Update on Nephelometer Analysis: A potential approach for correcting the IMPROVE Equation

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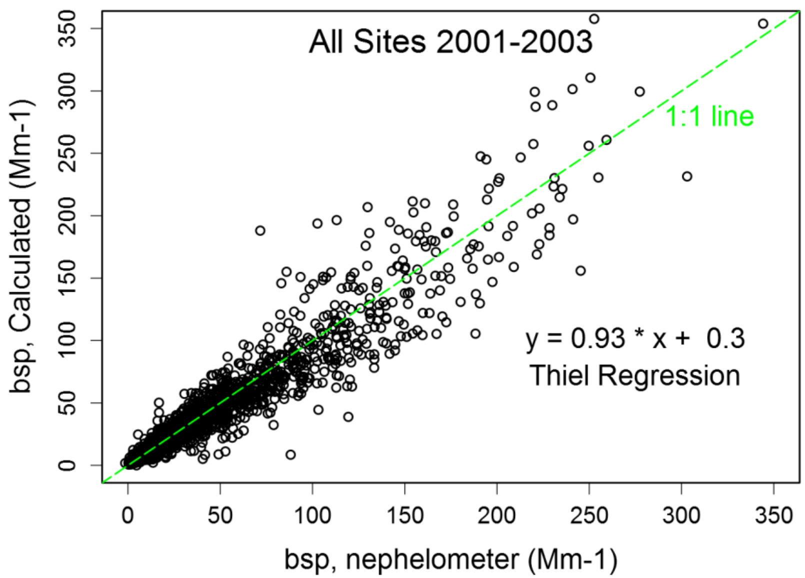
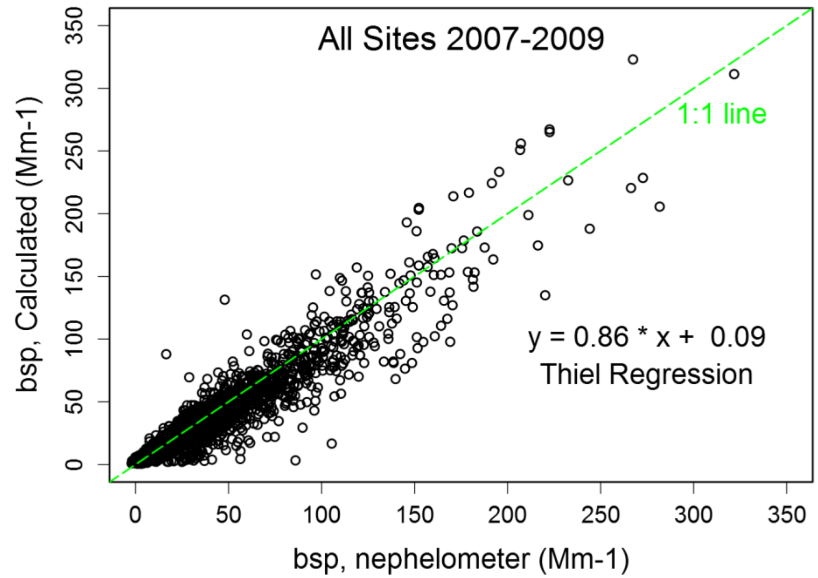
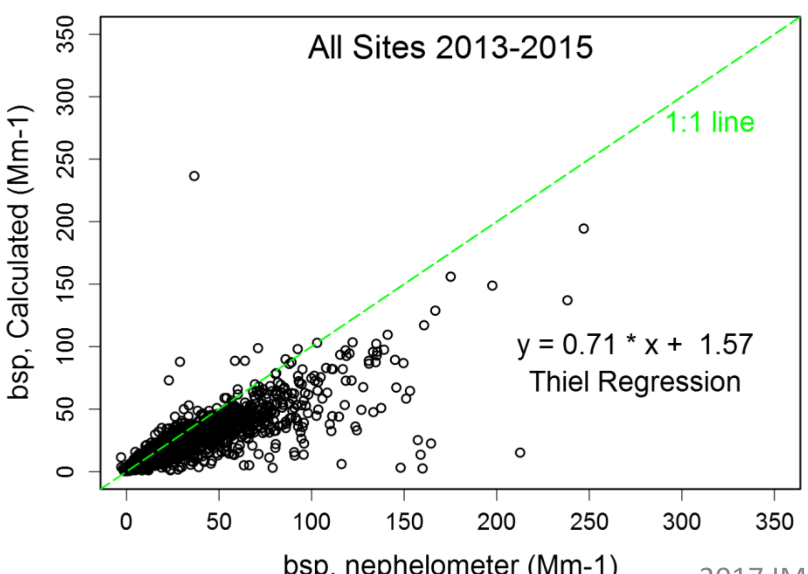
Recap from last year: Disagreement between measured & reconstructed light scattering



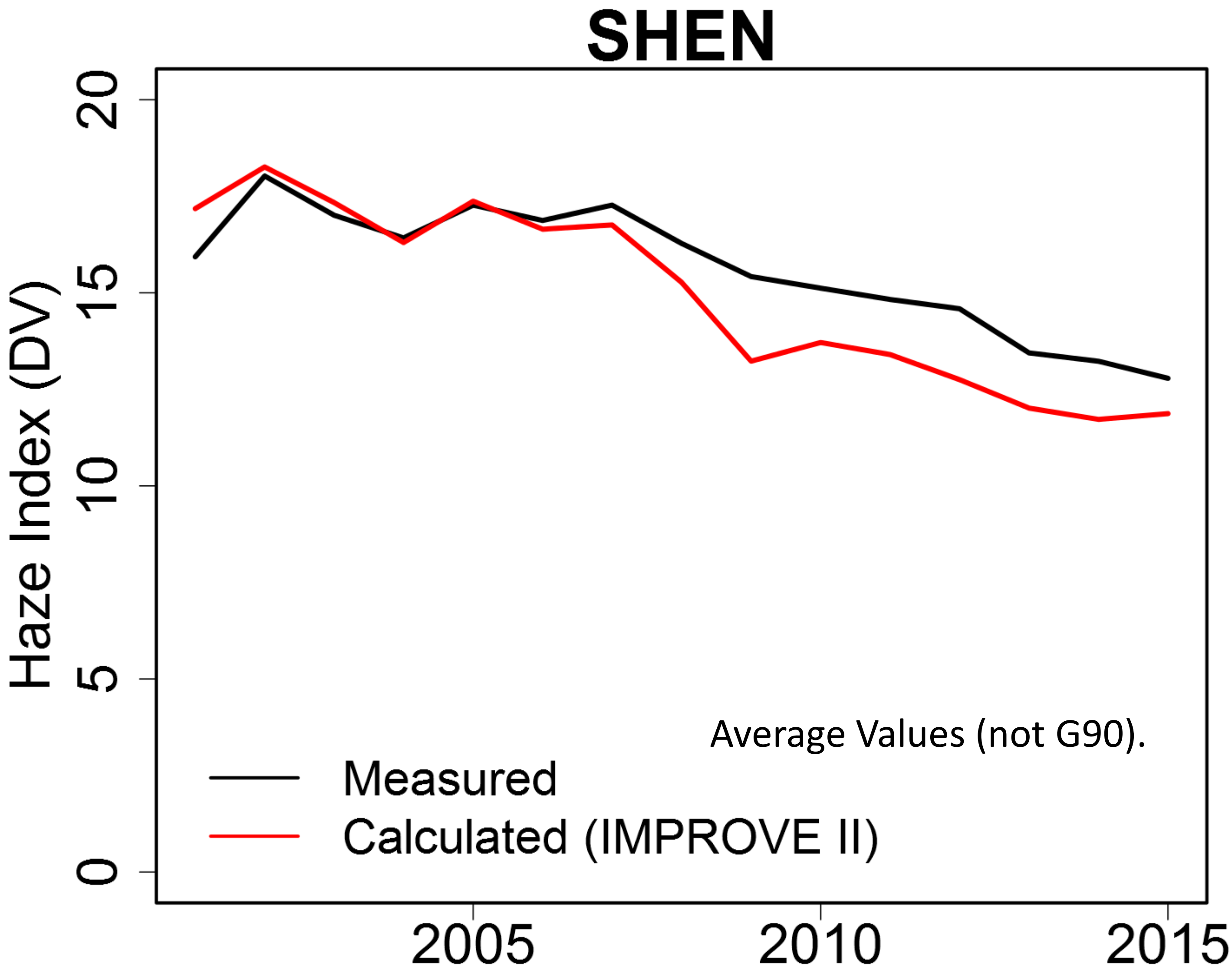




IMPROVE Equation is under-predicting measured scattering.

Agreement between measured and calculated b_{sp} is getting progressively worse.

Potential Implications



At parks with high conc. in early years + decreasing conc., progress may be over-estimated.

2^{nd} IMPROVE Equation

$$b_{sp} \approx 2.2 \times f_s(RH) \times [Small\ Sulfate] + 4.8 \times f_L(RH) \times [Large\ Sulfate]$$
$$+ 2.4 \times f_s(RH) \times [Small\ Nitrate] + 5.1 \times f_L(RH) \times [Large\ Nitrate]$$
$$+ 2.8 \times [Small\ Organic\ Mass] + 6.1 \times [Large\ Organic\ Mass]$$
$$+ 1 \times [Fine\ Soil] + 1.7 \times f_{SS}(RH) \times [Sea\ Salt] + 0.5 \times 0.6 \times [Coarse\ Mass]$$

Compare calculated scattering to measurements from nephelometer.

- Remove EC contribution to extinction
- Remove Rayleigh Scattering term
- Remove NO_2 absorption
- calculated coarse mass = 0.5 × 0.6 × [Coarse Mass]
  ➔ Account for truncation error in nephelometer

2^{nd} IMPROVE Equation

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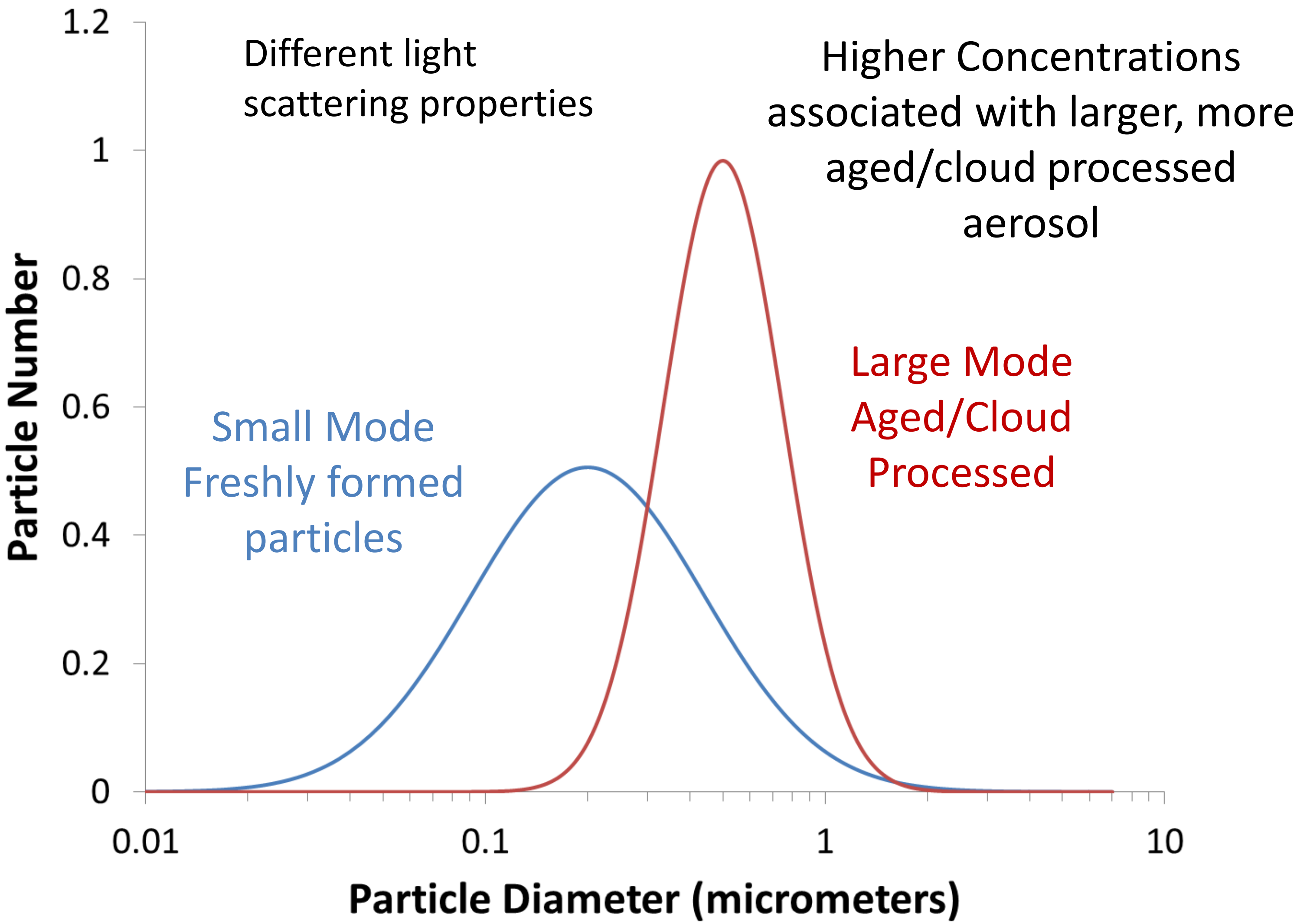
$$+ 2.8 \times [Small\ Organic\ Mass] + 6.1 \times [Large\ Organic\ Mass]$$
$$+ 1 \times [Fine\ Soil] + 1.7 \times f_{SS}(RH) \times [Sea\ Salt] + 0.5 \times 0.6 \times [Coarse\ Mass]$$

$$[Large\ Sulfate] = \frac{[Total\ Sulfate]}{20} \times [Total\ Sulfate]$$
$$[Small\ Sulfate] = [Total\ Sulfate] - [Large\ Sulfate]$$

If the total > 20 μg m^{-3}, all mass assumed to be in the large mode.

Large and Small Modes

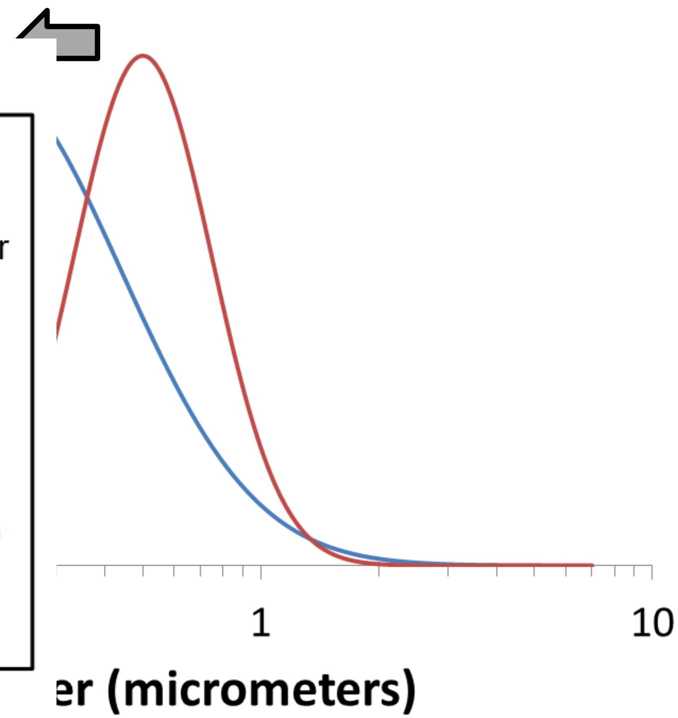


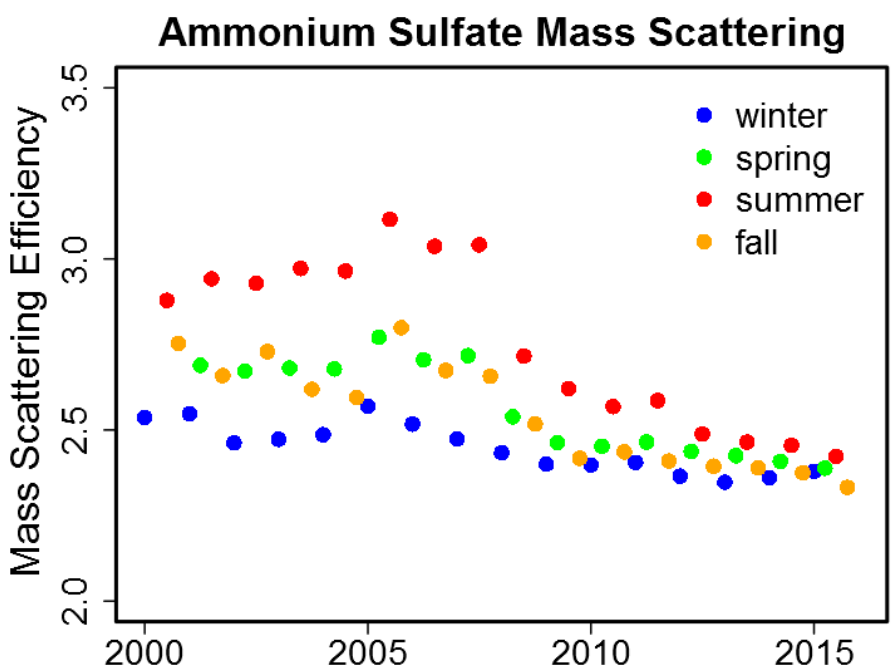
Large and Small Modes

As concentrations have decreased:

- less aerosol in the large mode
- relatively more in the smaller mode

Should there be a lower fraction of aged aerosol now?





Focus on the Split Component Factor of 20

$$[Large\ Sulfate] = \frac{[Total\ Sulfate]}{20} \times [Total\ Sulfate]$$

$$[Small\ Sulfate] = [Total\ Sulfate] - [Large\ Sulfate]$$

Would a value other than 20 improve agreement between measured and calculated b_{sp}?

Multiple Linear Regression

$b_{sp}^{*} = b_{sp} - [SOIL] - 1.7 \times fRH_{SS} \times [SS] - 0.6 \times 0.5 \times [CM]$

$AS = 2.2 \times fRH_s \times [ammSO_4]_s + 4.8 \times fRH_l \times [ammSO_4]_l$

$= 2.2 \times fRH_s \times (1 - \frac{[ammSO_4]}{x}) \times [ammSO_4] +$

$4.8 \times fRH_l \times \frac{[ammSO_4]}{x} \times [ammSO_4]$

$= \underbrace{2.2 \times fRH_s \times [ammSO_4]}_{\text{Move to } b_{sp}^{*} \text{ term.}} + \frac{1}{x} \times \underbrace{[ammSO_4]^2 \times (4.8 \times fRH_l - 2.2 \times fRH_s)}_{AS}$

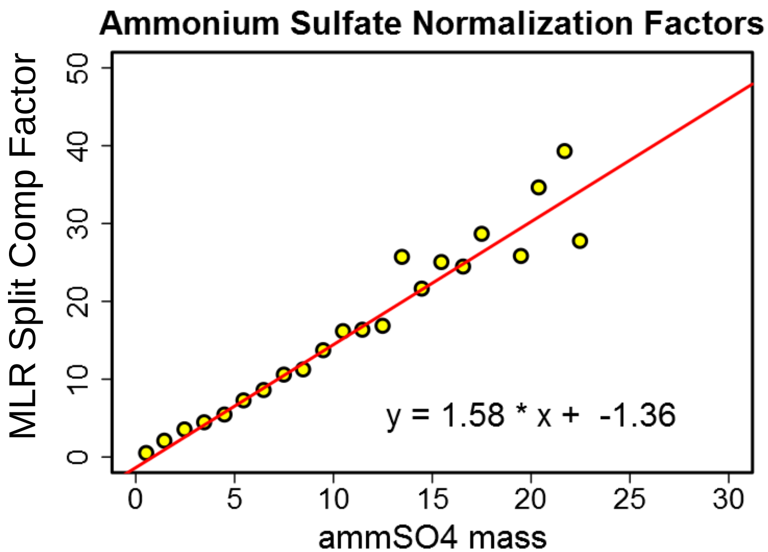
Repeat for $ammNO_3$ and OMC.

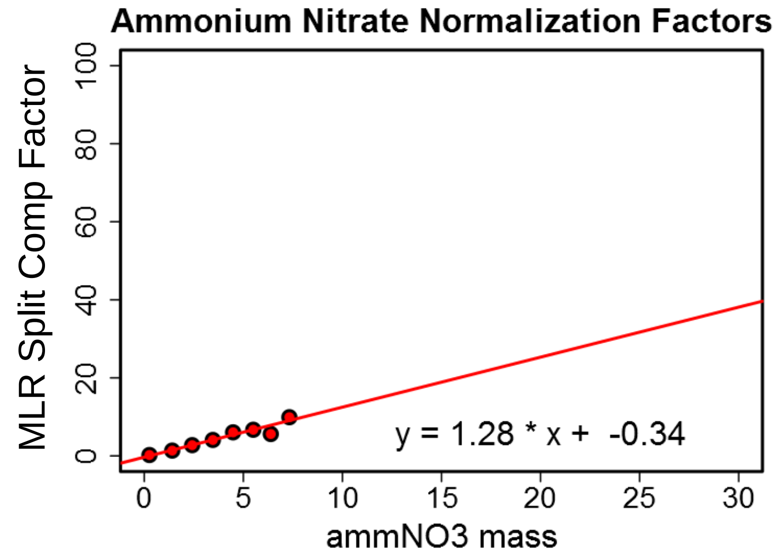
Regression: $b_{sp}^{*} = c_1 \times AS + c_2 \times AN + c_3 \times OMC$

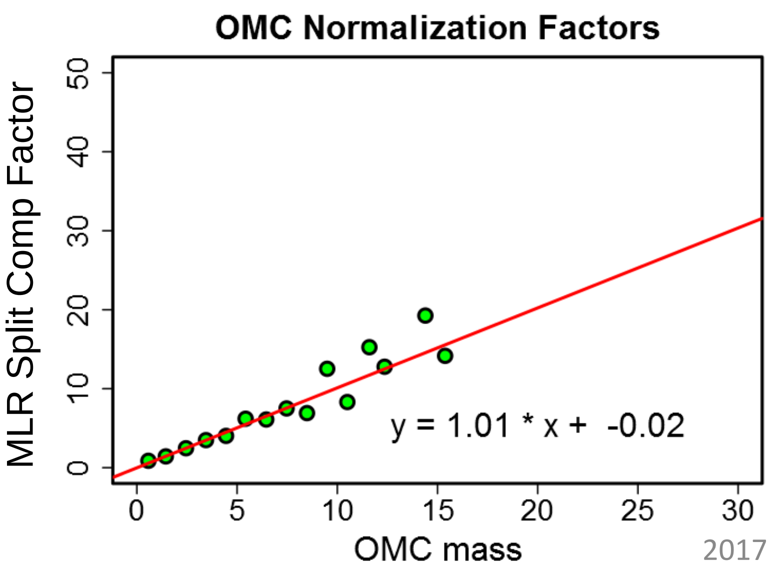
Normalization Factors: $ammSO_4 = \frac{1}{c_1}$; $ammNO_3 = \frac{1}{c_2}$; $OMC = \frac{1}{c_3}$

Focus on species with split component (ammSO4, ammNO3, OMC). Ignore all of the other issues that Jenny presented.

Initial Results







Best fits for Split Component Factors scale with mass.

These relationships change over time (not shown).

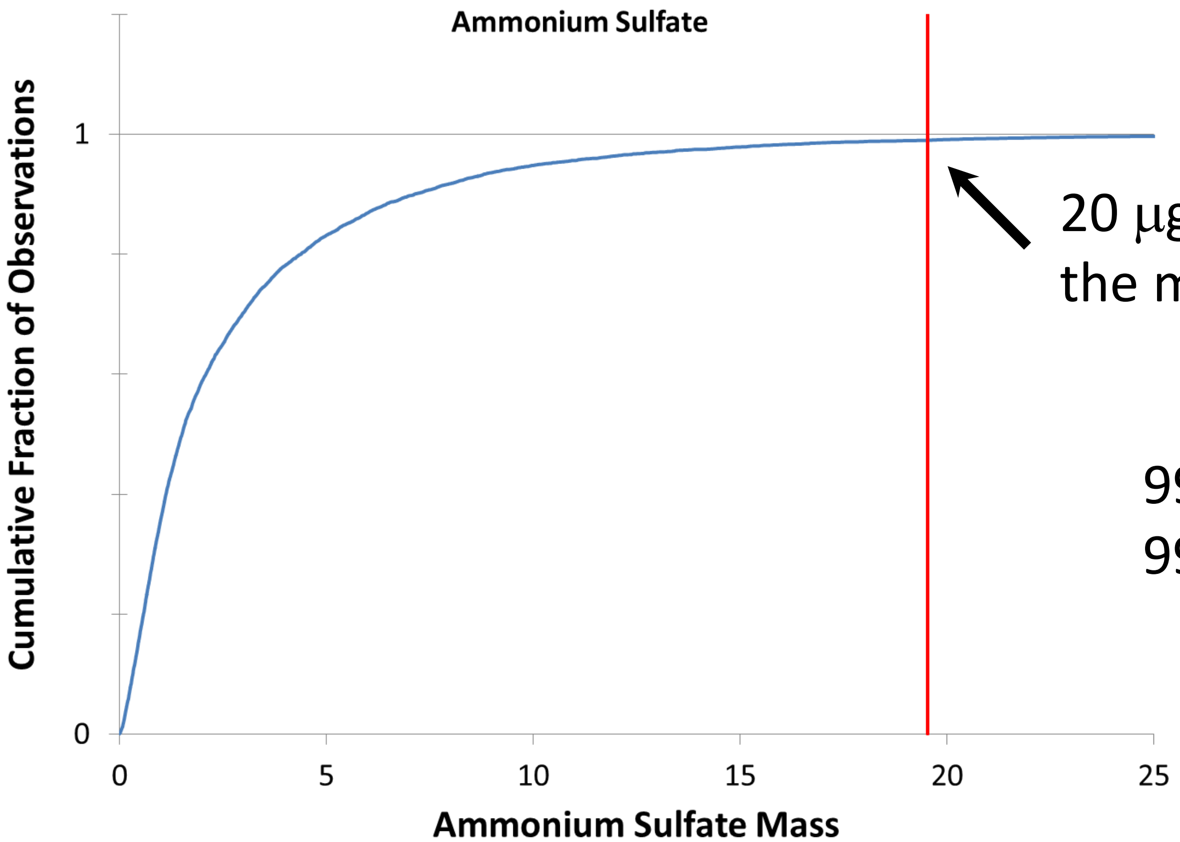
How do we deal with a mass-dependent split component factor that has changed over time?

What were the IMPROVE masses for the data used in developing the 2nd IMPROVE Equation?

Data used roughly from ~1995-2003

ACAD	Acadia National Park	Maine
BIBE	Big Bend National Park	Texas
BOWA	Boundary Waters Canoe Area	Minnesota
CORI	Columbia River Gorge	Washington
DOSO	Dolly Sods/Otter Creek Wilderness	West Virginia
GICI	Gila Wilderness	New Mexico
GRCA	Grand Canyon National Park	Arizona
GRGU	Great Gulf Wilderness	New Hampshire
GRSM	Great Smoky Mountains	Tennessee
JARB	Jarbidge Wilderness	Nevada
LOPE	Lone Peak Wilderness	Utah
LYBR	Lye Brook Wilderness	Vermont
MACA	Mammoth Cave National Park	Kentucky
MORA	Mount Rainier National Park	Washington
MOZI	Mount Zirkel Wilderness	Colorado
OKEF	Okefenokee National Wildlife Refuge	Georgia
SHEN	Shenandoah National Park	Virginia
SHRO	Shining Rock Wilderness	North Carolina
SNAP	Snoqualamie Pass Wilderness	Washington
THIS	Three Sisters Wilderness	Oregon
UPBU	Upper Buffalo Wilderness	Arkansas

Measurements used for 2nd IMPROVE Equation paper (Pitchford et al.) (~1995-2003)

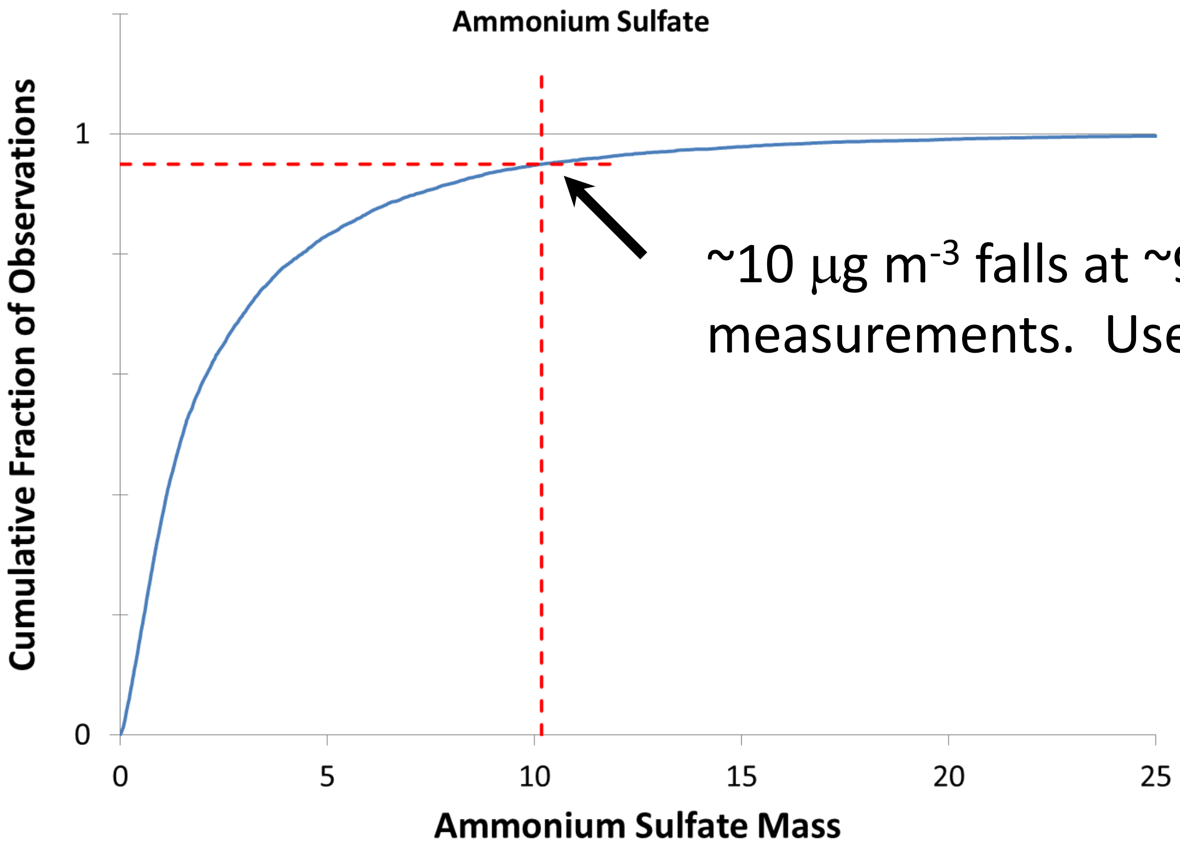


20 $\mu g\ m^{-3}$ falls at ~99% of the measurements

99% ammNO3: 5 $\mu g\ m^{-3}$

99% OMC: 10 $\mu g\ m^{-3}$

Measurements used for 2nd IMPROVE Equation paper (Pitchford et al.)

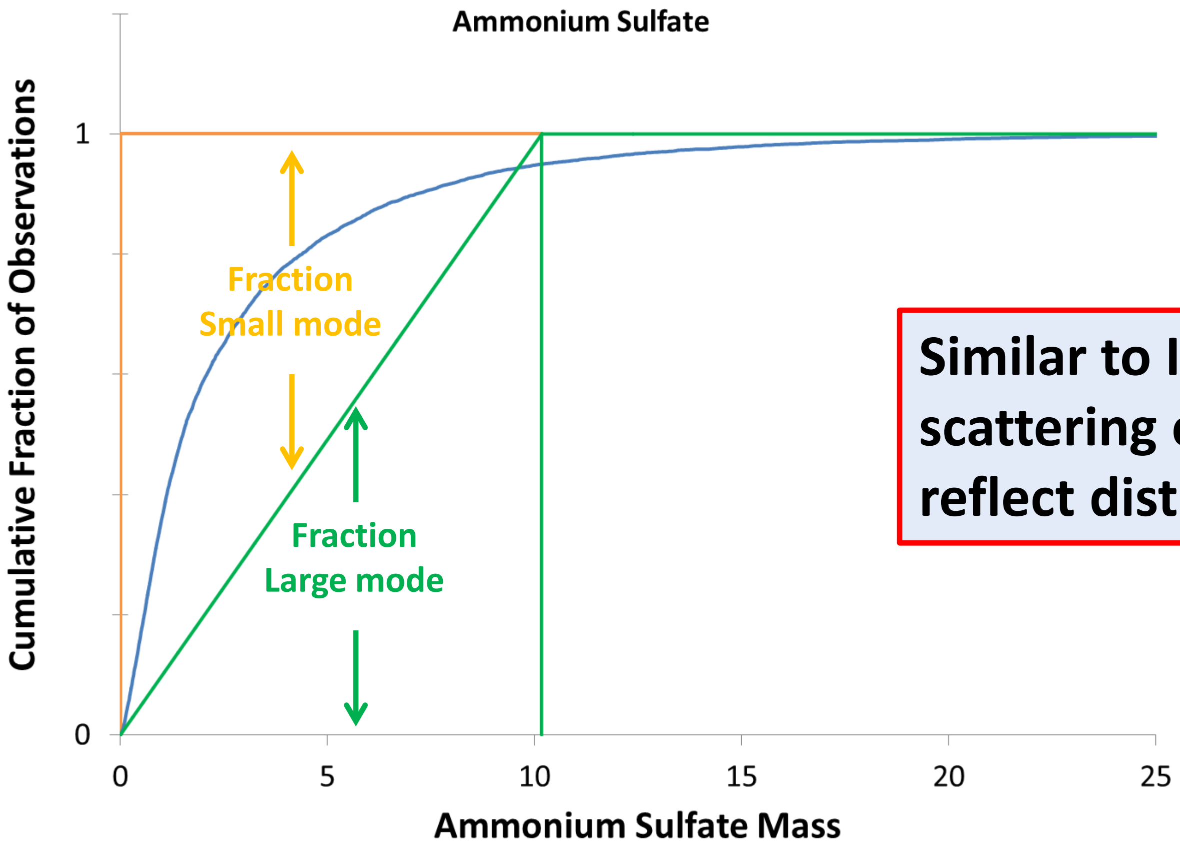


~10 μg m^{-3} falls at ~95% of the measurements. Use 95%.

*95% gives better results for later years.

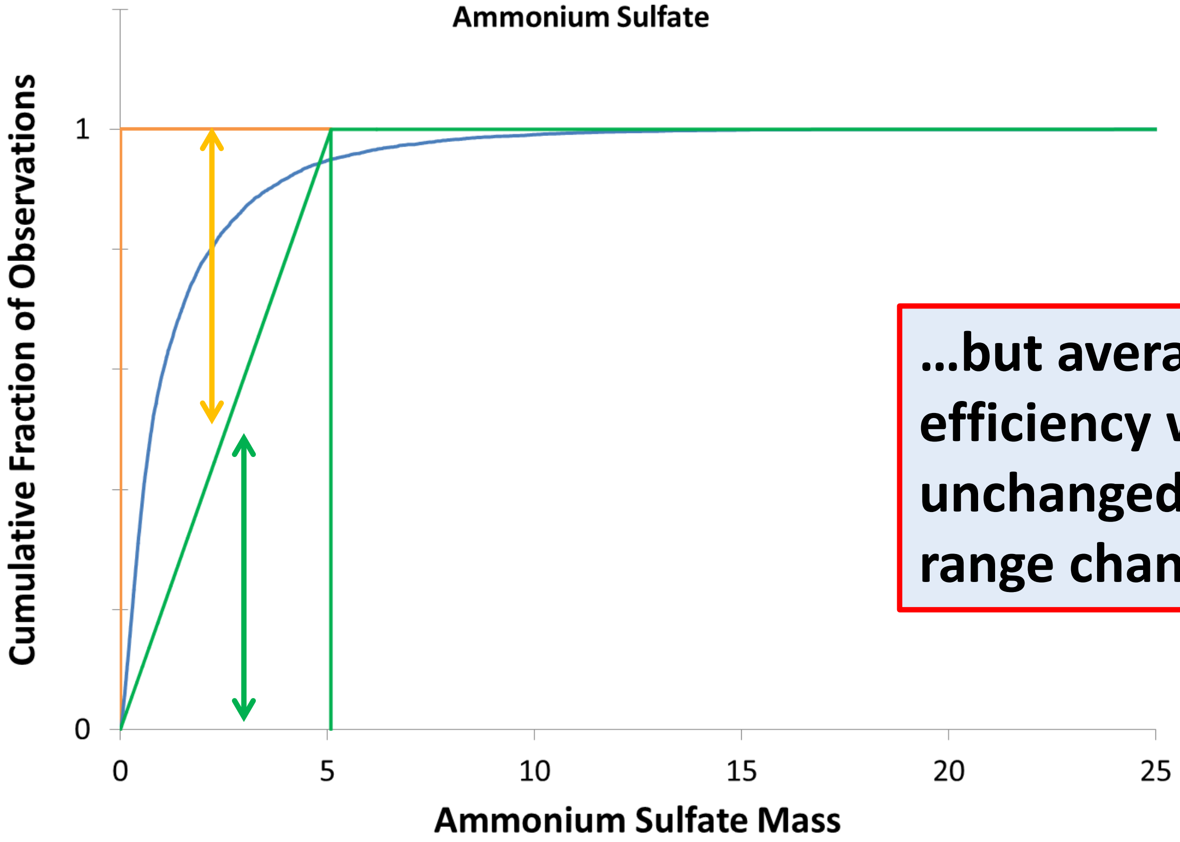
Use 95% of annual mass distribution as the denominator in the size split calculation.

95% of mass distribution as split component denominator



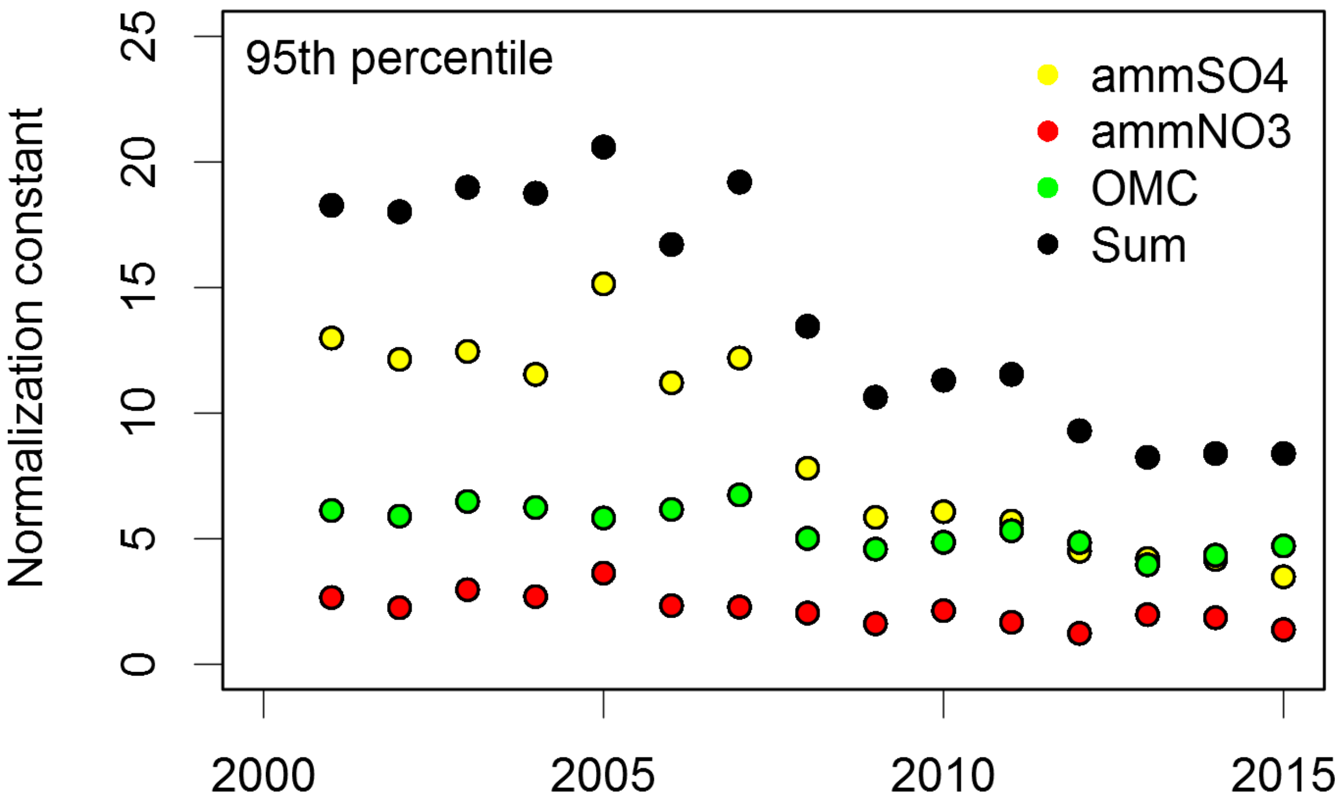
Similar to IMPROVE II, each year scattering calculations will reflect distribution of masses…

95% of mass distribution as split component denominator



...but average mass scattering efficiency will remain roughly unchanged even as the mass range changes over the years.

Use 95% of species mass at a given site on a given year as Split Component Factor

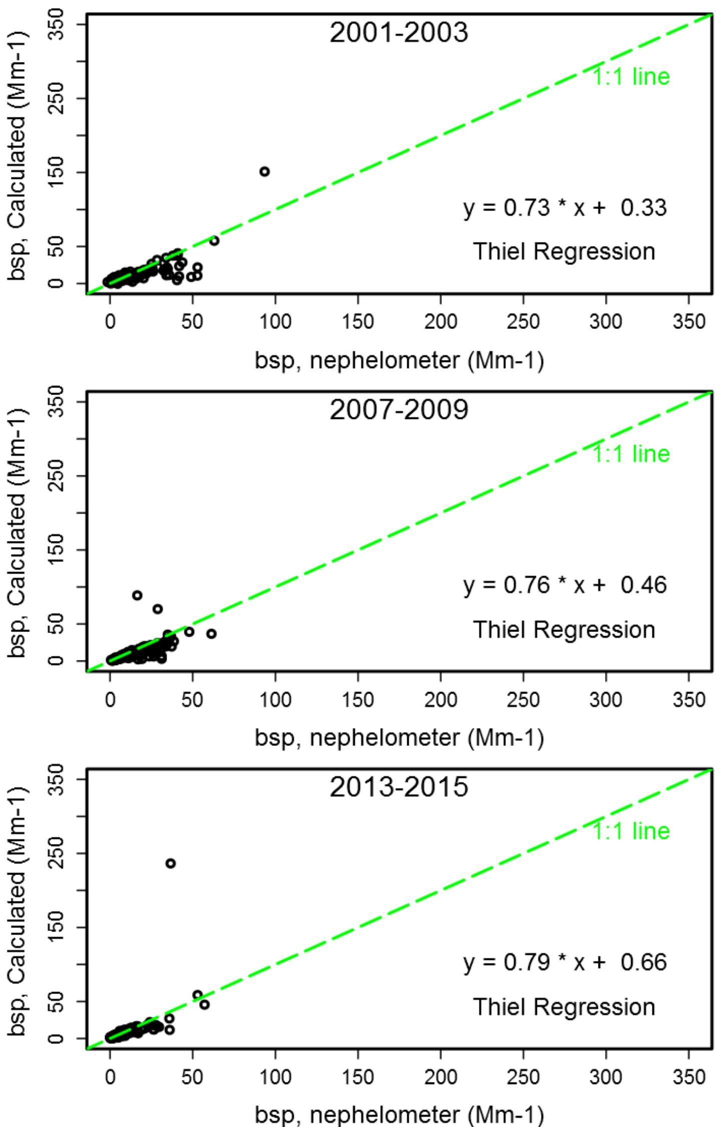


ACAD – Acadia
BIBE – Big Bend
GLAC – Glacier
GRBA – Great Basin
GRCA – Grand Canyon
GRSM – Great Smoky
MACA – Mammoth Cave
MORA – Mount Rainier
NACA – National Capital (DC)
ROMO – Rocky Mtn
SHEN - Shenandoah

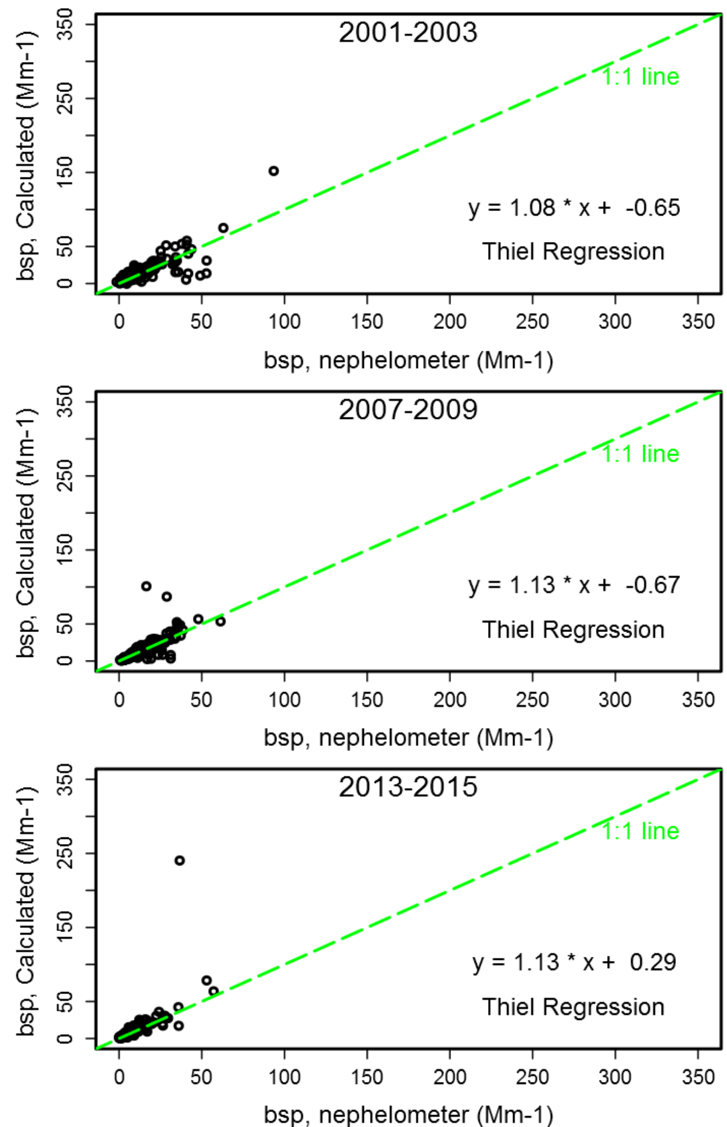
Plot shows 95% for all parks explored. For analysis, will use 95% on a park by park basis for each species.

Measured vs Calculated for Grand Canyon

2nd IMPROVE Equation



95% for Split Component



At GRCA:

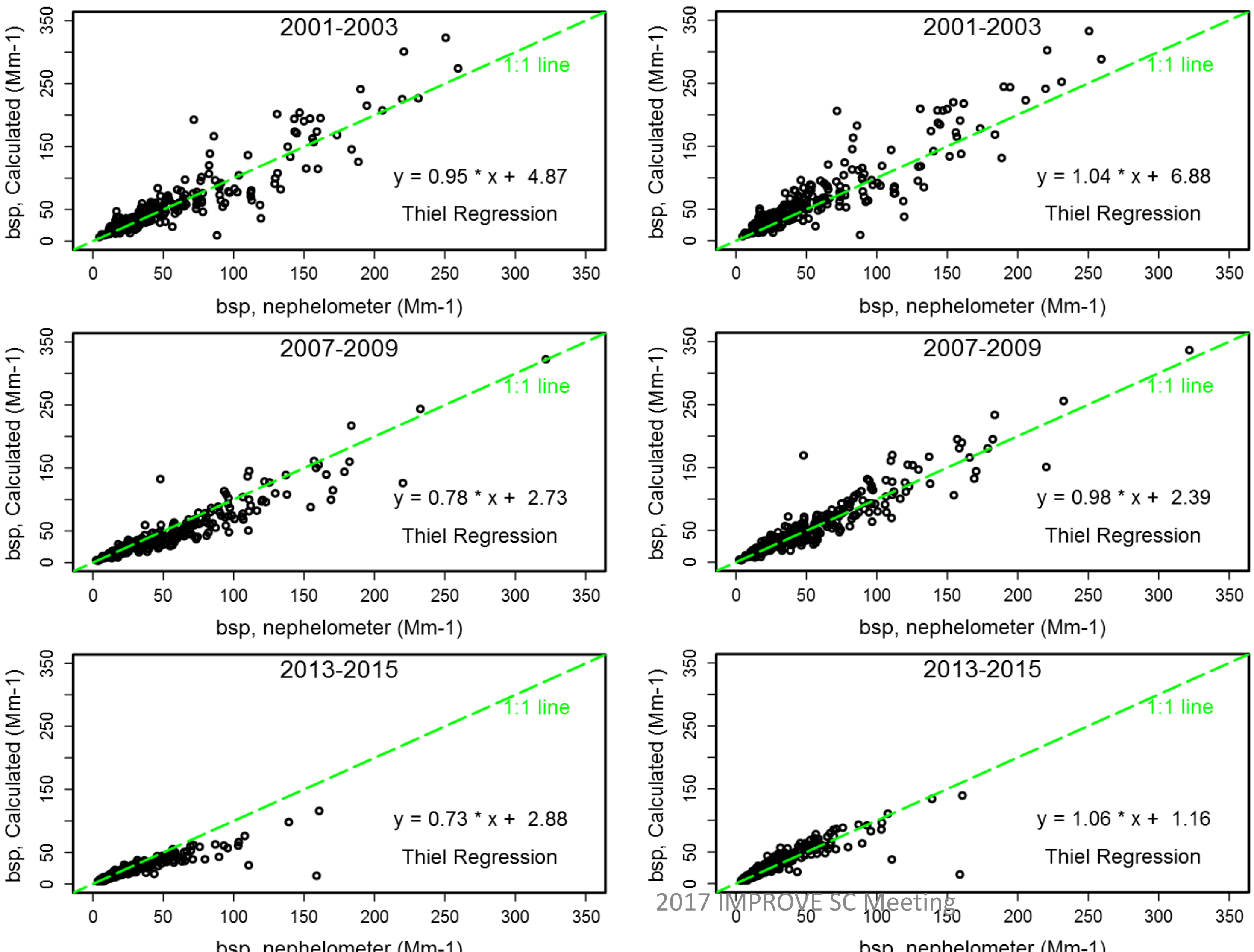
Low Mass.

IMPROVE II underestimates scattering, even in early years.

Revised approach overestimates scattering, but better agreement.

Measured vs Calculated for Shenandoah

2nd IMPROVE Equation | **95% for Split Component**



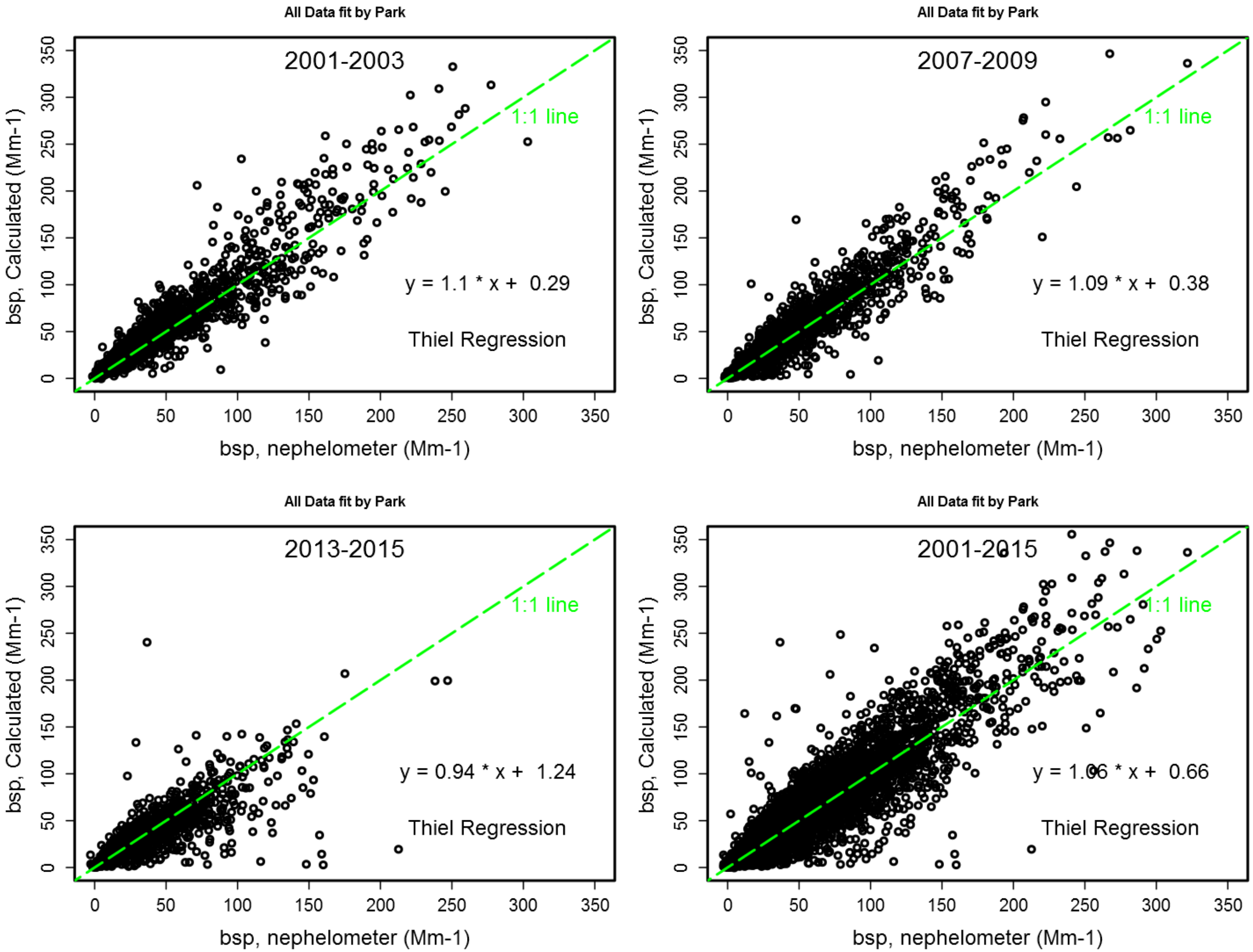
At SHEN:

Higher Mass in early years.

IMPROVE II underestimates scattering in later years.

Revised approach has much better agreement.

Measured vs Calculated for all Parks



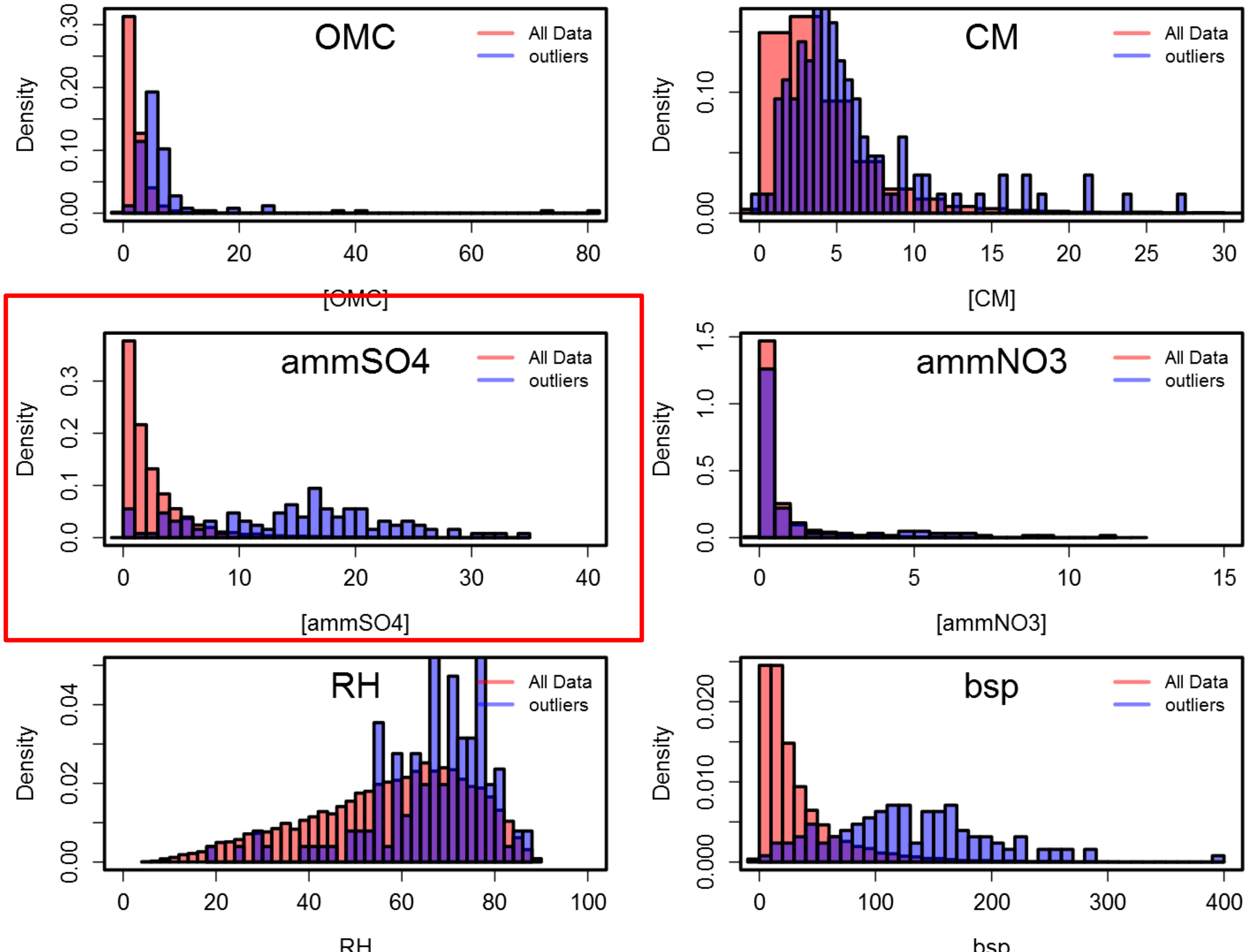
At all parks, overall better agreement.

Temporal trend still apparent.

Changing Roc?

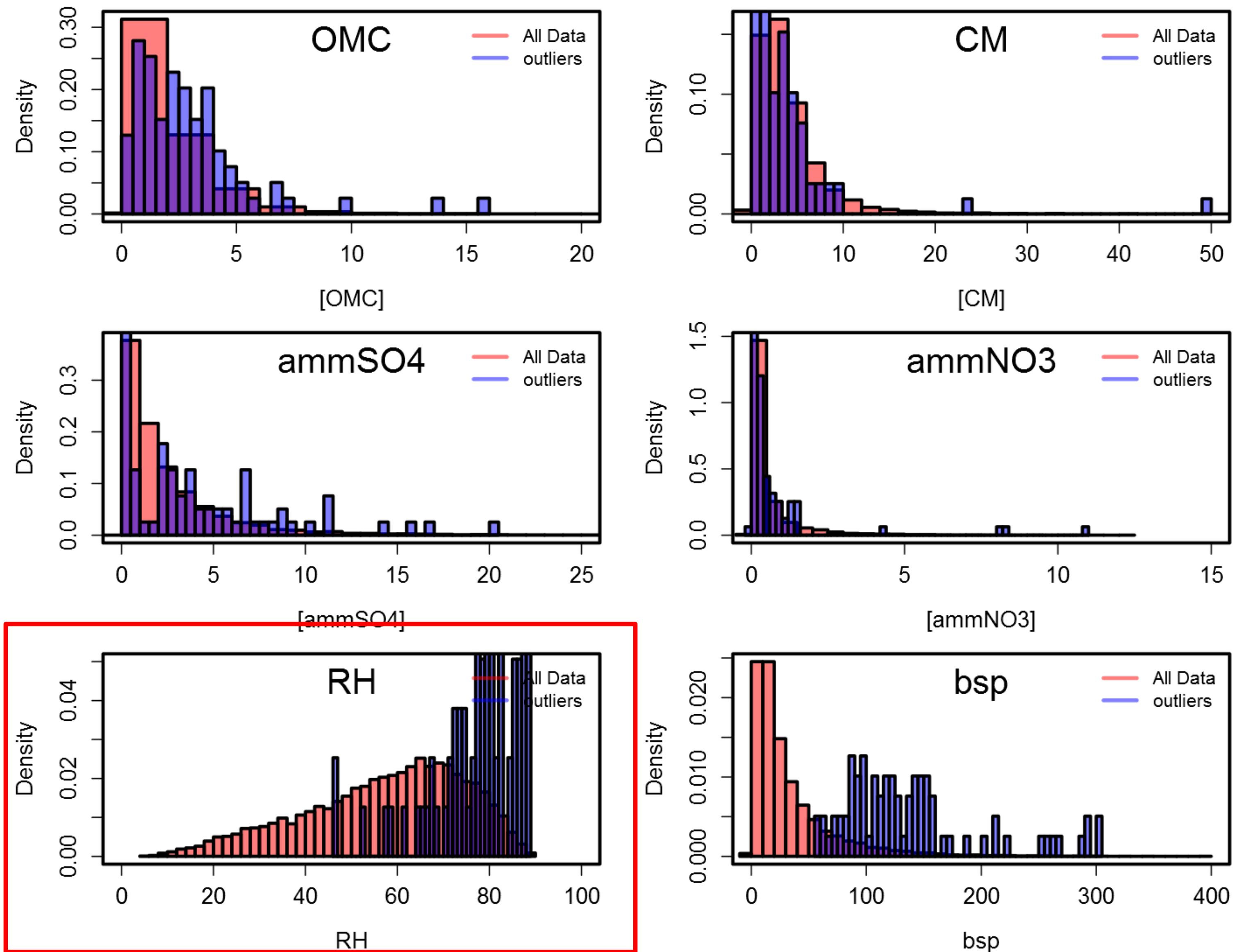
Something else?

Outliers: Recon. bsp – Meas. bsp **> 50 Mm^{-1}**
i.e. when we're over-predicting (absolute)



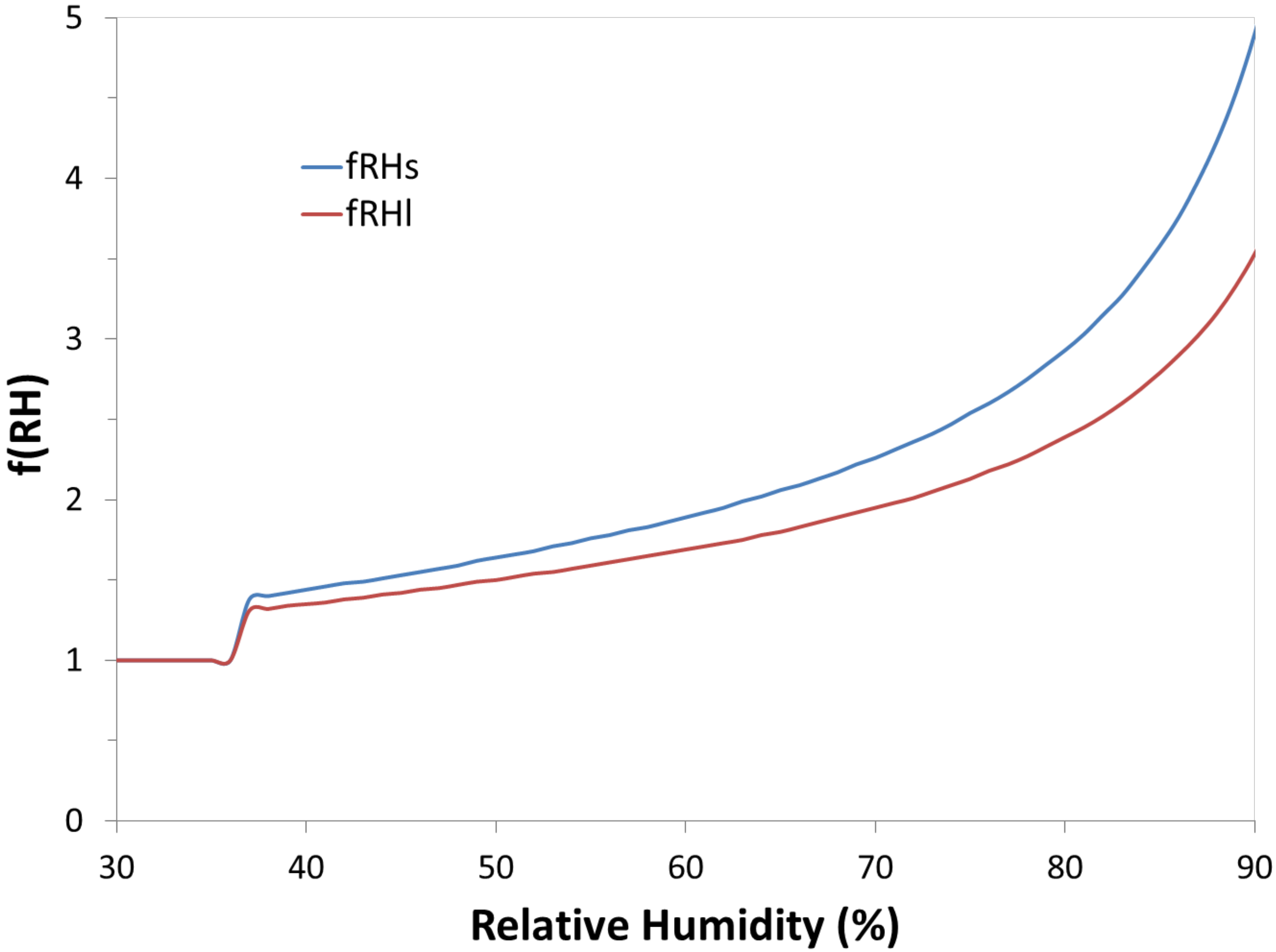
95th% size split denominator is too low for sulfate in some cases, leading to too much mass in large mode, and scattering that is higher than actually observed.

Outliers: Recon. bsp – Meas. bsp **< 50 Mm^{-1}**
i.e. when we're under-predicting (absolute)



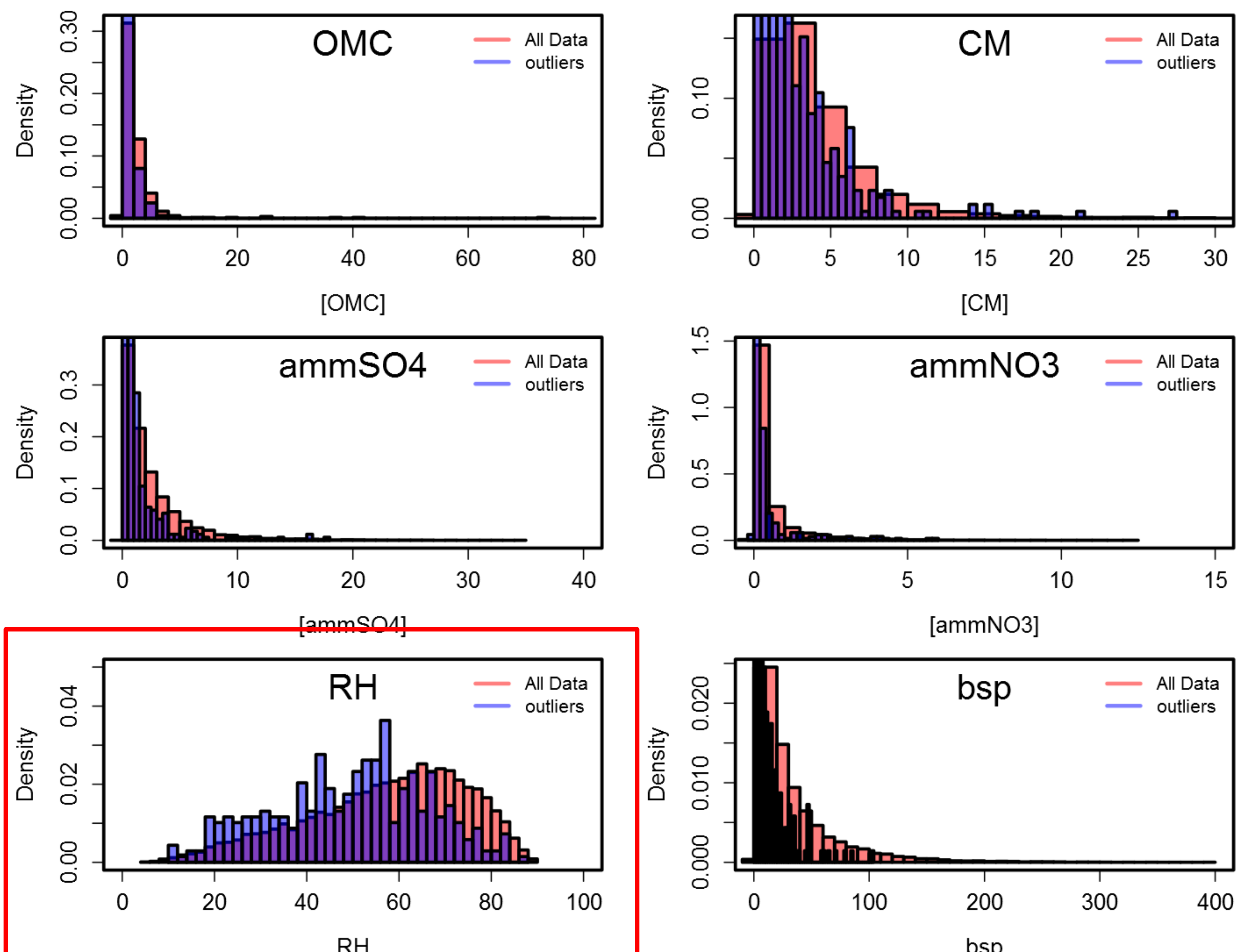
Large under-predictions at high RH.

Outliers: Recon. bsp – Meas. bsp < 50 Mm^{-1} i.e. when we're under-predicting (absolute)



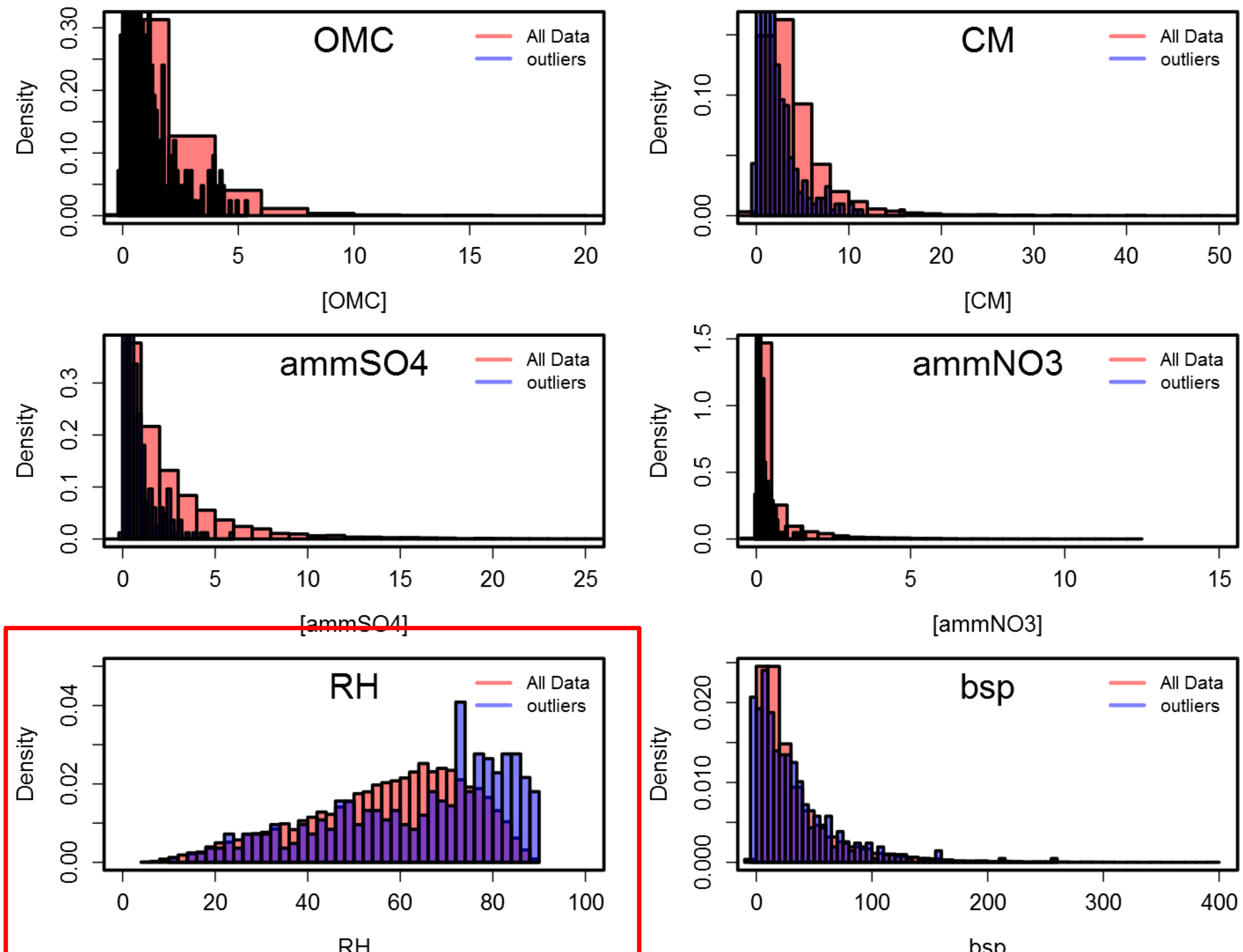
Large under-predictions at high RH. fRH is significantly lower in large mode.

Outliers: When over-predicting (relative) by > factor of 2



Over-predictions at low RH. Little role for fRH factor. Does this suggest we've moved too much into the large mode?

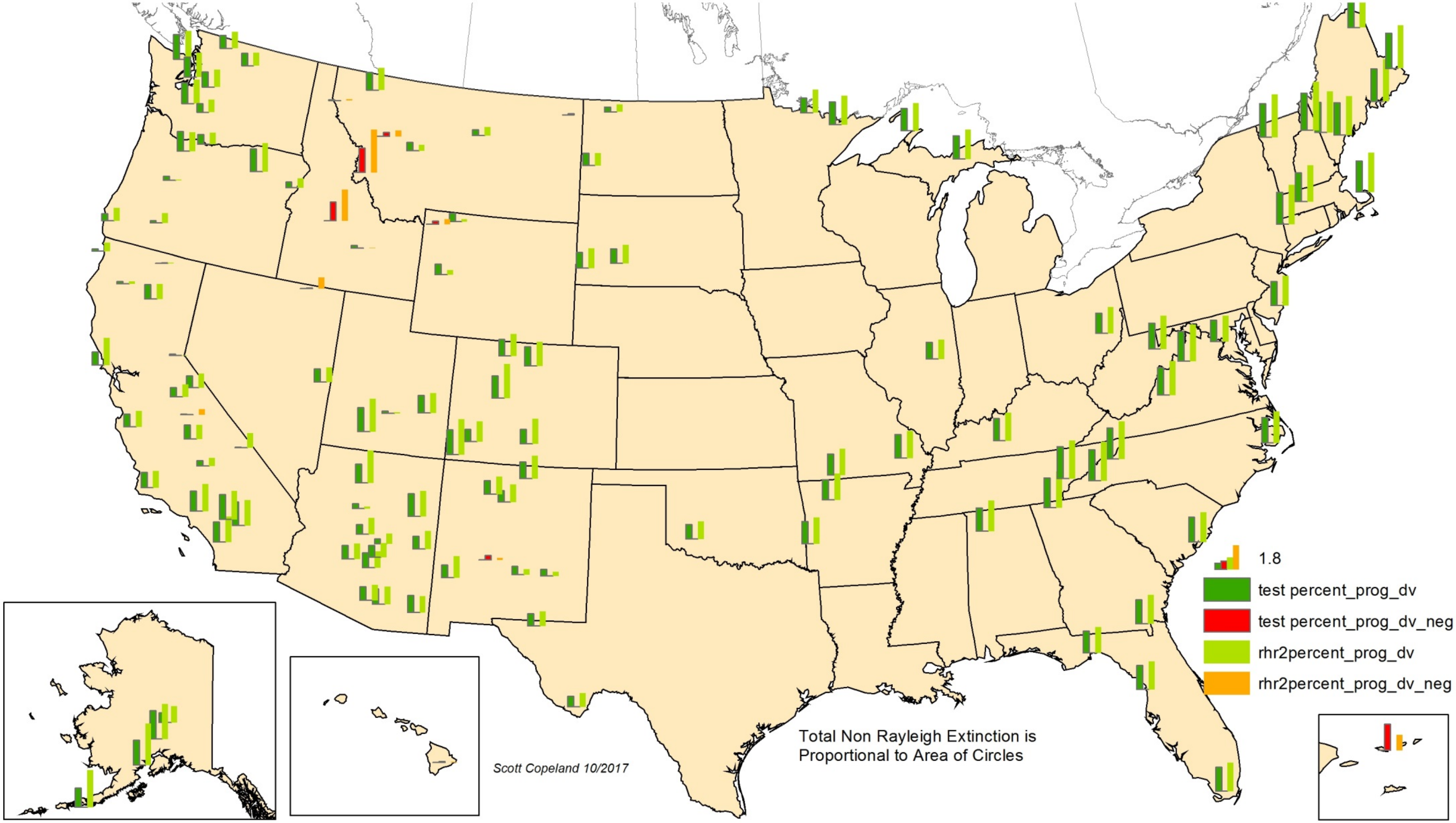
Outliers: When under-predicting (relative) by > factor of 2



Under-predictions at high RH

IMPROVE Data - 95% sm/lg and ROC compared to RHR2

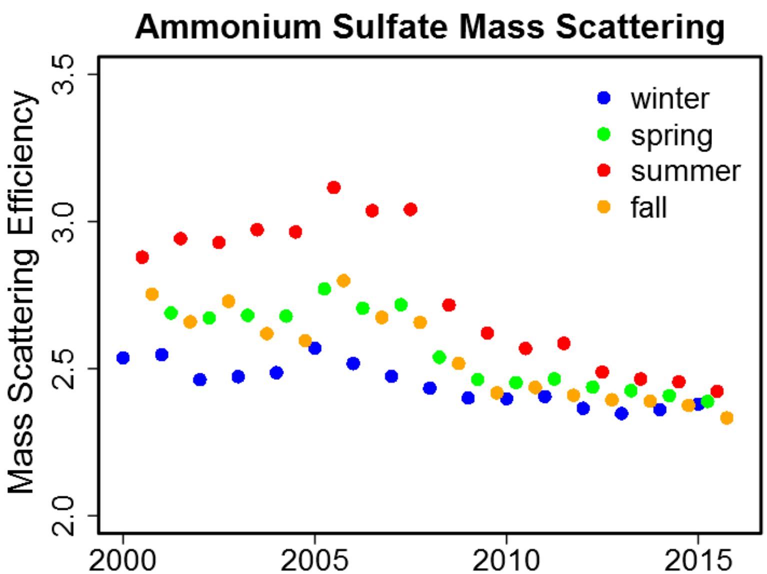
DRAFT % of Glide Slope Change in Haziest Days dv

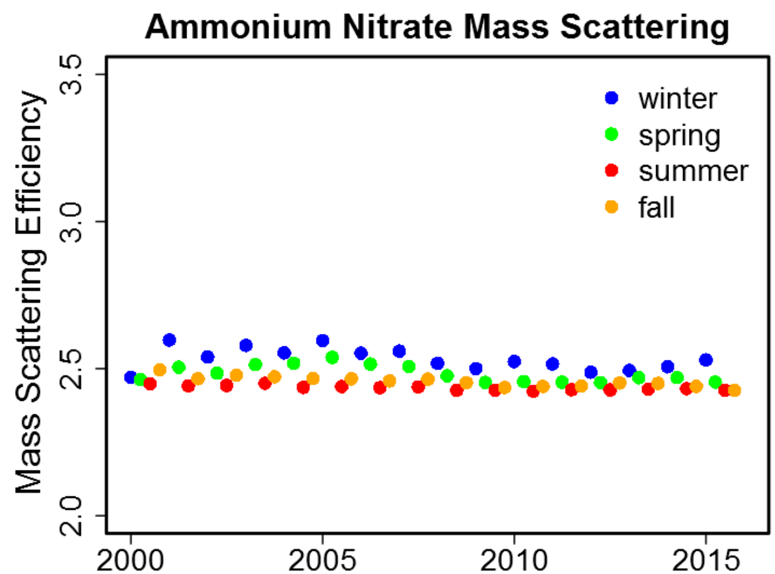


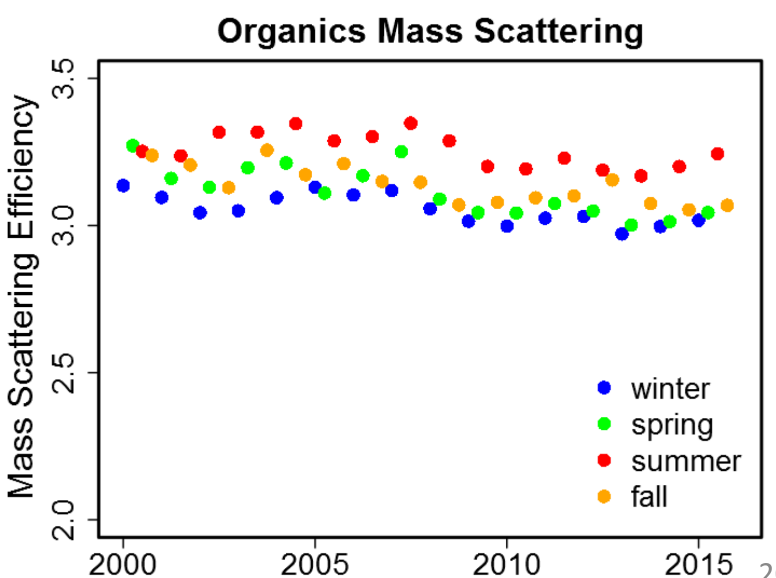
Path Forward?

- The annual 95% mass offers a potential approach to scaling the split component denominator.
- Optimizing this value is dependent on resolving issues with mass closure.
- This approach ignores any other issues with IMPROVE II, and may be compensating for other potential issues with the equation.
- Would like to first validate that size distributions have remained roughly the same at parks.

Size Split may be moving too much aerosol to small mode (lower mass extinction coefficient)







Ex. Sulfate Scattering Efficiency

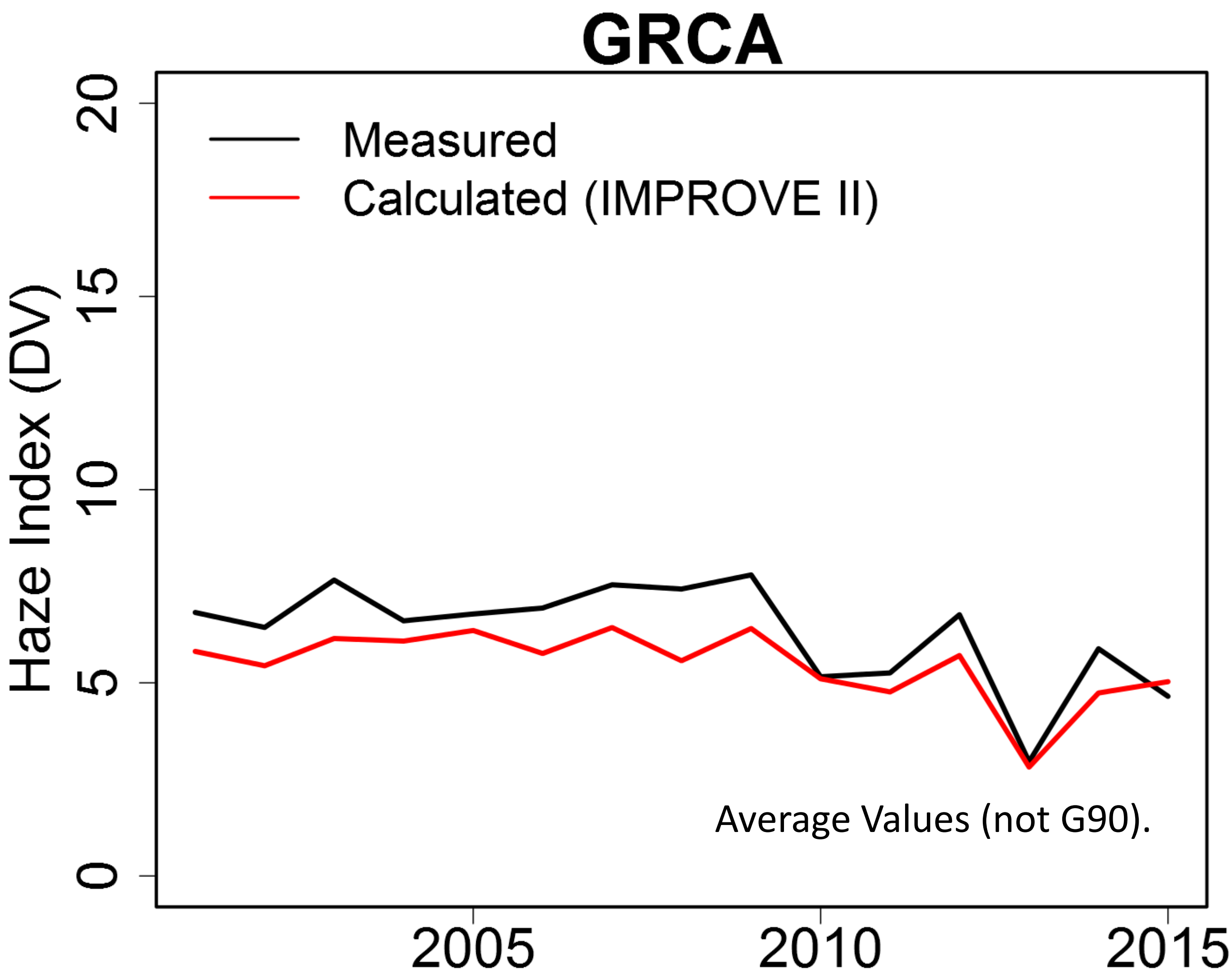
$$\mathbf{2.2 \times f_s(RH) \times [Small\ Sulfate]}$$
$$\mathbf{+4.8 \times f_L(RH) \times [Large\ Sulfate]}$$

Mass Scattering Efficiency =

fraction small × small mode scattering efficiency +

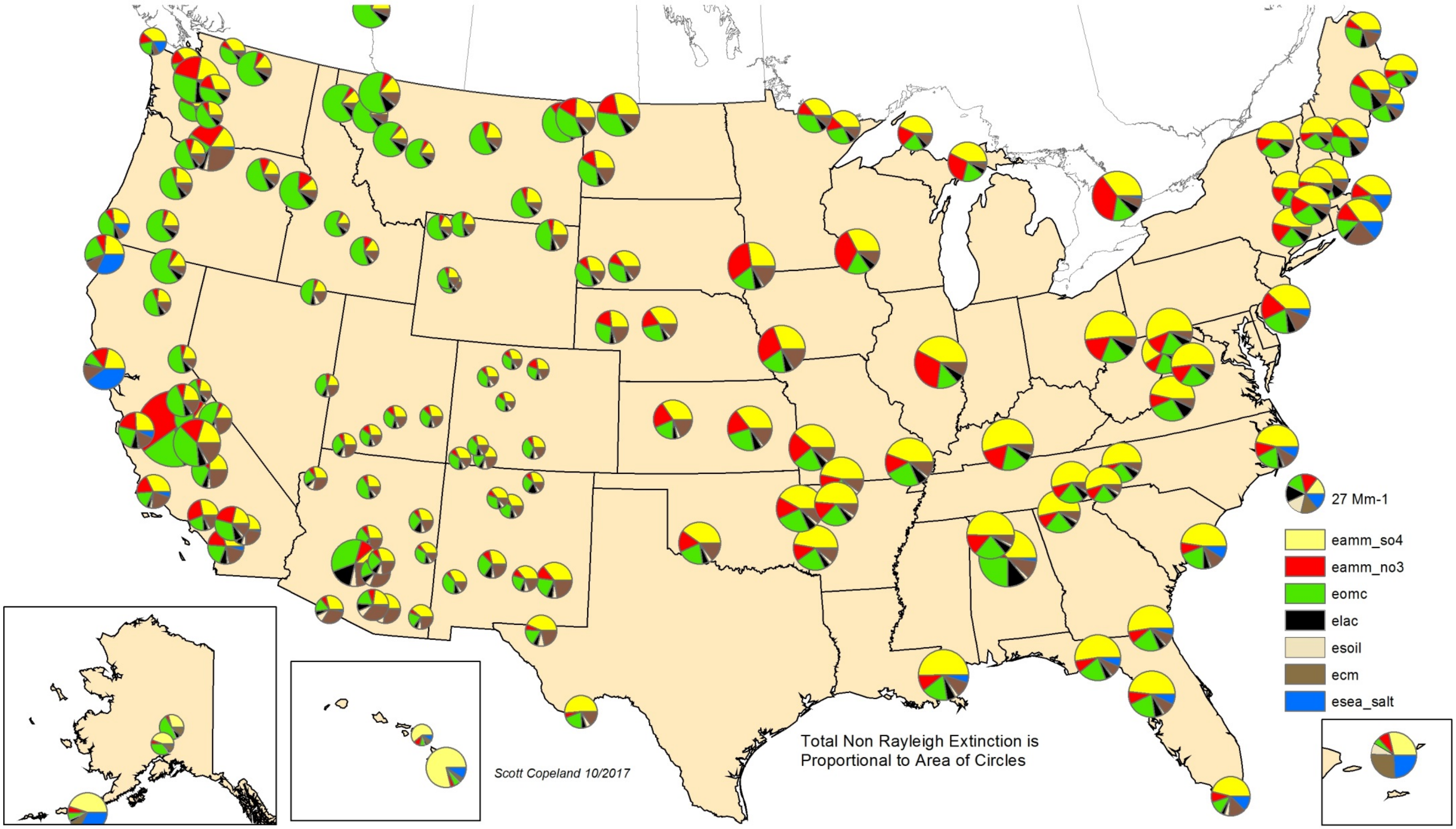
fraction large × large mode scattering efficiency

Potential Implications



At parks with low conc. in early years, baseline period may have been under-estimated.

2016 Annual Mean Aerosol Extinction RHR2



2016 Annual Mean Aerosol Extinction sm/lg and ROC correction

DRAFT