Changes in visibility and local radiative forcing in the Southeast U.S. linked to decreased aerosol sulfate mass

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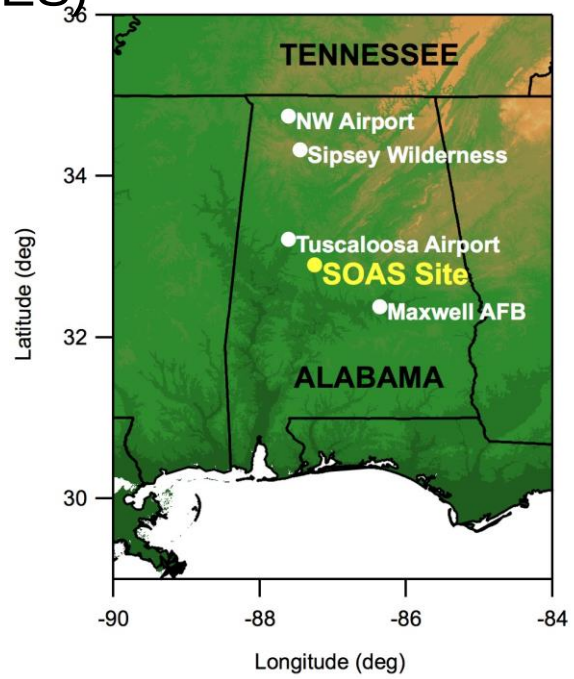
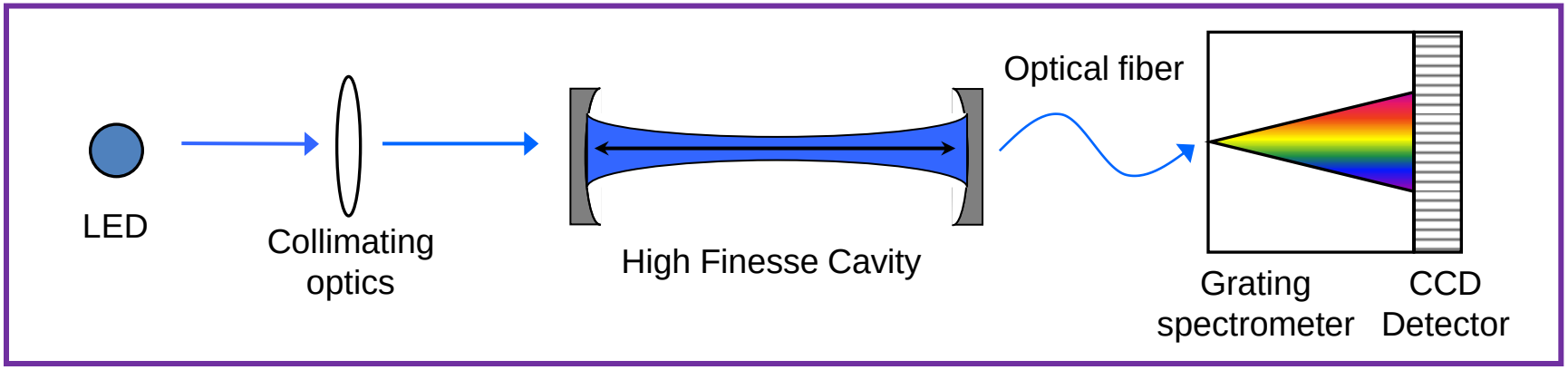
U.S. DEPARTMENT OF COMMERCE

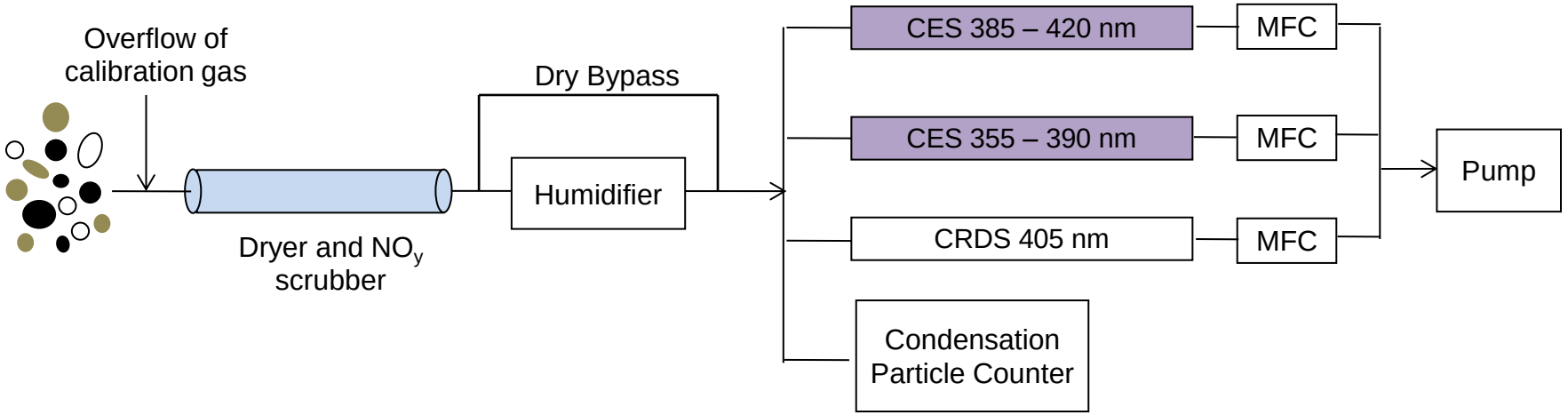
Introduction

- Sulfate is a major contributor to $PM_{2.5}$ mass in US
  - Very hygroscopic
- Southeastern Oxidant and Aerosol Study (SOAS)
  - Aerosol dominated by ammonium sulfate and organics
  - Humid
- BroadBand Cavity Enhanced Spectrometer (BBCES)
  - Dry and humidified aerosol extinction from 355-420 nm
- Aerosol Mass Spectrometer: CU Boulder
- Historical Data
  - SEARCH: Collocated with SOAS site
  - IMPROVE: Sipsey Wilderness
  - Visibility: Airports



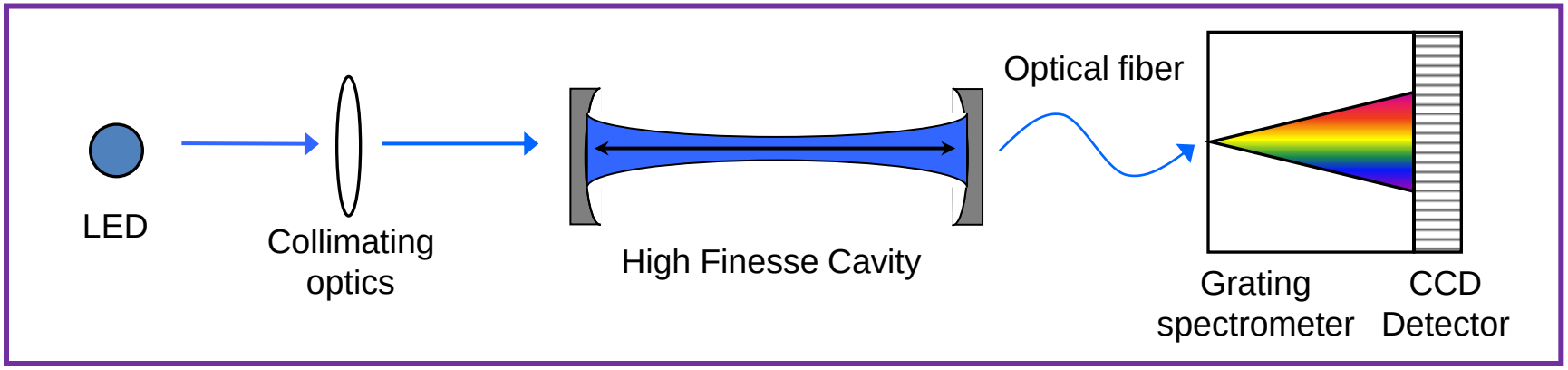
BBCES

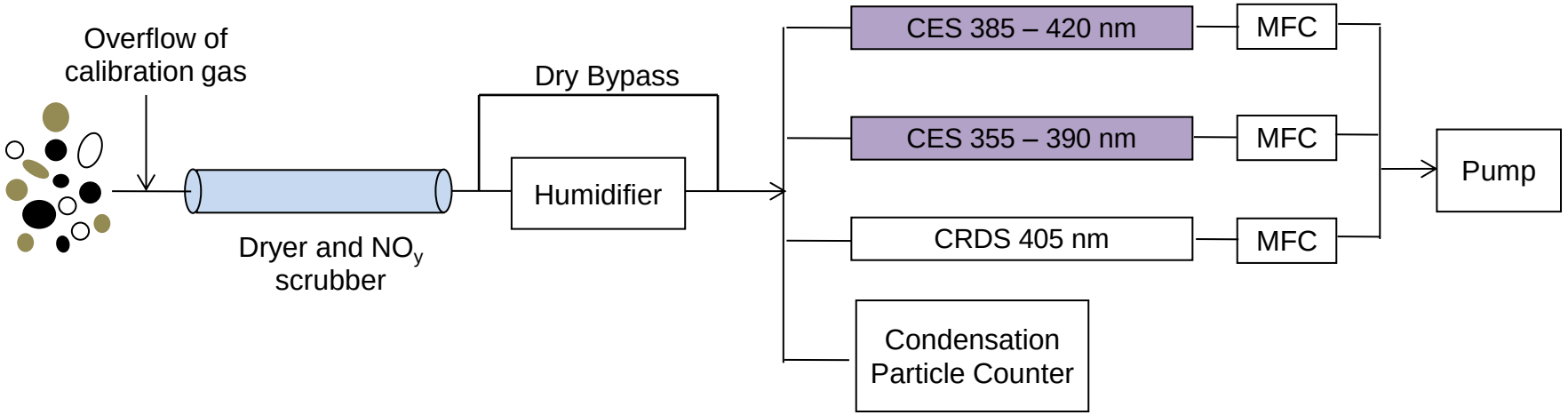




Washenfelder et al., 2013

BBCES

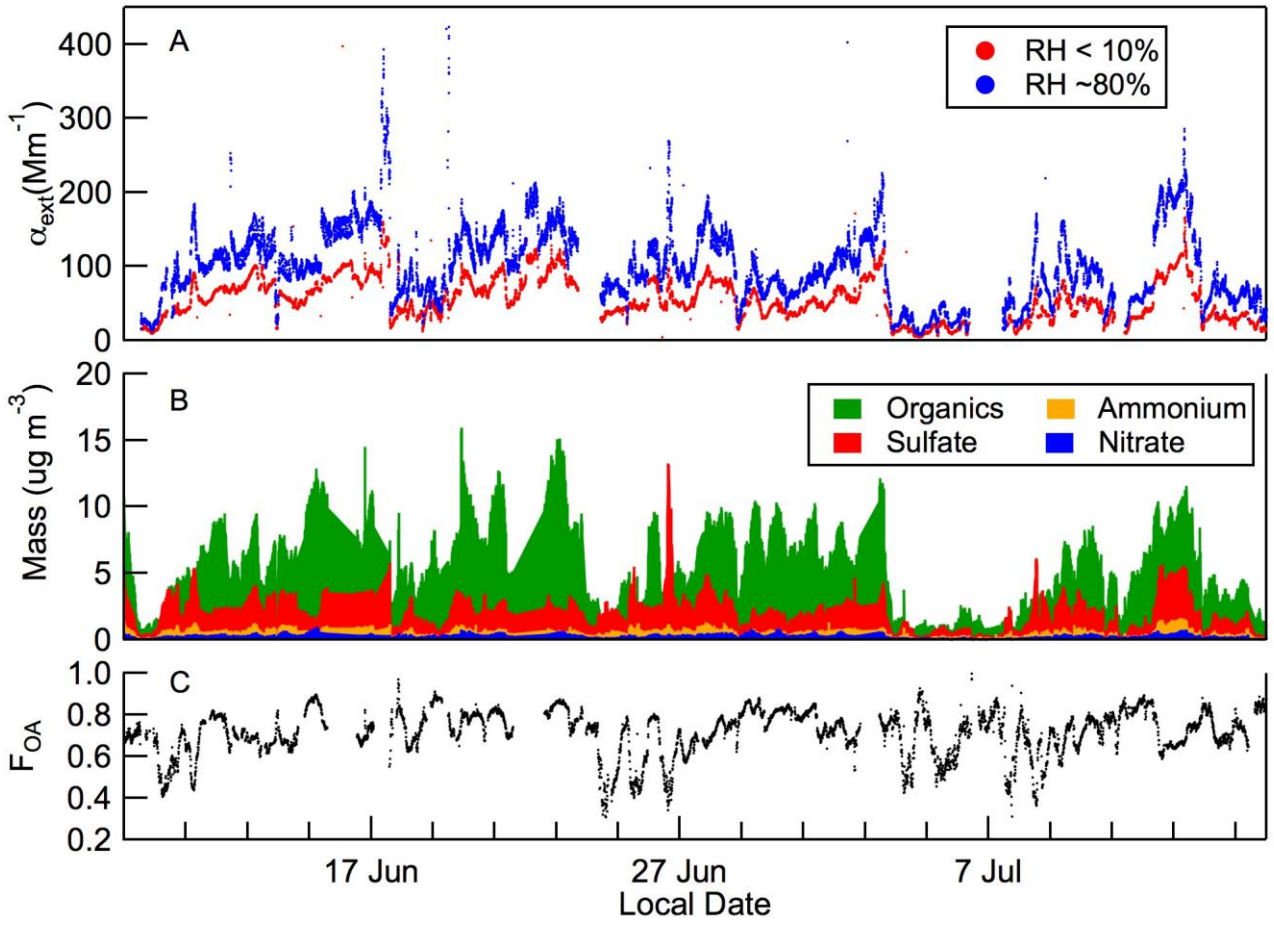




Total Extinction (α, cm^{-1})= Scattering + Absorption

$$a(\lambda) = R_L \left(\frac{1 - R(\lambda)}{d} + a_{Rayleigh,ZA}(\lambda) \left(\frac{I_{ZA}(\lambda) - I(\lambda)}{I(\lambda)} \right) \right)$$

Aerosol Optical and Chemical Characteristics During SOAS



Variable Extinction

Dominated by Sulfate and Organics

$$F_{OA} = \frac{[OrganicAerosol]}{[OrganicAerosol] + [Sulfate]}$$

Dependence of Water Uptake on Chemical Composition

$$\frac{a_{ext}(RH)}{a_{ext}(RH_{ref})} = \left[\frac{(100 - RH)}{(100 - RH_{ref})}\right]^{g_{ext}}$$

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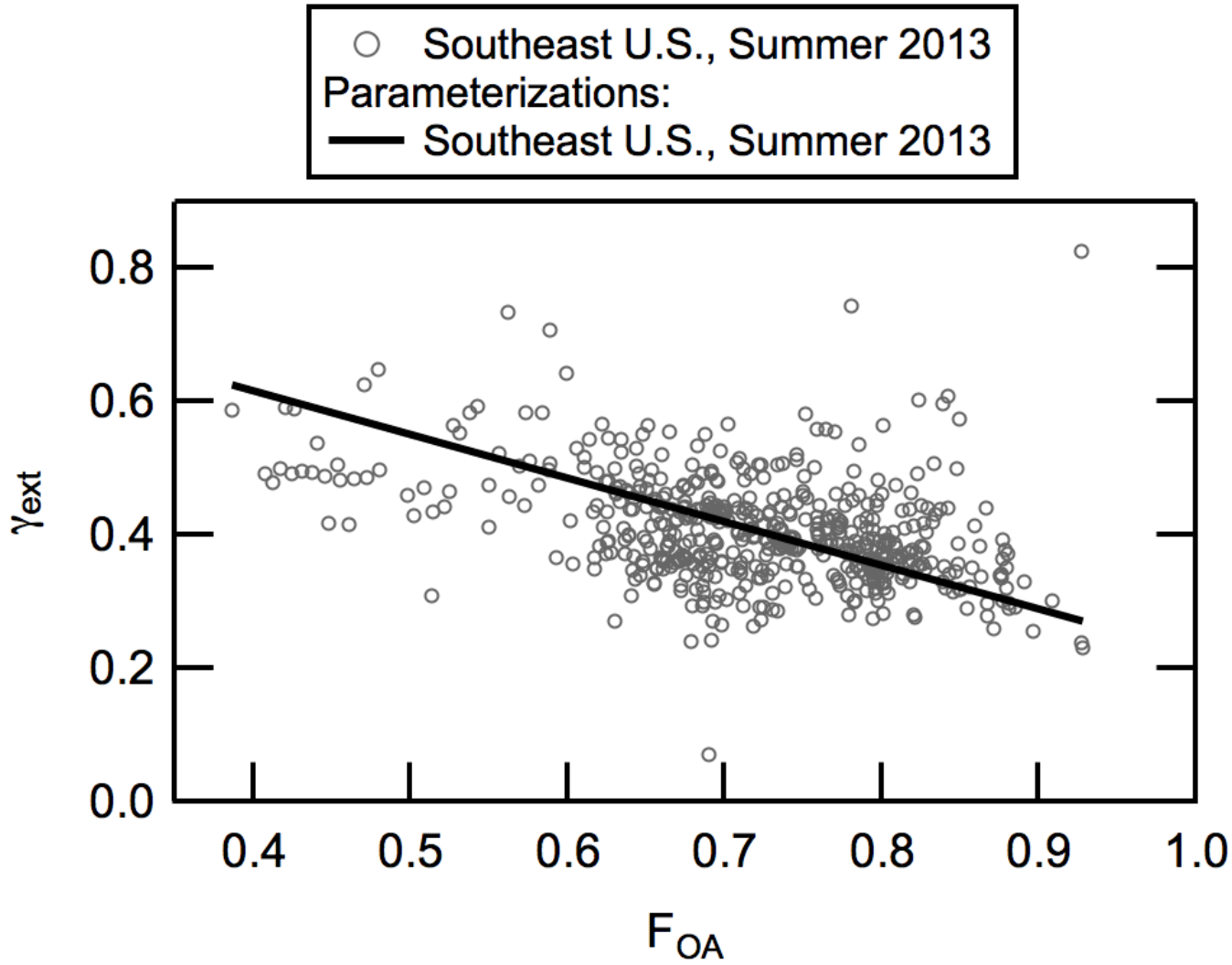

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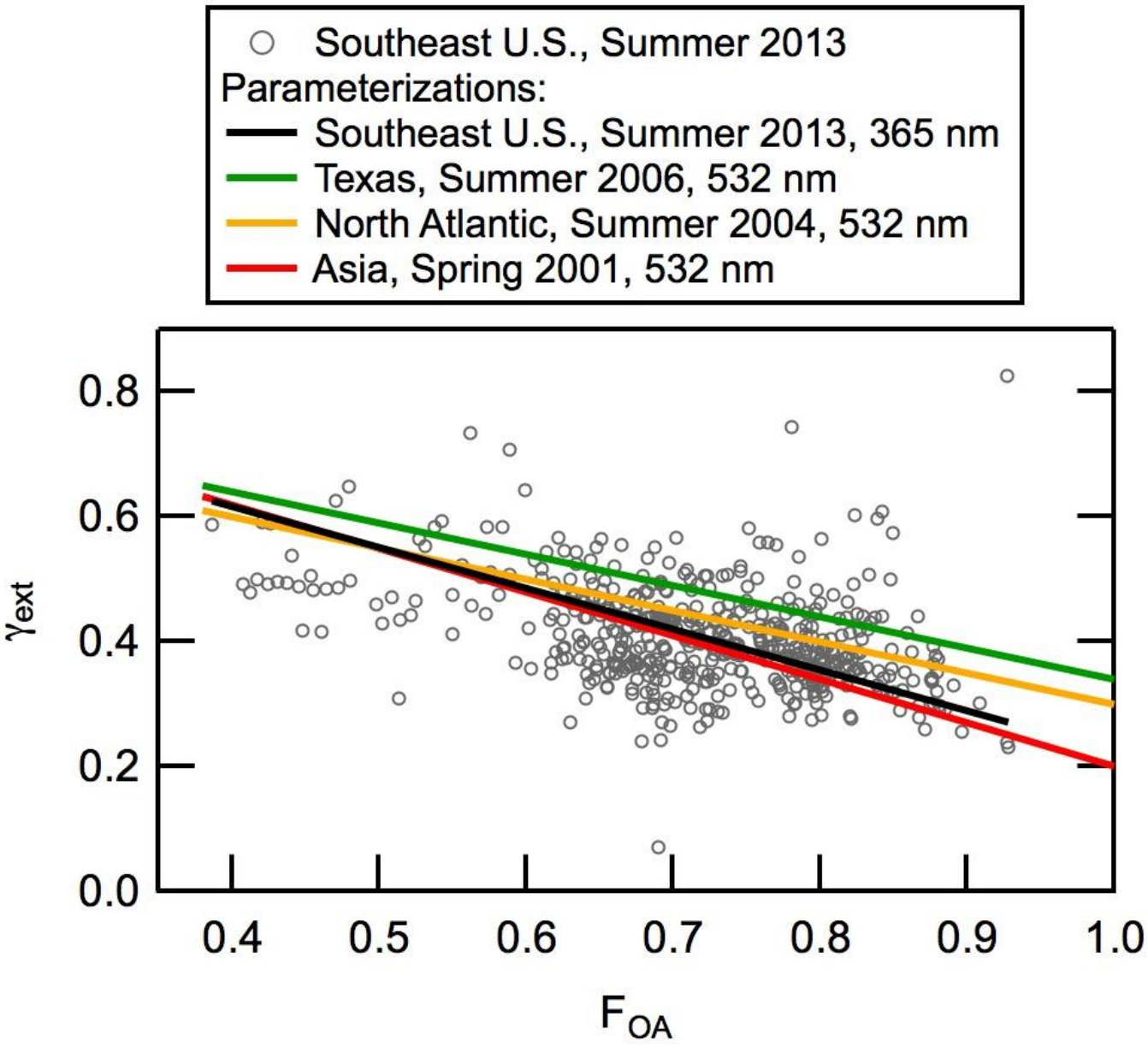
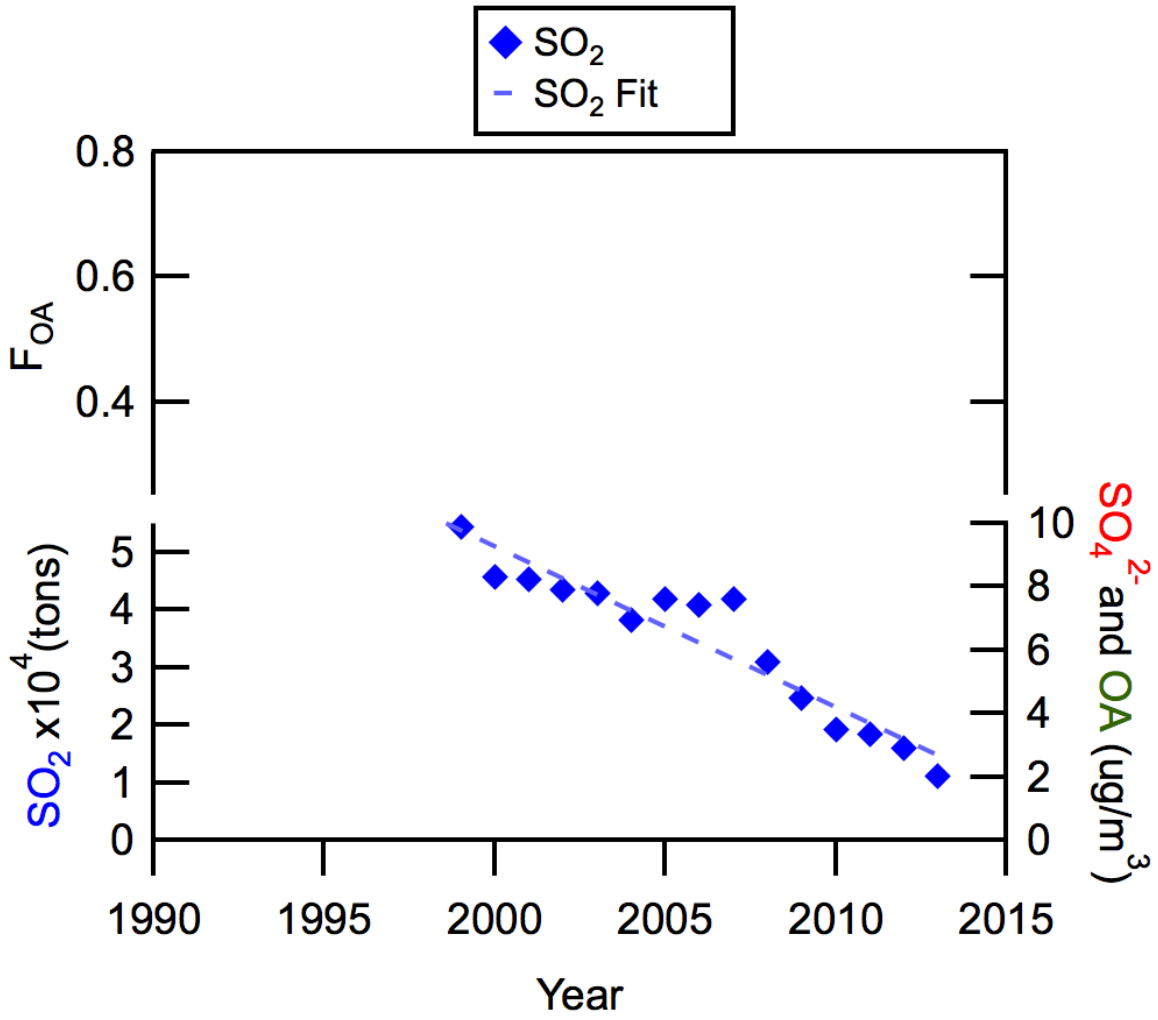
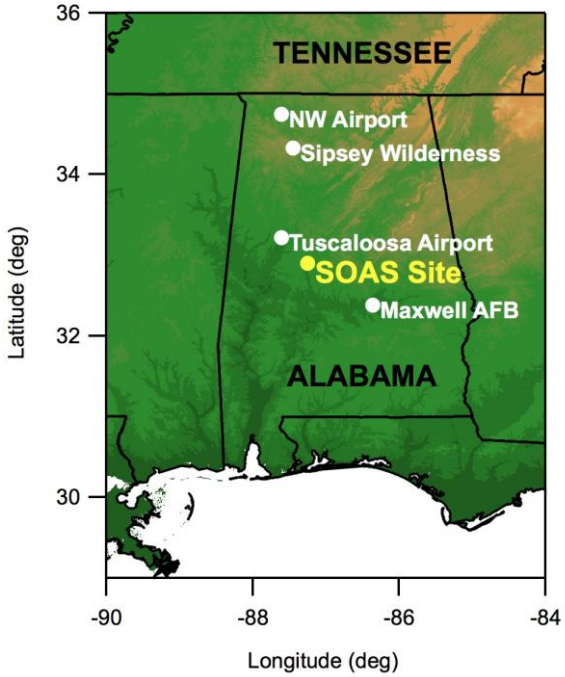


Historical Data



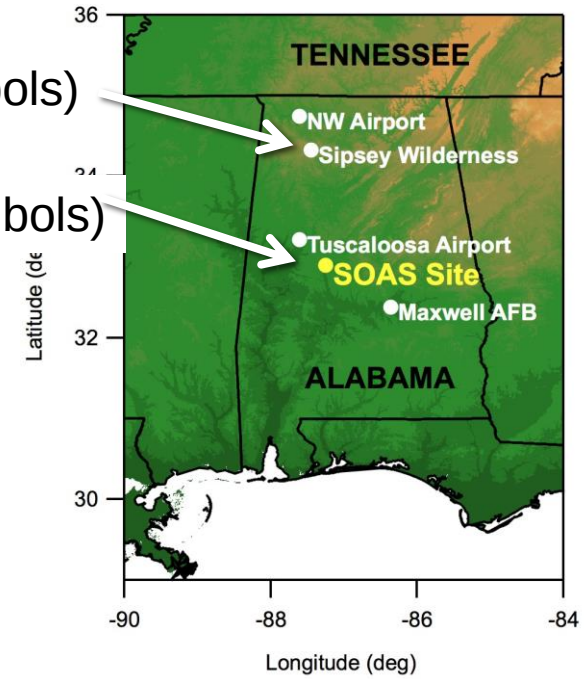


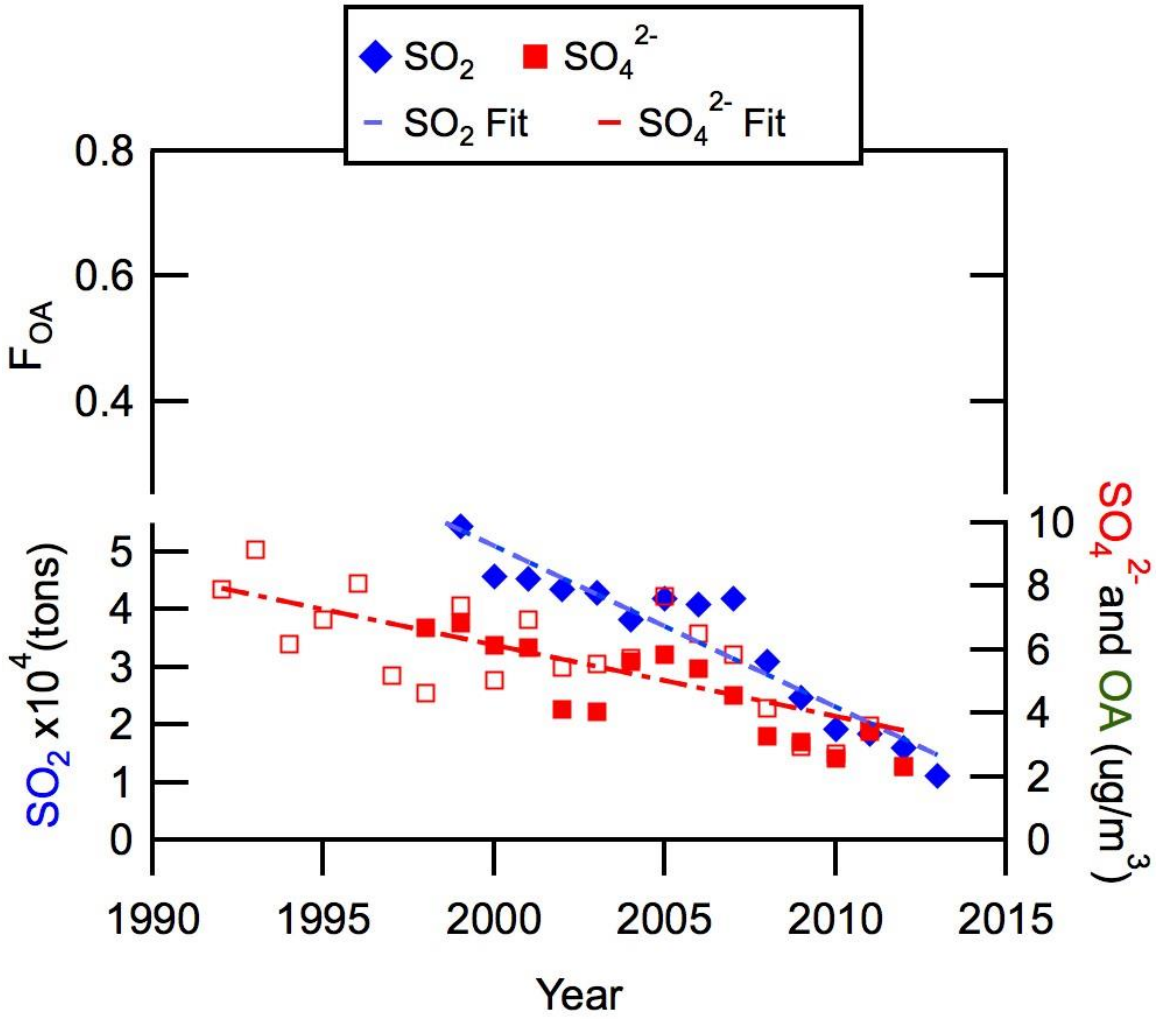
SO_2 data: JJA Averages in Alabama from NEI

Historical Data

IMPROVE (open symbols)

SEARCH (closed symbols)





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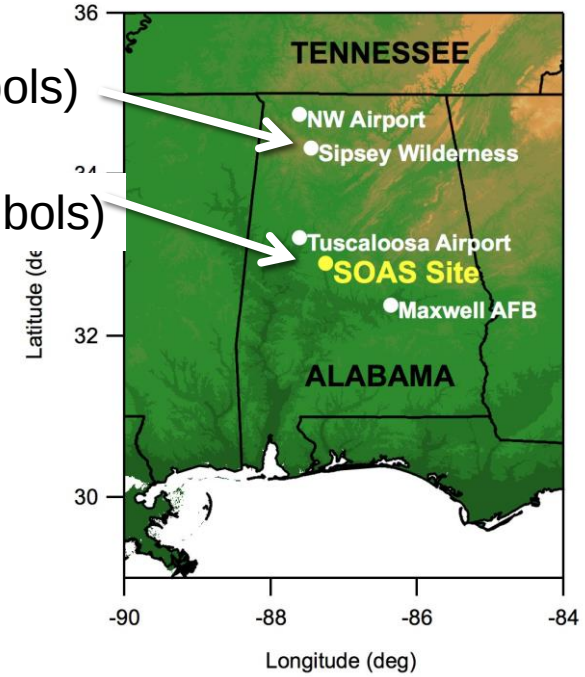

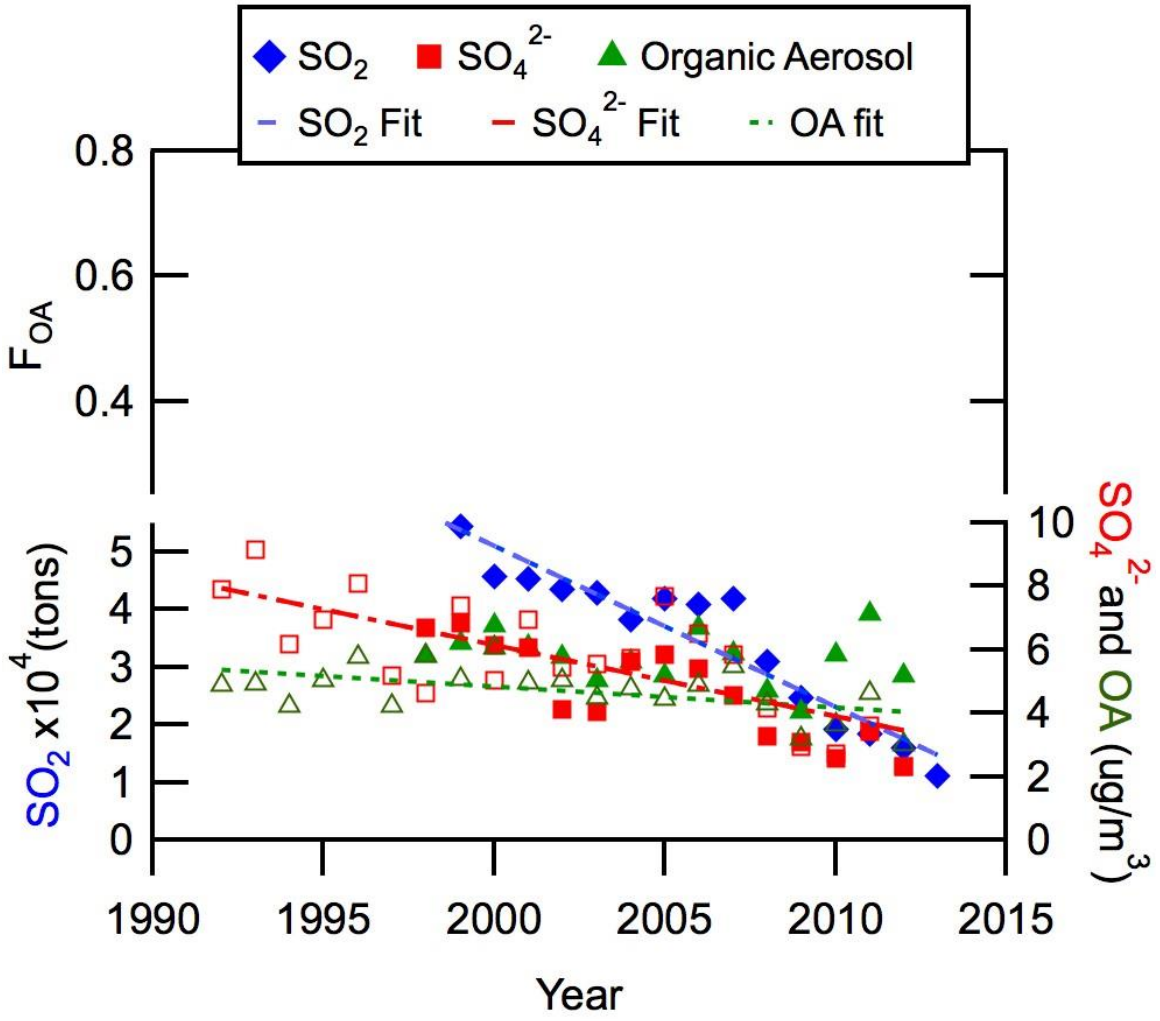

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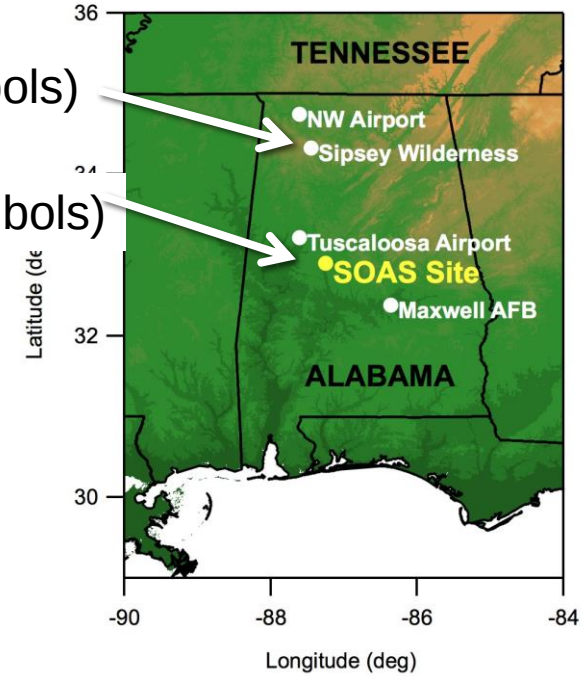

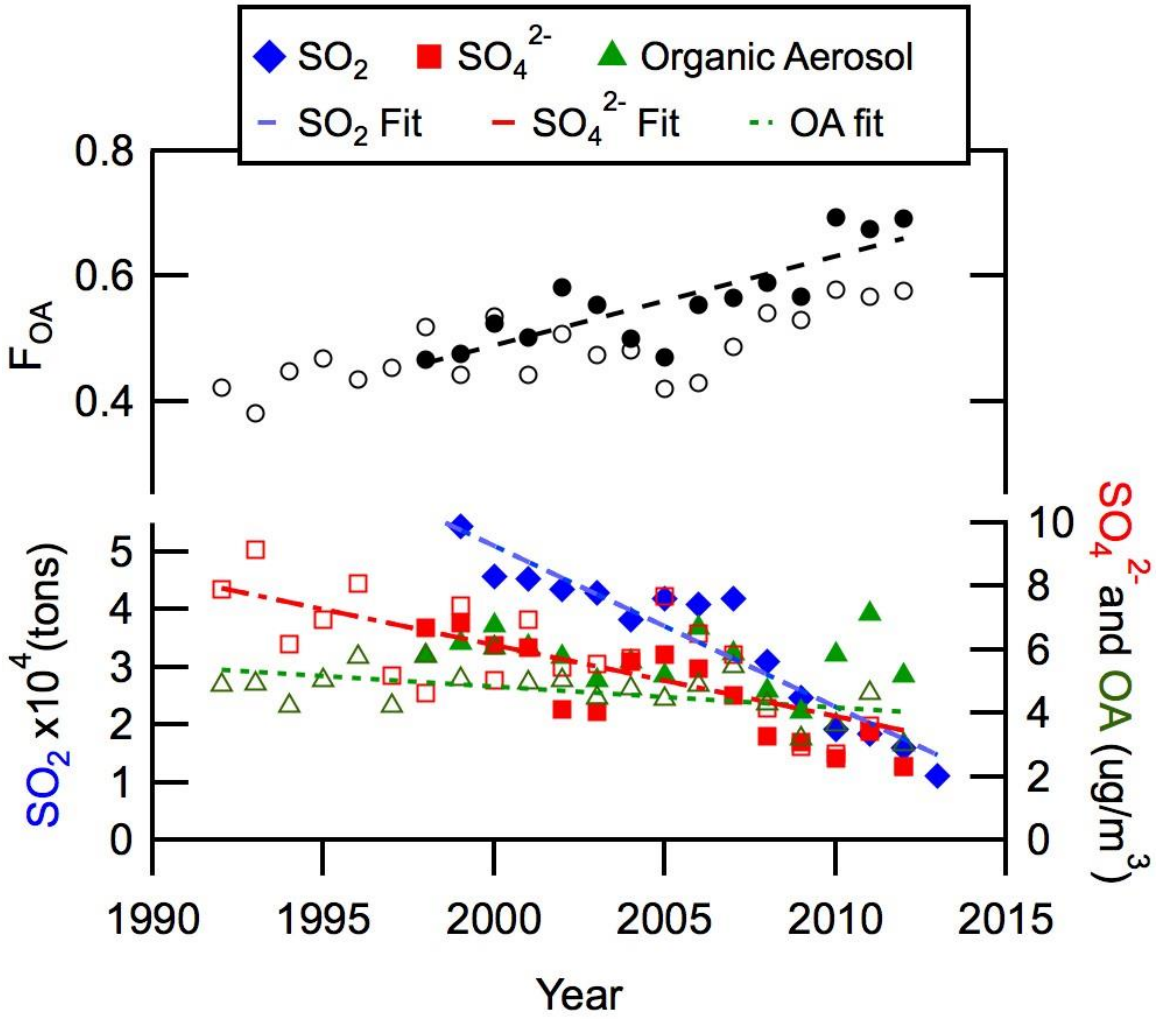

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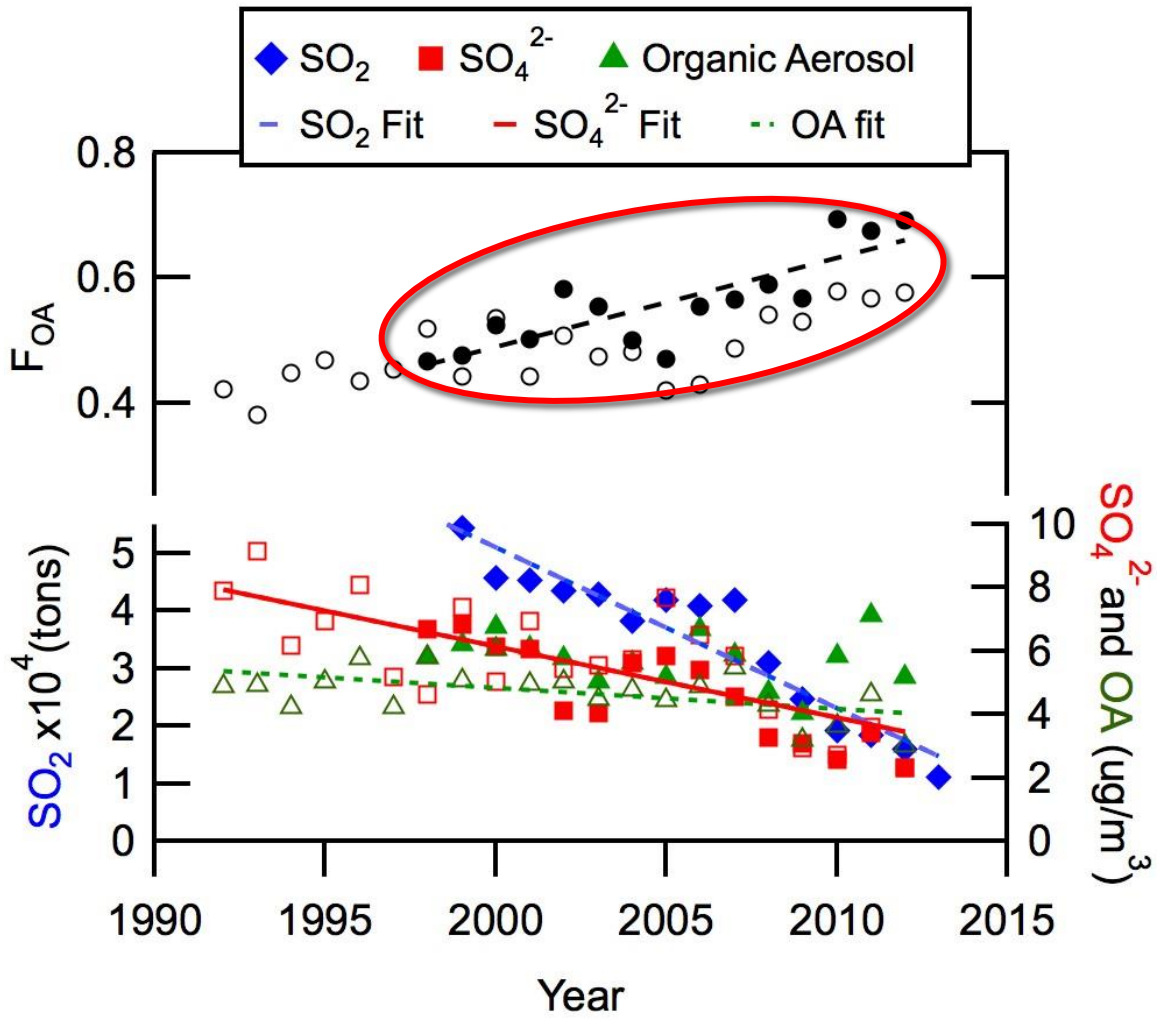

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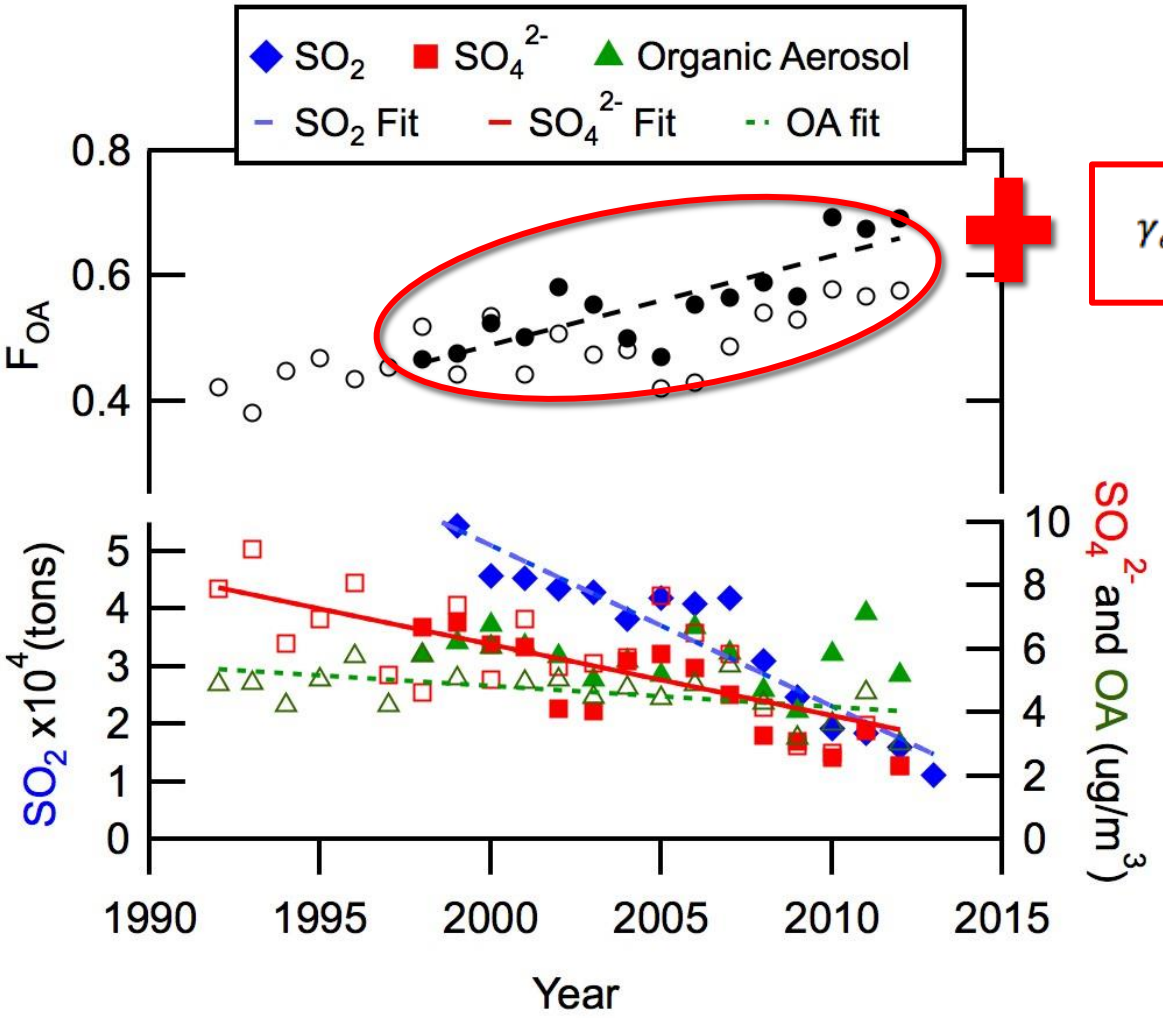
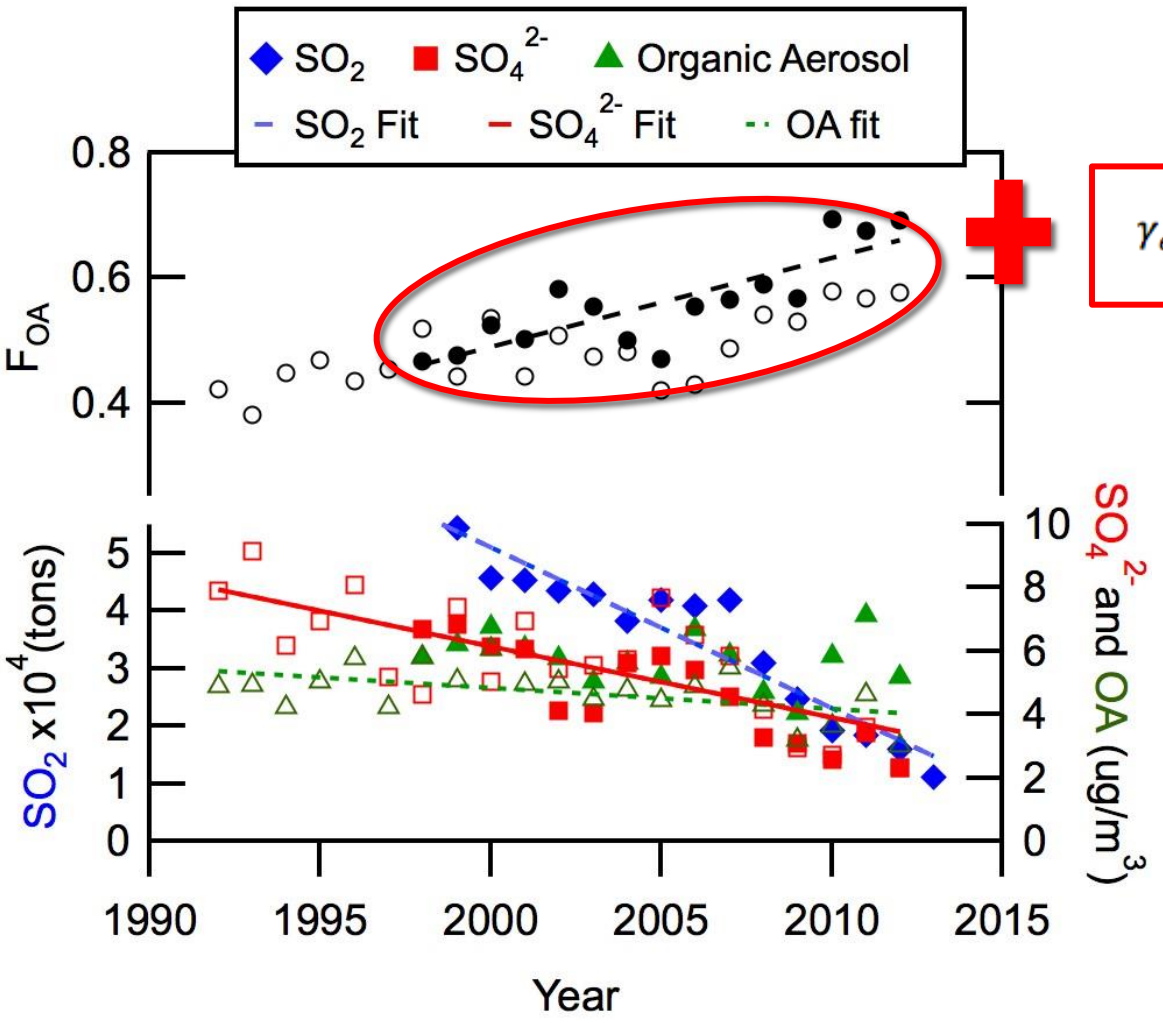


$$\gamma_{ext} = 0.88(\pm 0.03) - 0.65(\pm 0.04) \times F_{OA}$$

Historical Data



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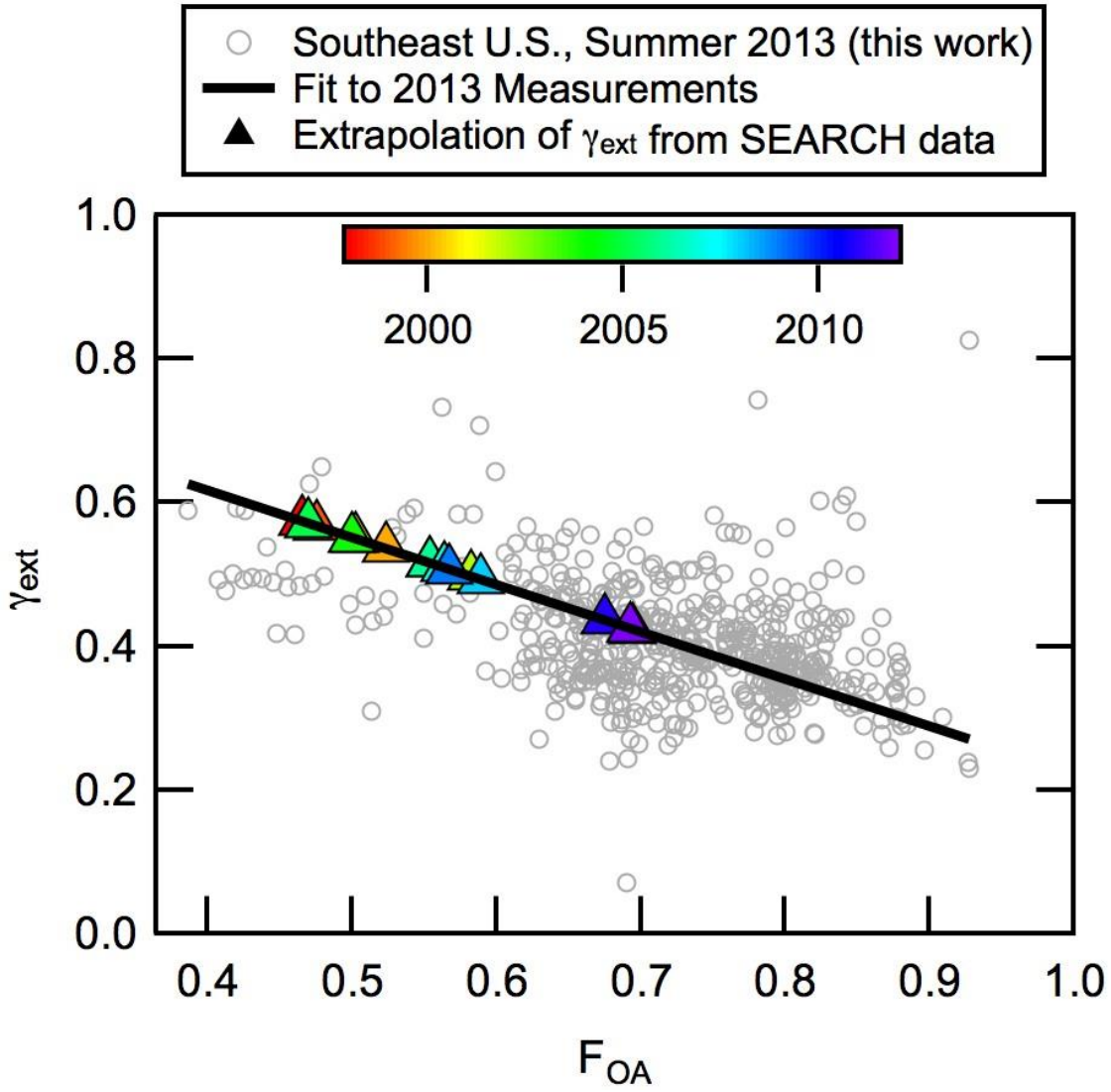
Historical Gamma Values

Decreased Extinction due to Decreased Hygroscopicity



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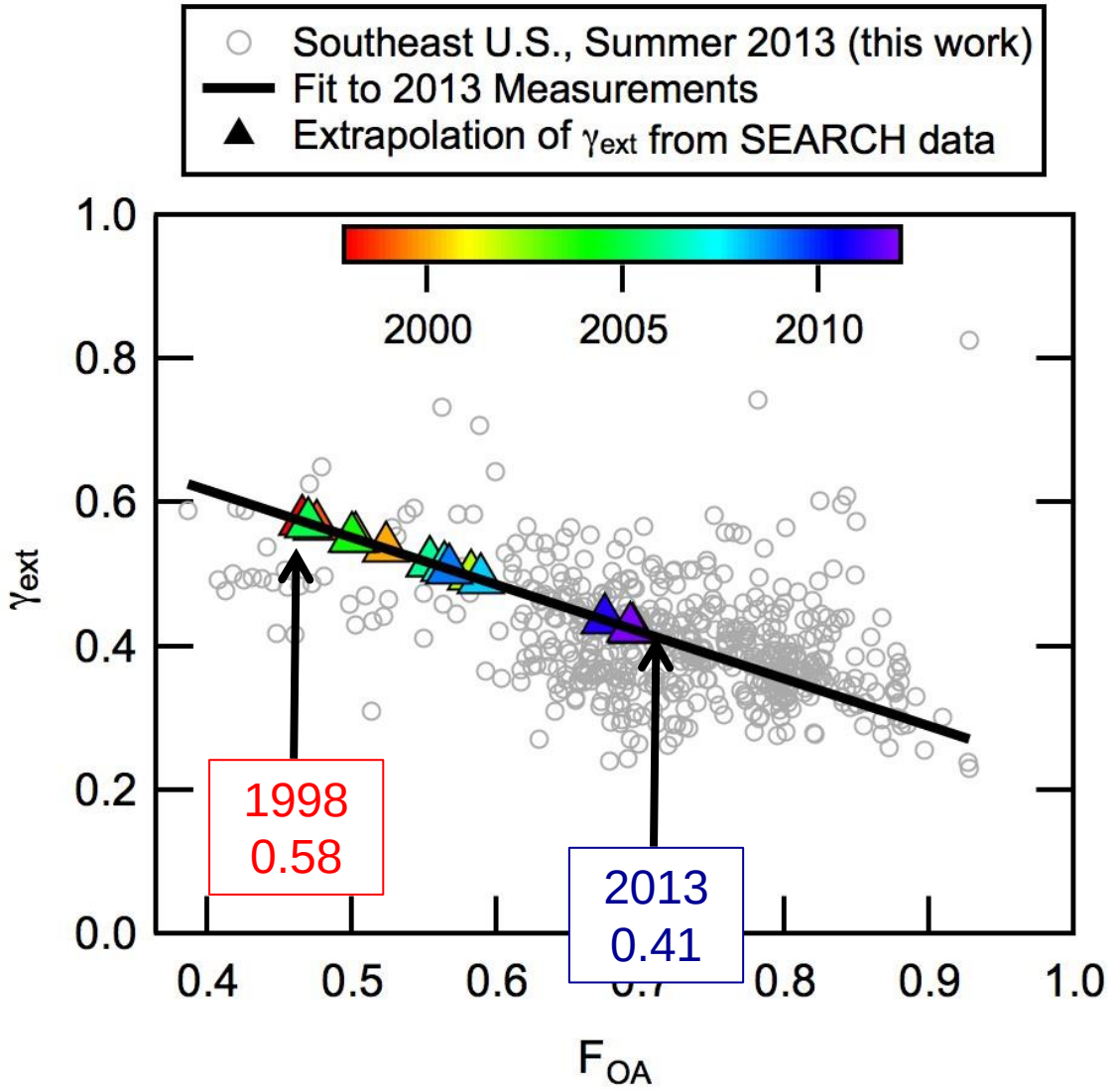
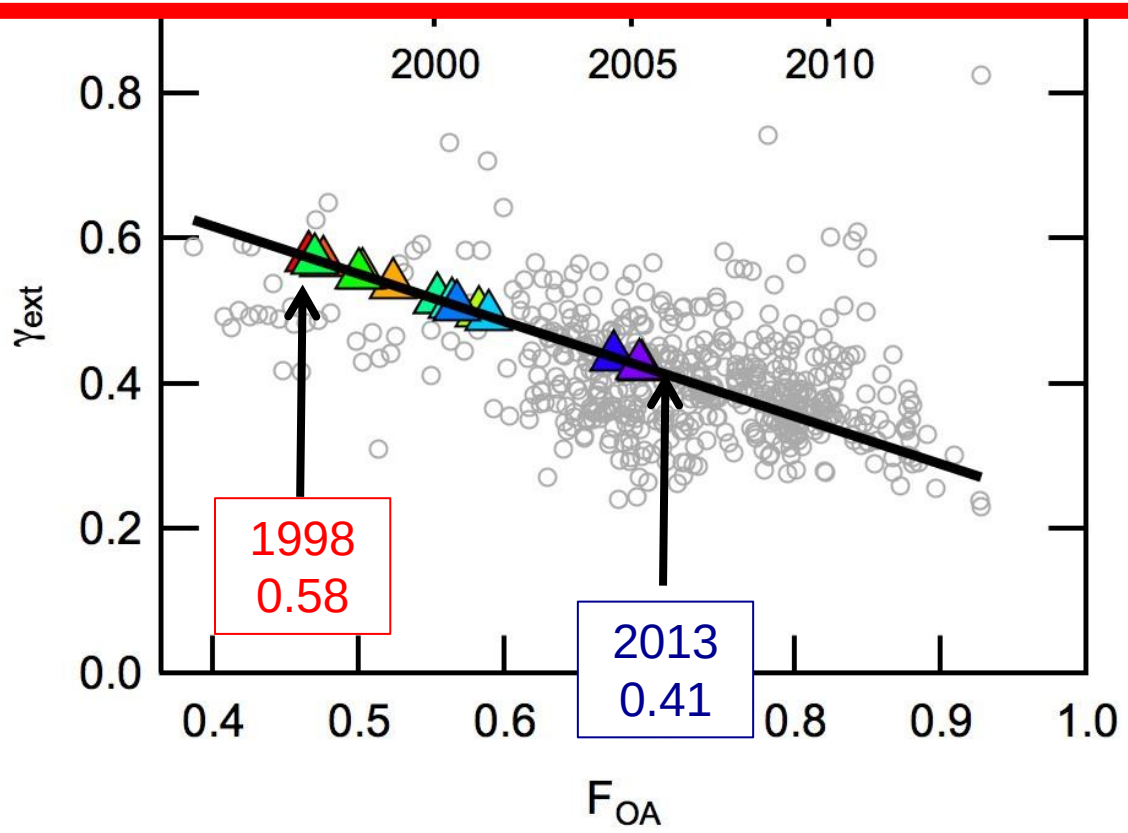


Decreased Extinction due to Decreased Hygroscopicity

-1.6(±0.3)% yr^{-1} decrease in the ambient extinction from 1998 to 2013 due to reduced hygroscopicity (sulfate)



Decreased Extinction due to Decreased Aerosol Mass Calculated from SEARCH Filter Data

$$
\begin{aligned}
b_{ext} = {} & 2.2 \times [\text{Small } (NH_4)_2SO_4] + \\
& 4.8 \times [\text{Large } (NH_4)_2SO_4] + \\
& 2.4 \times [\text{Small } (NH_4)NO_3] + \\
& 5.1 \times [\text{Large } (NH_4)NO_3] + \\
& 2.8 \times [\text{Small } POM] + \\
& 6.1 \times [\text{Large } POM] + \\
& 10 \times [LAC] + \\
& 1 \times [\text{Soil}] + \\
& 1.7 \times [\text{Sea Salt}] + \\
& 0.6 \times [\text{Coarse Mass}] +
\end{aligned}
$$

$$
\text{For } [\text{Total}] < 20\ \mu g/m^3 \begin{cases} [\text{Large}] = \dfrac{[\text{Total}]}{20} \times [\text{Total}] \\ [\text{Small}] = [\text{Total}] - [\text{Large}] \end{cases}
$$

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\text{For } [\text{Total}] \geq 20\ \mu g/m^3,\ [\text{Large}] = [\text{Total}]
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Pitchford et al., 2012

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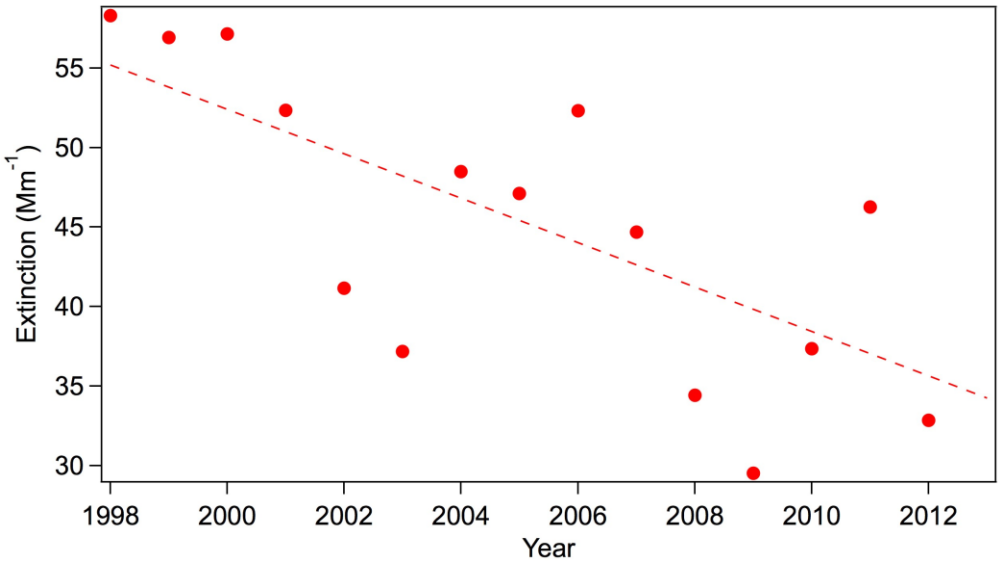

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Pitchford et al., 2012

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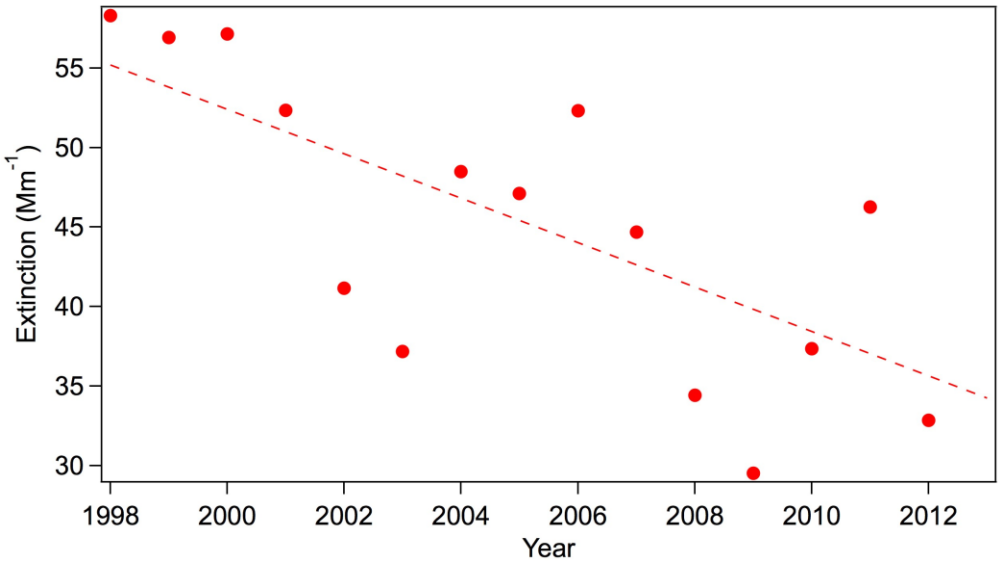
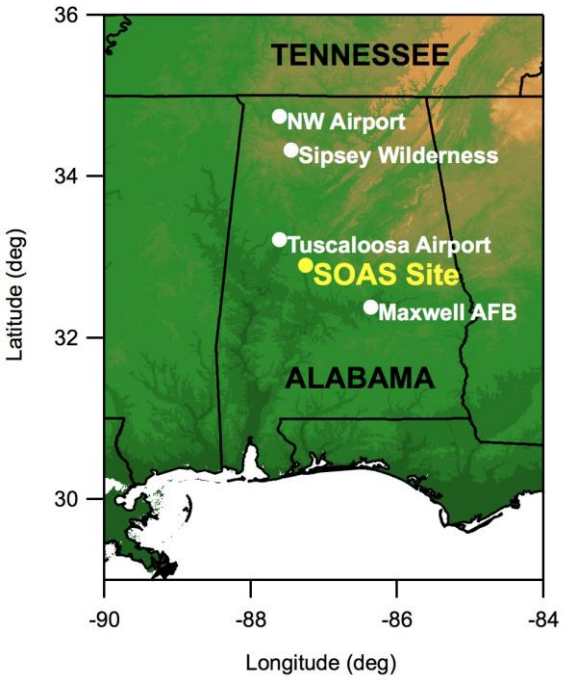


For [Total] $< 20\ \mu g/m^3$ { [Large] $= \frac{[\text{Total}]}{20} \times [\text{Total}]$

For

-3.1(±0.8)% yr^{-1} decrease in the ambient extinction from 1998 to 2013 due to decreased aerosol mass

Historical Trends in Visibility



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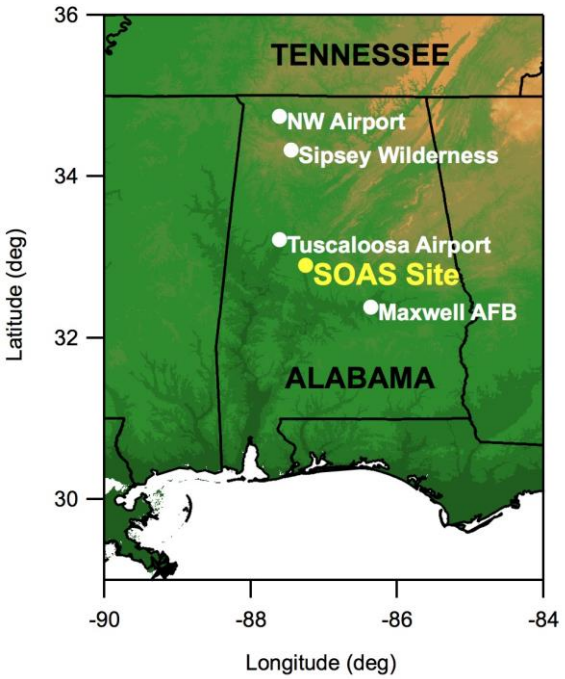

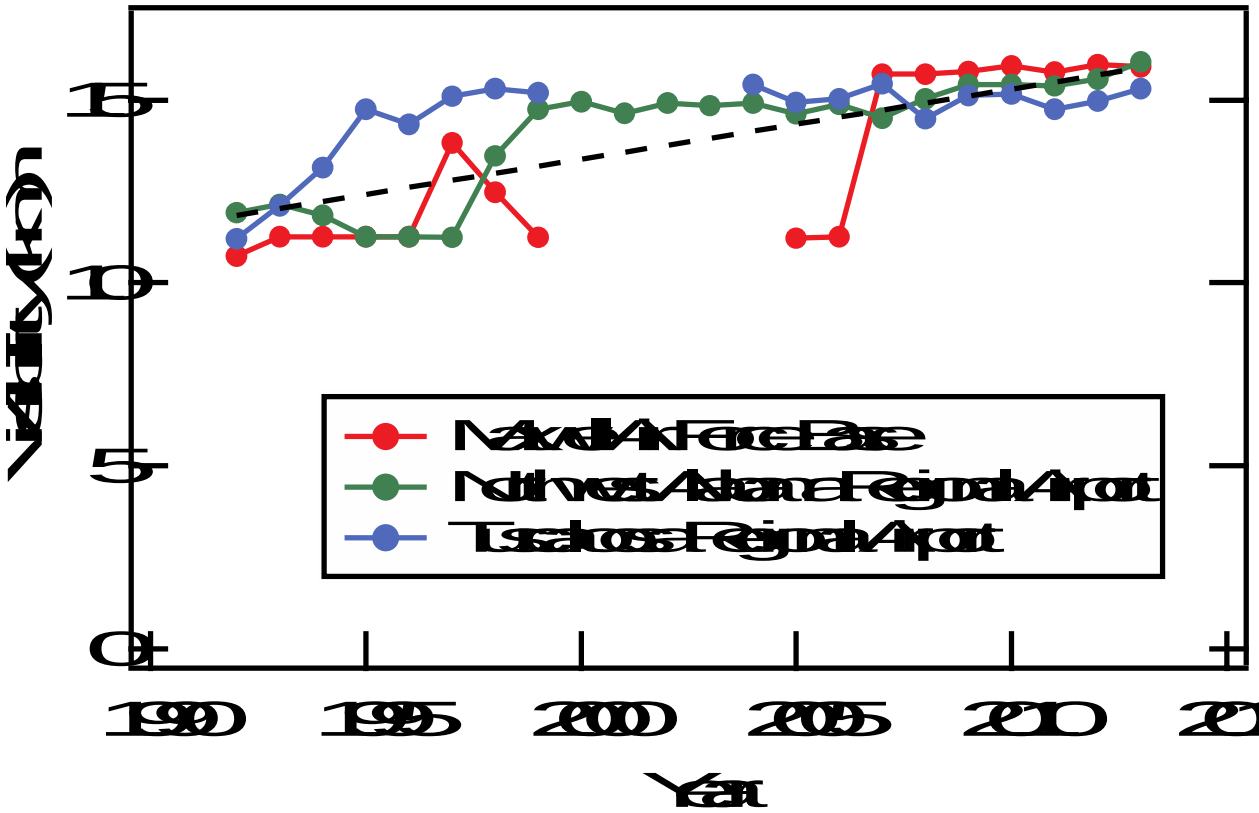

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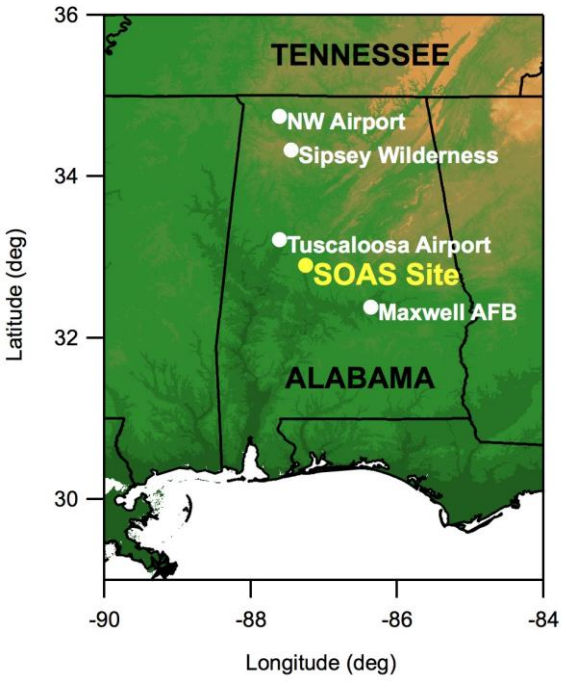
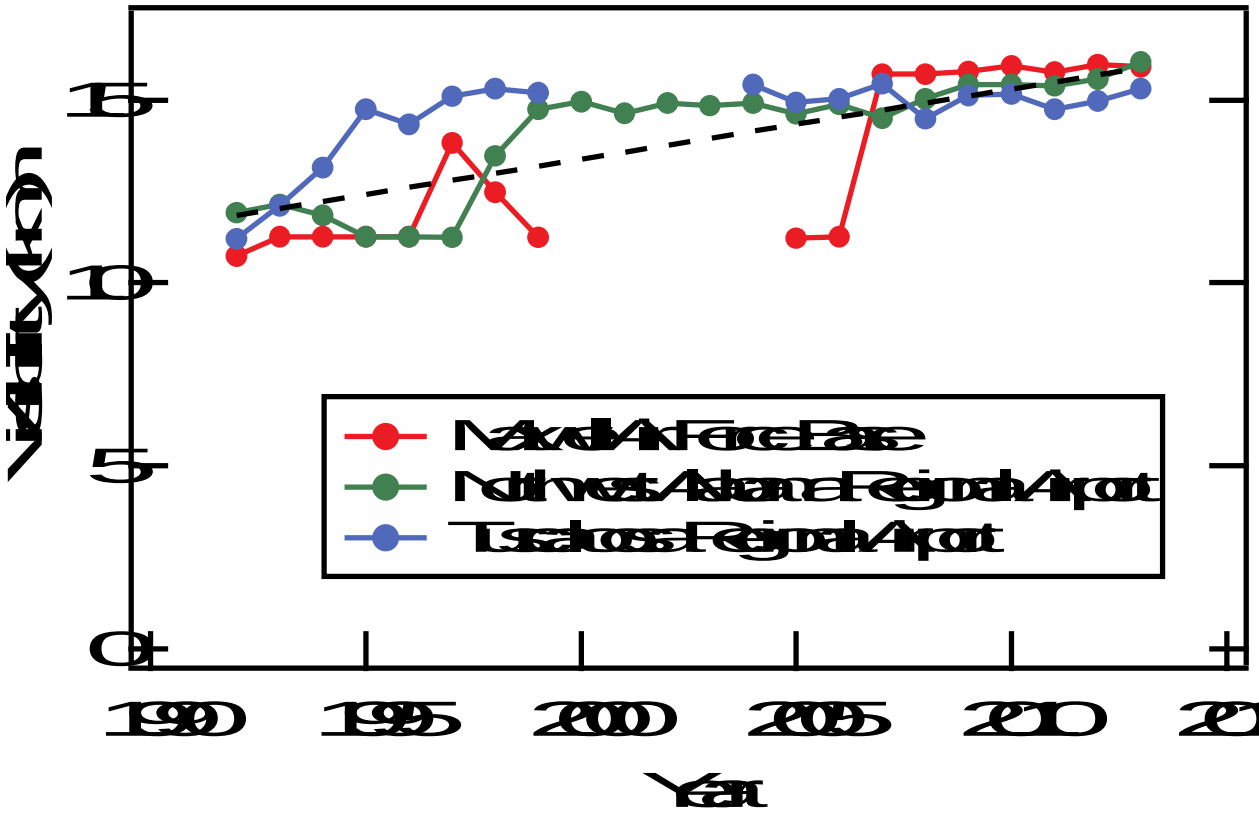


- 1.0(±0.2)% yr^{-1} improvement in visibility
- Visibility only reported up to 16 km



Conclusions

- Measurements of RH, aerosol extinction and F_{OA} from SOAS applied to historical aerosol composition data
- Trend of decreasing γ_{ext} with increasing F_{OA} observed
- Trend analysis from 1998-2013 of aerosol sulfate and organics:
  - -1.6(±0.3)% yr^{-1} decrease due to decreased hygroscopicity
  - -3.1(±0.8)% yr^{-1} decrease due to decreased aerosol mass
  - >1.0(±0.2)% yr^{-1} improvement in visibility
- Radiative forcing calculations underway

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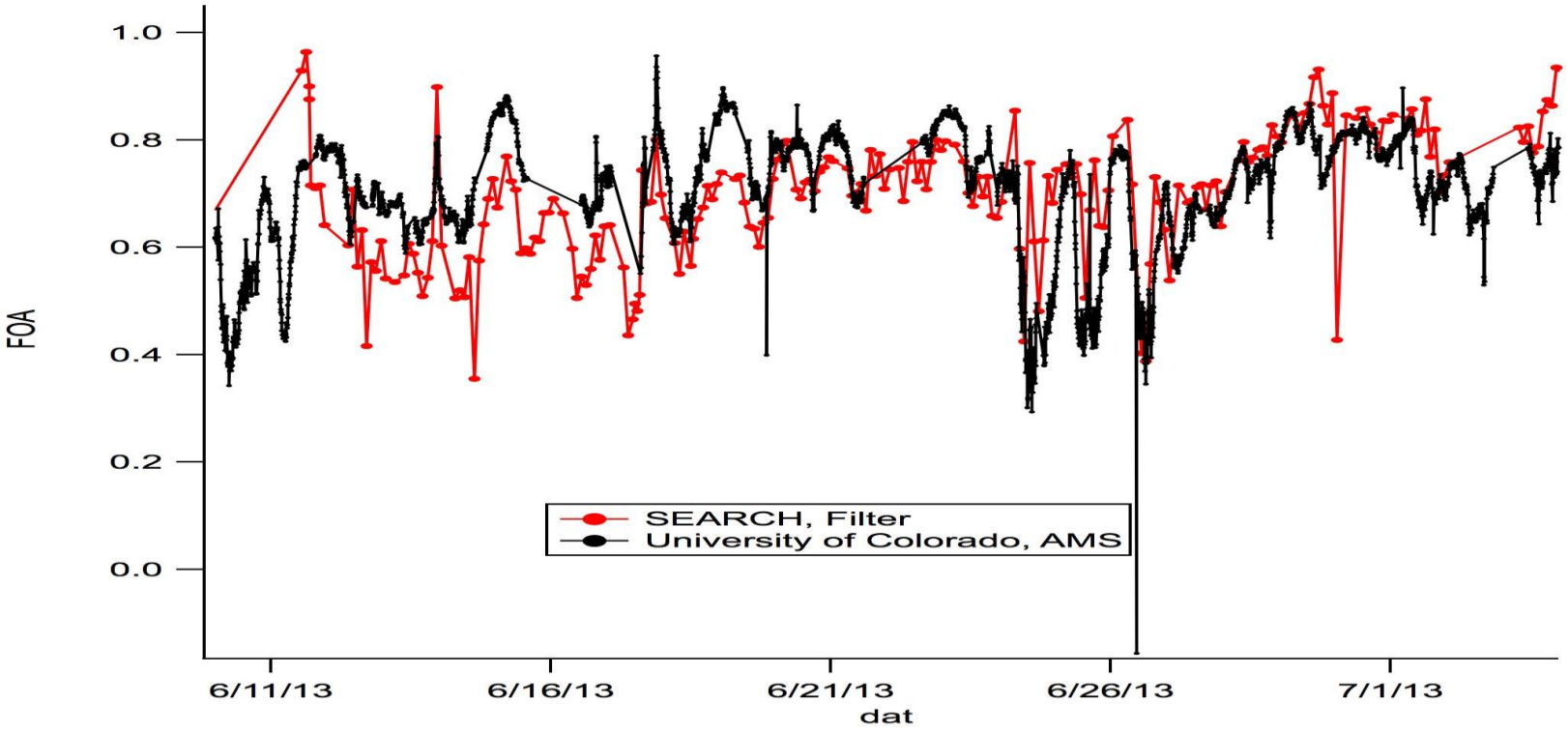
Acknowledgements

- Lead investigators for the SOAS campaign; AnnMarie Carlton, Allen Goldstein and Jose Jimenez
- SEARCH monitoring network
- Funding from the NOAA Atmospheric Chemistry, Carbon Cycle and Climate Program


1.0
0.8
0.6
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FOA
SEARCH, Filter
University of Colorado, AMS
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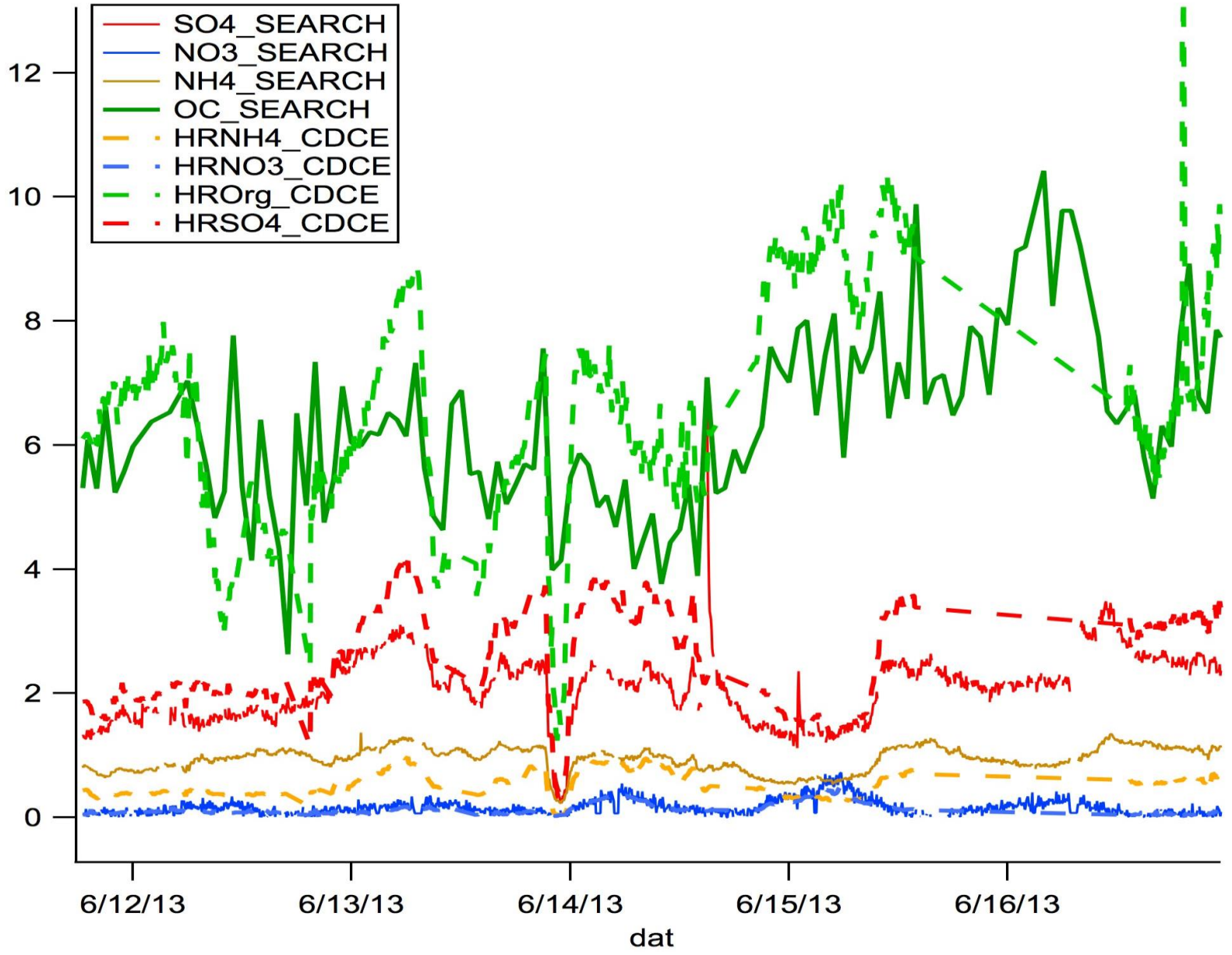
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