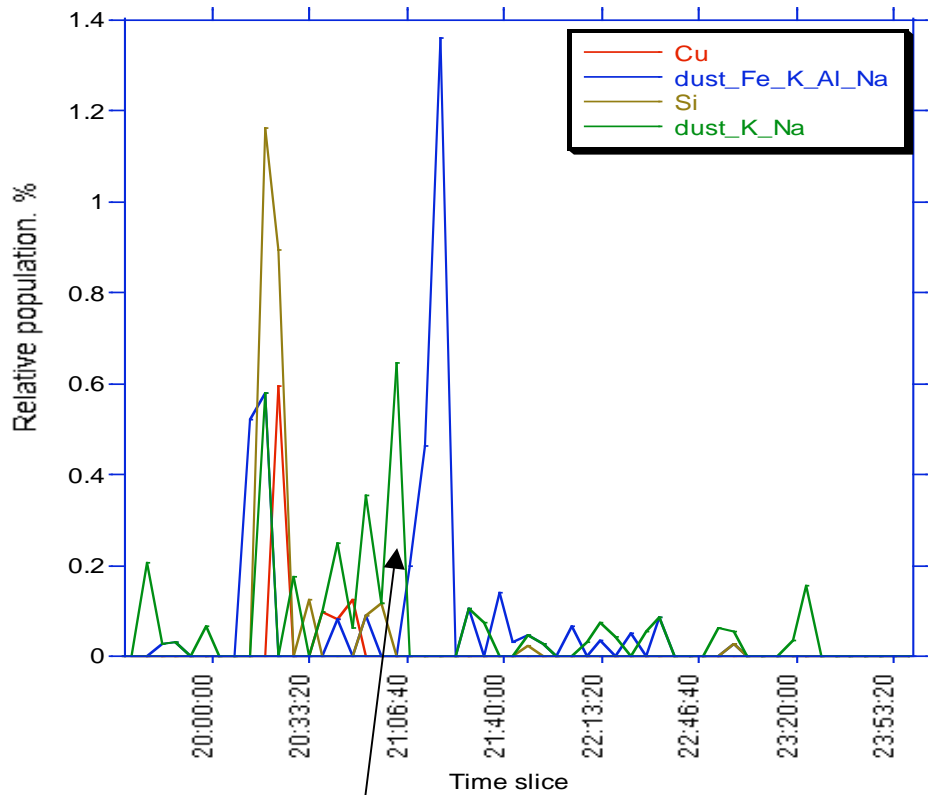
# Organic/soot enhancement correlates with smaller particles that are darker in the blue (405 nm)

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



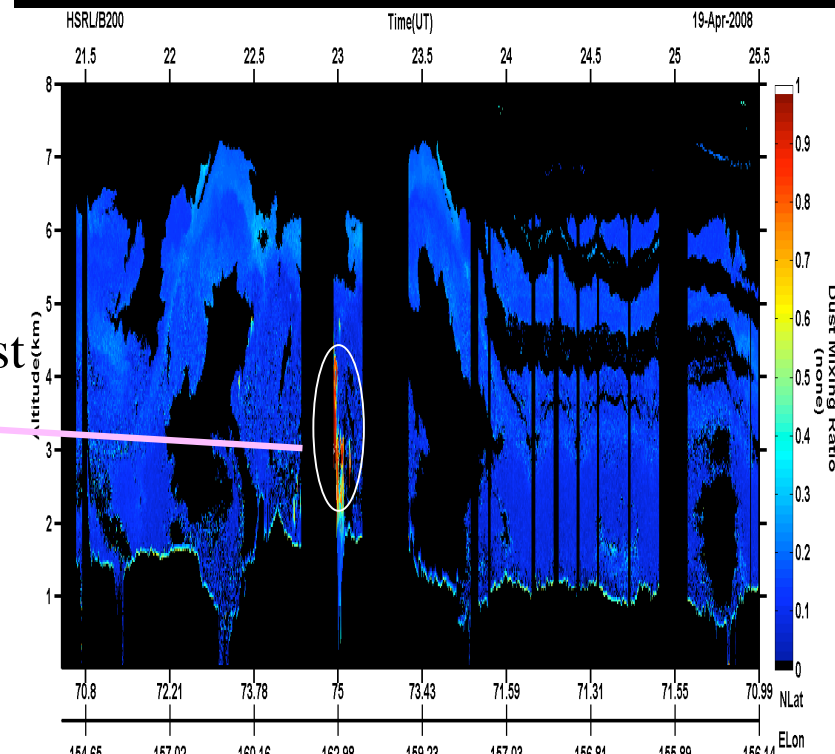
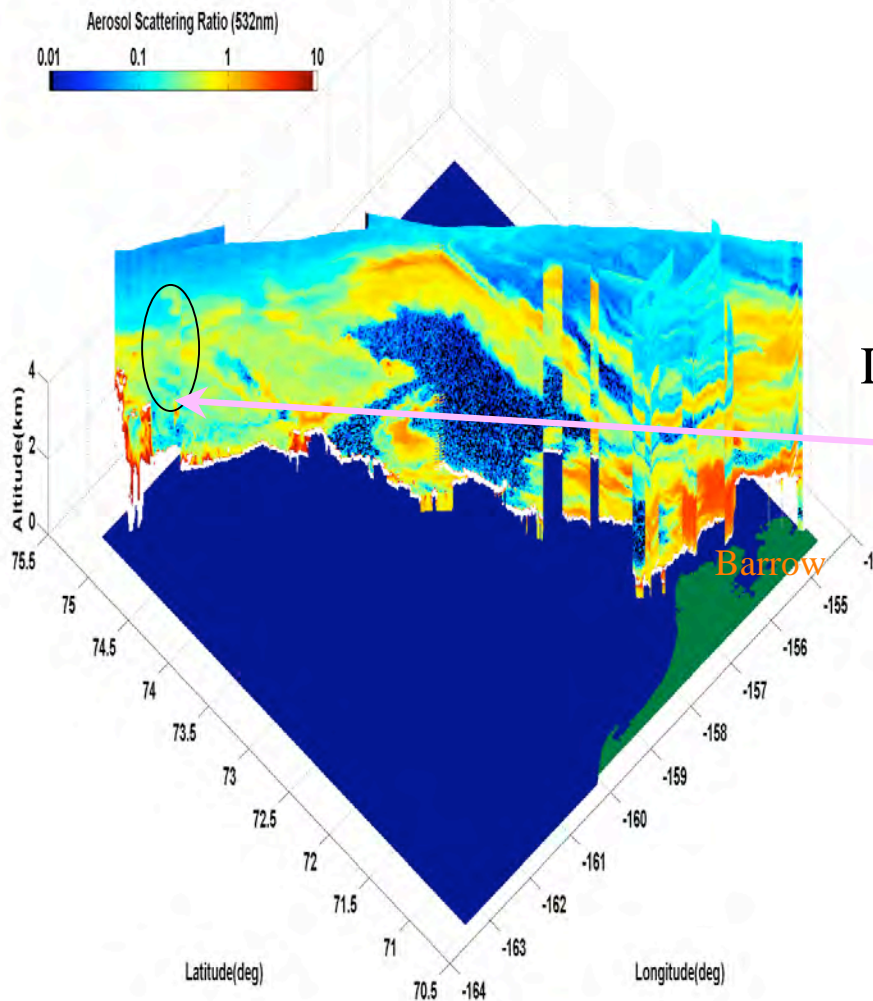
- Increase in Soot & Organic Components (x2)
- Mean diameter decreases from 220nm to 175nm
- Angstrom exponent increases by 2.5
- 405 nm absorption increases x 4, scattering decreases x 1/4
- SSA drops from 0.95 to 0.65

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



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

## 532nm Scattering Ratio



Dust Fraction from Depolarization Ratio 532 nm

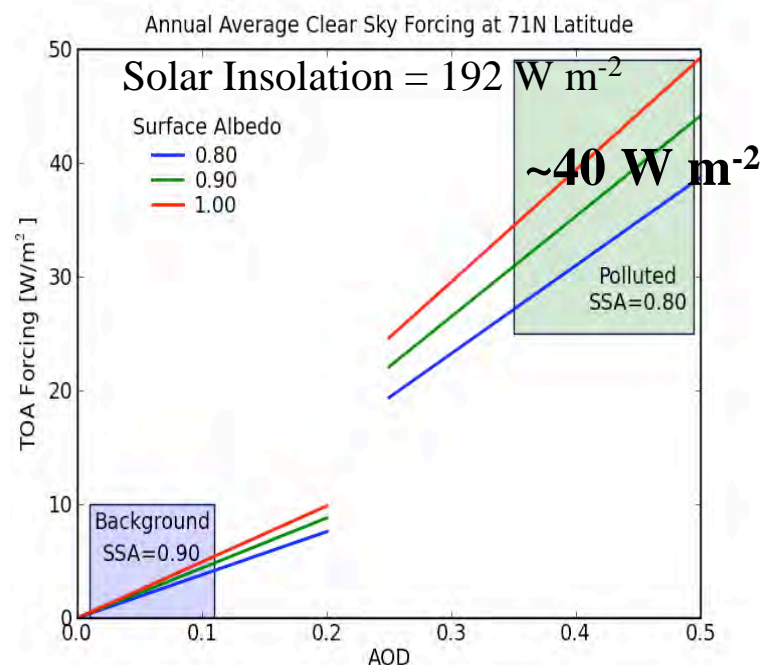
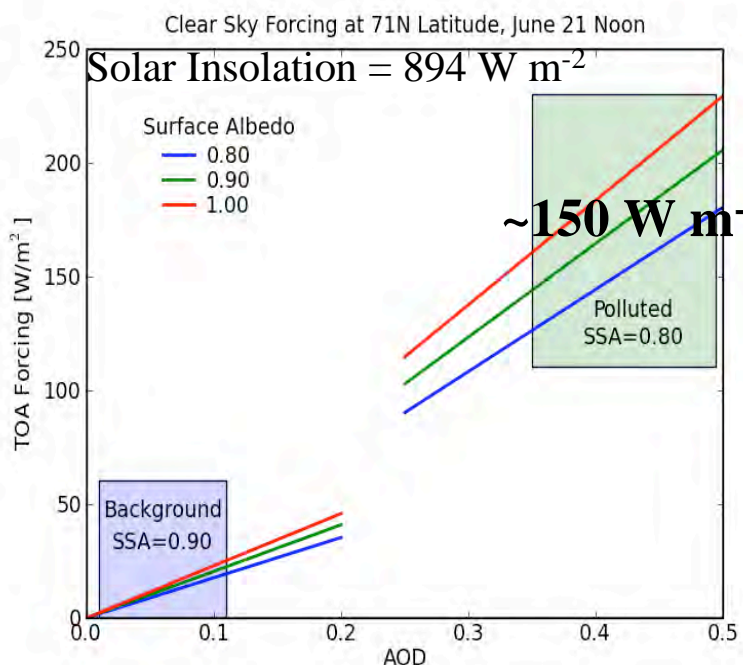
Scale: Ground (Barrow), in situ profile, LIDAR, satellite



# UNCLASSIFIED 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

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



Perturbation  $\Delta\text{GISS}^{\text{Mod}}$  is 0.92 Wm<sup>-2</sup> (Spr.), 0.3 Wm<sup>-2</sup> (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.**

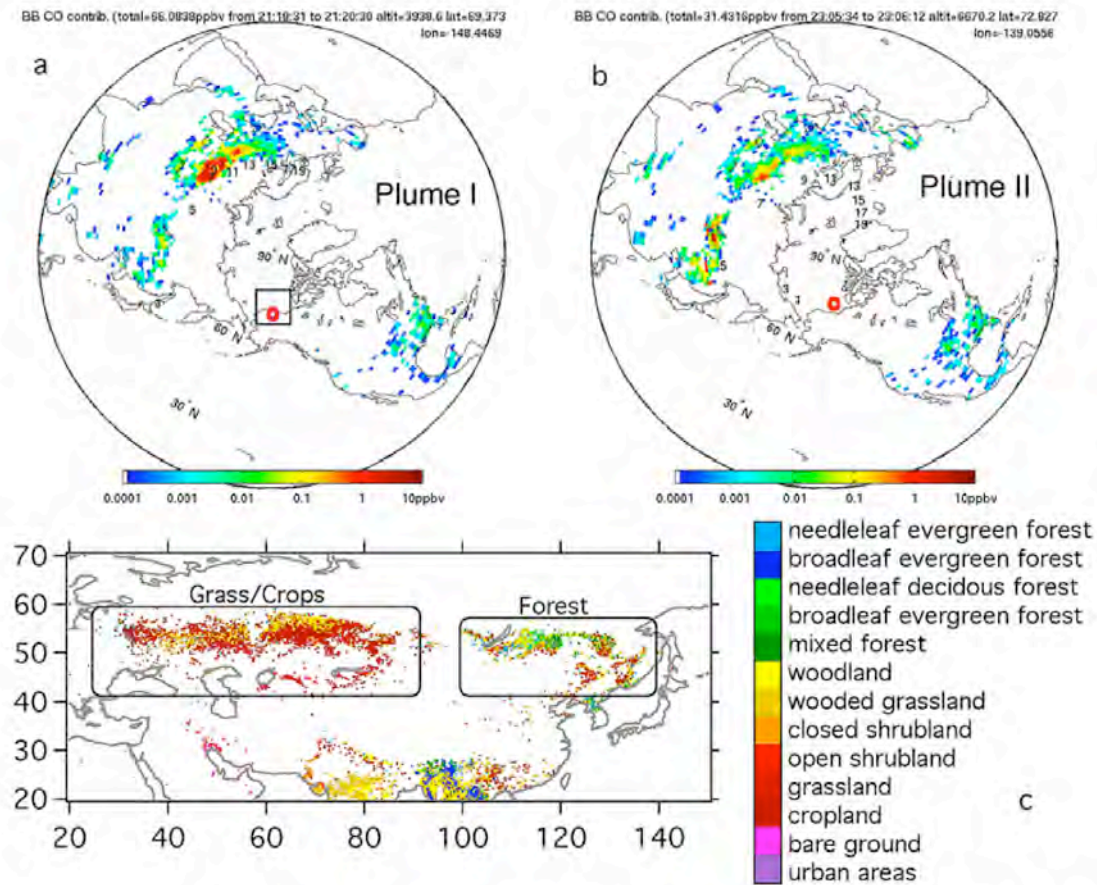


# Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008

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

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 logarithmic scale. The white box in Figure 2a indicates the area where the ARCPAC flights took place and all the BB plumes were observed. (c) MODIS fire detection between April 1, 2008 and April 20, 2008. The color code indicates the dominant land cover type.

# Conclusions

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- Arctic haze optical properties consistent with layers of aged biomass-fires, dust and sulfate
- Internally mixed Soot-Sulf, Sulf-BB, Sulf-Nit-Org
- Optical property gradients correlated to changes in chemical composition and size distributions: Needs Closure Studies
- Our SSA imply large episodic forcing above Arctic in April that are not captured in current climate change projection models
- Follow-up: (1) Microphysical-chemistry-optical closure with WRF (Fast) (2) Capture episode in CCSM4 (Ghan) & GISS model (Shindell) for Arctic forcing (Ferrare) (3) Atmospheric heating rates from absorbing layers (4) Ice nucleation (Brooks)