Climate implications of the unique optical properties of wildfire smoke.

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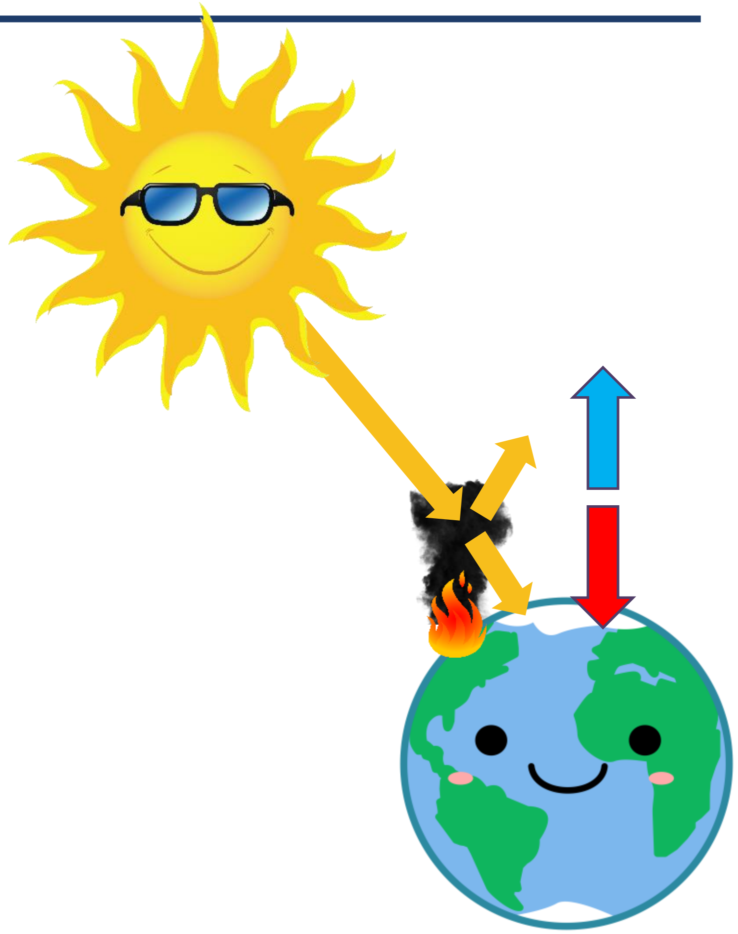
Key takeaways

- Smoke from wildfires has important effects on the global radiative balance.
- How the smoke affects the radiative balance depends on the optical properties of the particles, especially single scattering albedo and asymmetry parameter.
- Measurements of smoke asymmetry parameters show that more light is scattered back into space than would be predicted by models.
- We estimate the effect of this backscatter on the direct radiative forcing is to underestimate the cooling caused by fresh smoke by 20%.



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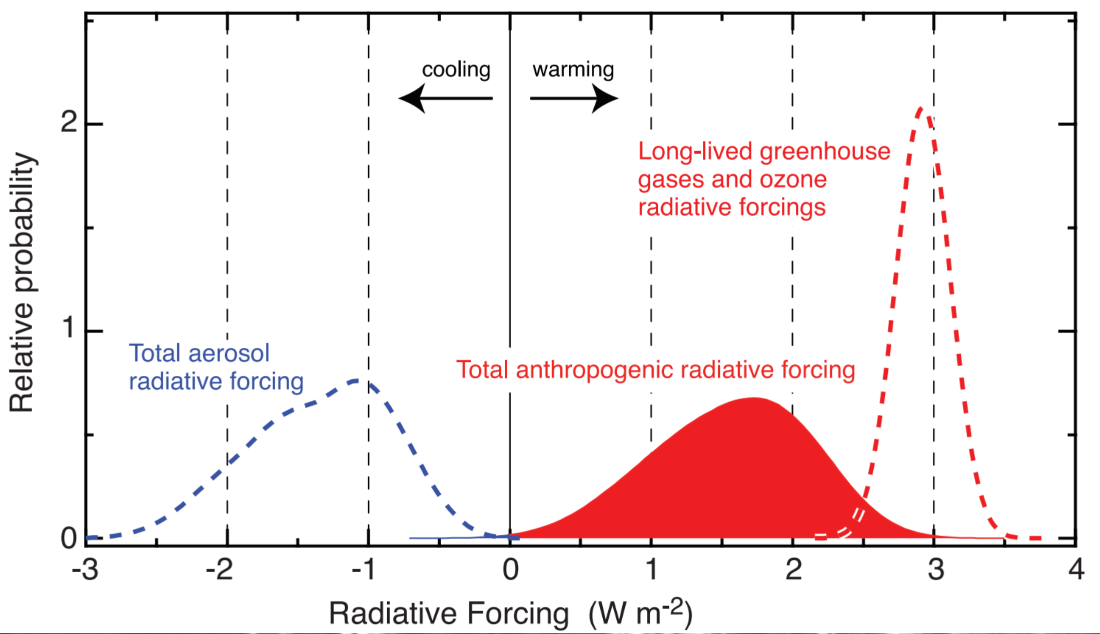
Background

- Uncertainty associated with aerosols is a large source of uncertainty for the degree of climate change, overall.
- Wildfires are increasing in frequency and intensity as a result of climate change.
- Smoke from wildfires can change the global radiative budget two ways:
  - Indirectly by changing cloud properties.
  - Directly by absorbing sunlight or scattering sunlight back into space.
- **This work looks at the direct radiative effect of smoke based on measurements of smoke optical properties during FIREX-AQ.**



IPCC 4, WG1 Fig 2.20

SSA and g are important optical properties of smoke

- **Single scattering albedo (SSA or ω)** describes the probability that a photon that interacts with a smoke particle will be absorbed (warming effect) or scattered (cooling effect, maybe.)

  $\omega = 1$ → All scattered and none absorbed

  $\omega = 0.5$ → Half scattered and half absorbed

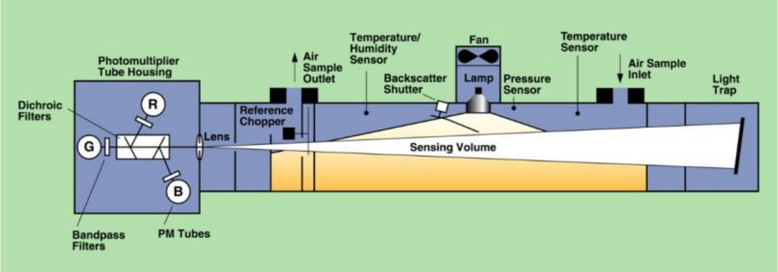
- The **scattering phase function (P_{11})** describes the intensity of scattered light as a function of scattering angle.
  - Computationally expensive for climate models
- Instead, climate models use **the asymmetry factor (g),** describes probability that if a photon is scattered, will it go back into space (cooling effect) or continue towards earth (no effect)?

  $g = 1$ → all scattered light continues forward (towards earth, depending on solar angle)

  $g = 0$ → scattered light is evenly split between forward and backward

  $g = -1$ → all scattered light goes backwards

Backscatter fraction vs asymmetry parameter



- **Backscatter fraction, b,** is routinely measured by TSI Integrating Nephelometers. Simply the ratio of hemispheric backscattered light to total scattered light.

$$b = \frac{\sigma_{bsp}}{\sigma_{sp}}$$

- **Asymmetry parameter, g,** is useful in climate models because it is more easily relatable to up-scatter fraction. But, it is rarely measured directly.
  - It can be calculated from Mie theory as a function of particle size, number, shape, and composition.
  - Or derived from the measured backscatter fraction, assuming a Henyey-Greenstein phase function.

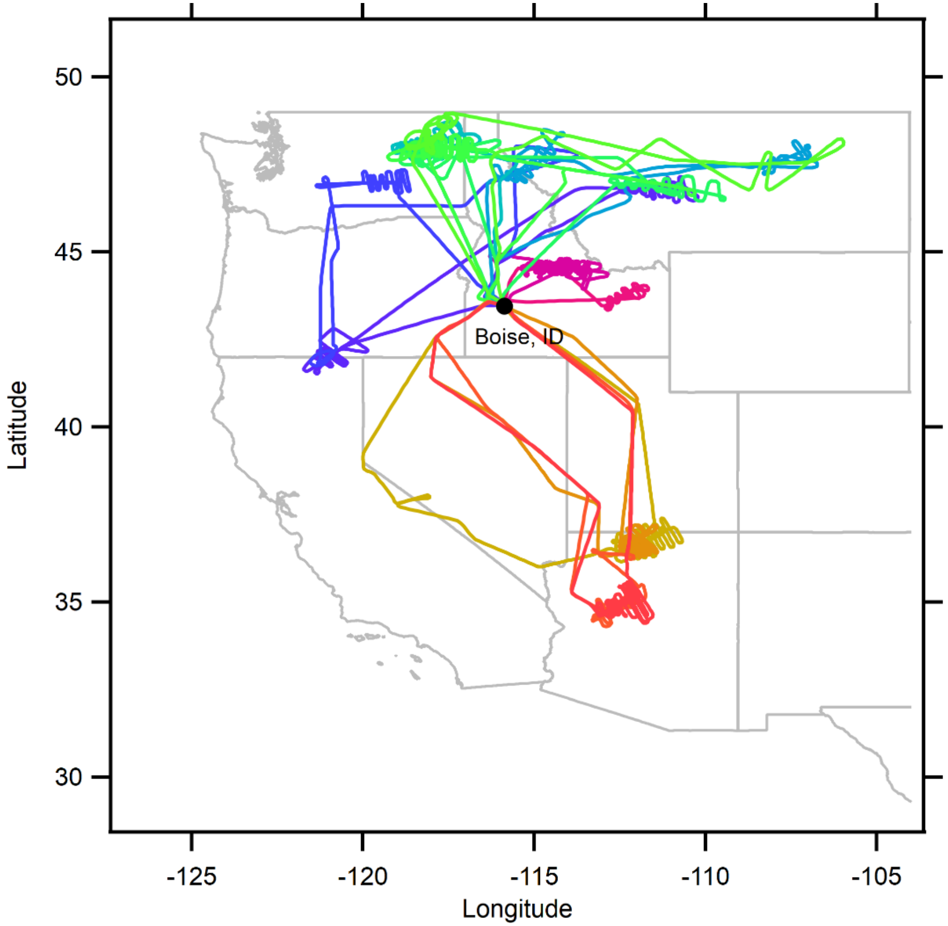
$$g = \frac{1}{2} \int_0^\pi \cos\theta \, P(\theta) \, \sin\theta \, d\theta$$

Heintzenberg, J. and R. J. Charlson (1996). Journal of Atmospheric and Oceanic Technology

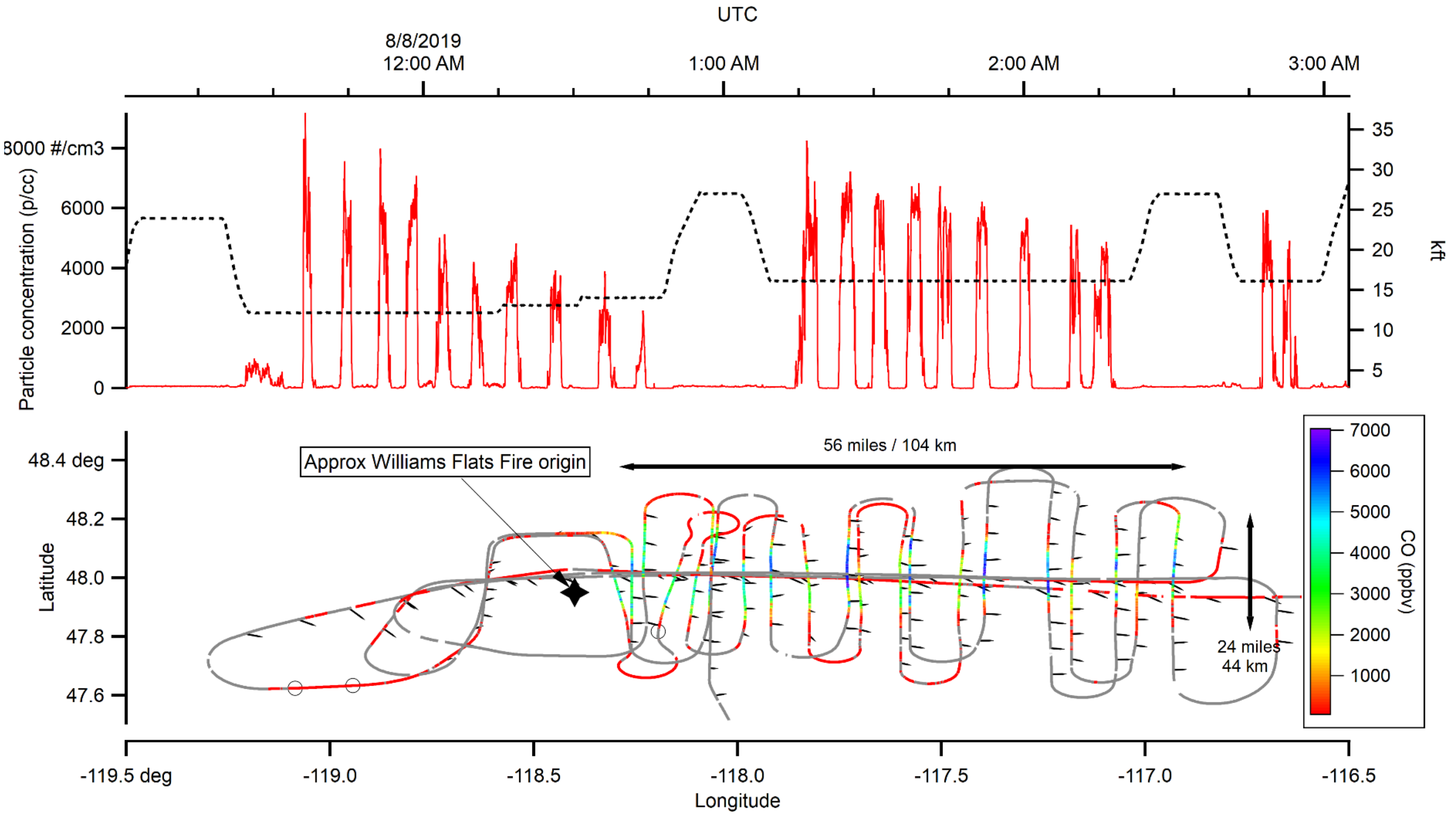
Methods and Measurements

Measurements of smoke optical properties

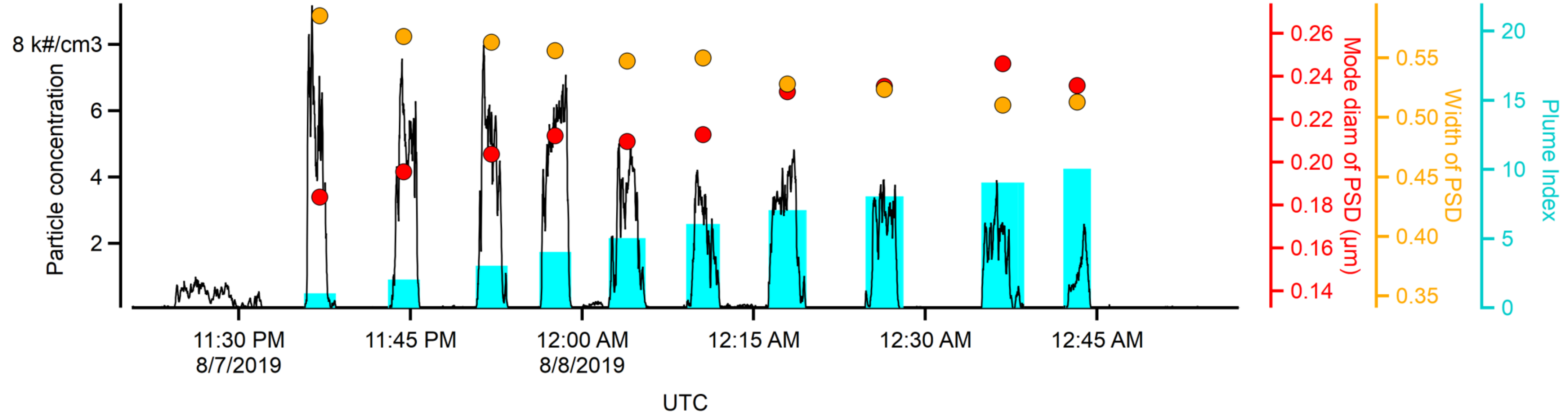
- FIREX-AQ was an airborne mission to study wildfire smoke in the western USA.
- Measured g and SSA at 405 nm and 660 nm.
- Collaborators measured particle size distribution and backscatter fraction.



Aircraft transected smoke plumes downwind of fire.



Each plume transect has a unique particle size distribution (PSD).



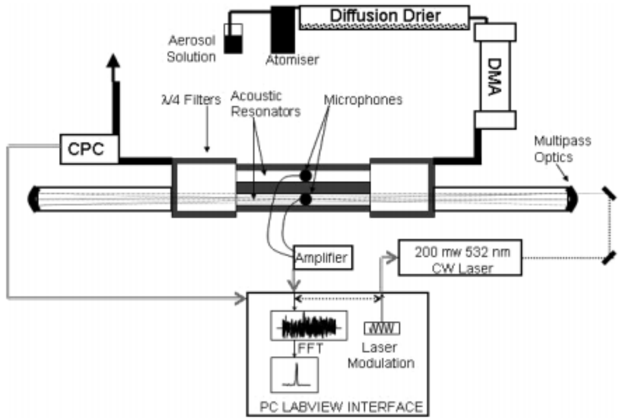
Measurements of single scatter albedo (SSA)

Single scattering albedo (ω) is the ratio of scattering to scattering plus absorption.

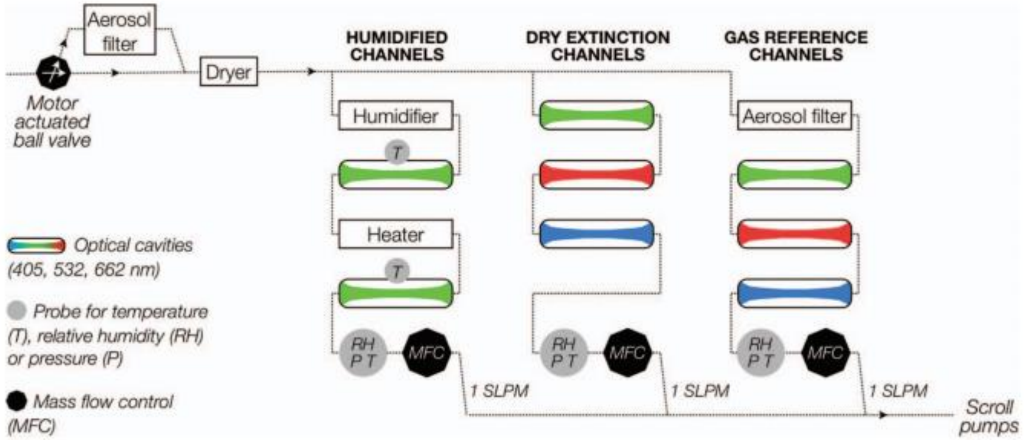
$$\omega = \frac{\sigma_{scat}}{\sigma_{scat} + \sigma_{abs}} = \frac{\sigma_{scat}}{\sigma_{extinction}}$$

Absorption is measured via photoacoutic spectroscopy (PAS.)

Extinction is measured via cavity ringdown spectroscopy (CRDS.)



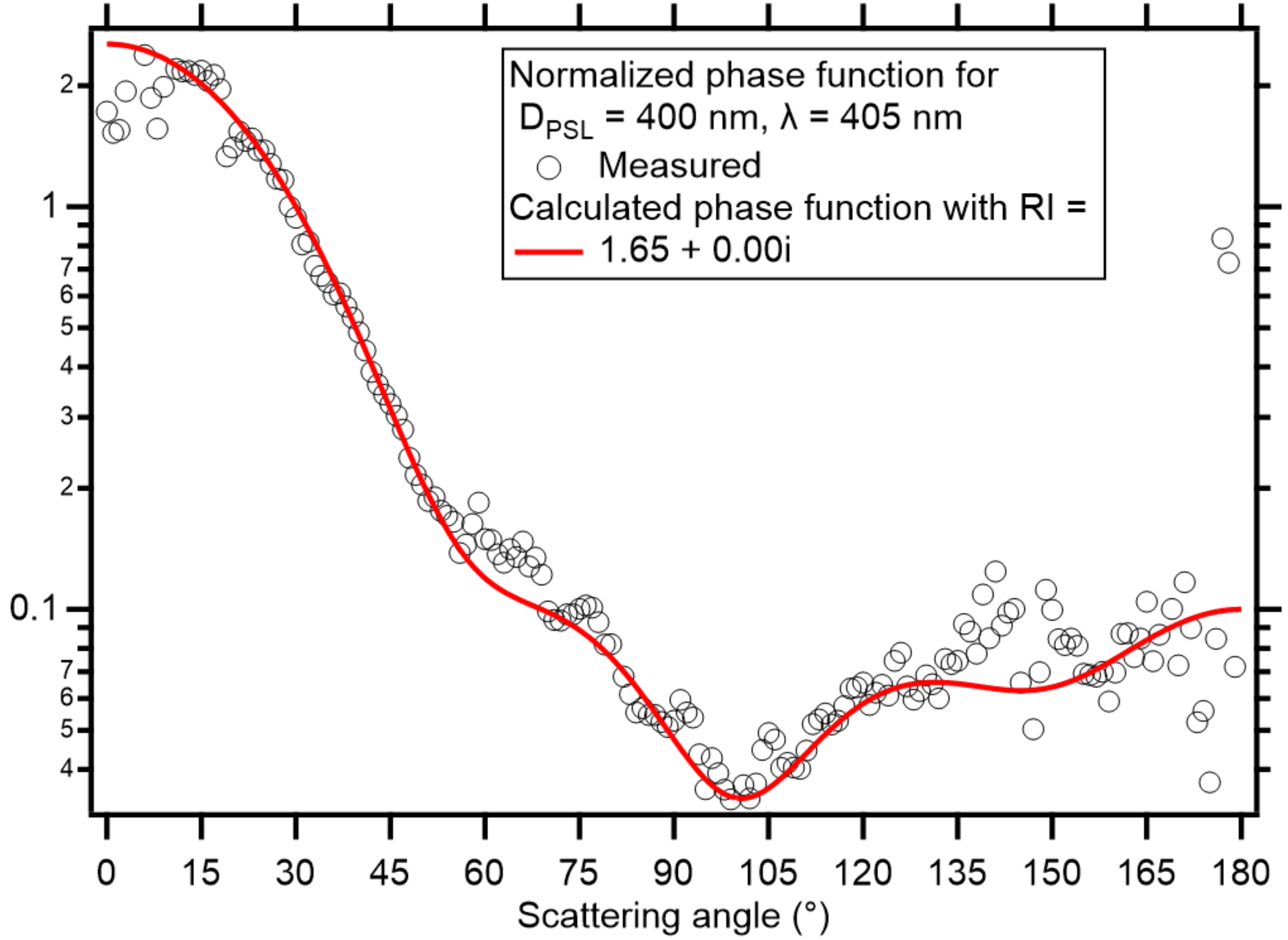
Lack et al. (2006) Aerosol Science and Technology



Langridge et al. (2011) Aerosol Science and Technology

Measurements of phase function (P_{11})

- Laser imaging nephelometer measures phase function for aerosol scattering.
- Markers show the normalized scattering intensity as a function of scattering angle.
- The red line shows the Mie theory predicted phase function.
- **We calculate the asymmetry parameter, g, directly from this phase functions.**



Calculating g for smoke using Mie theory

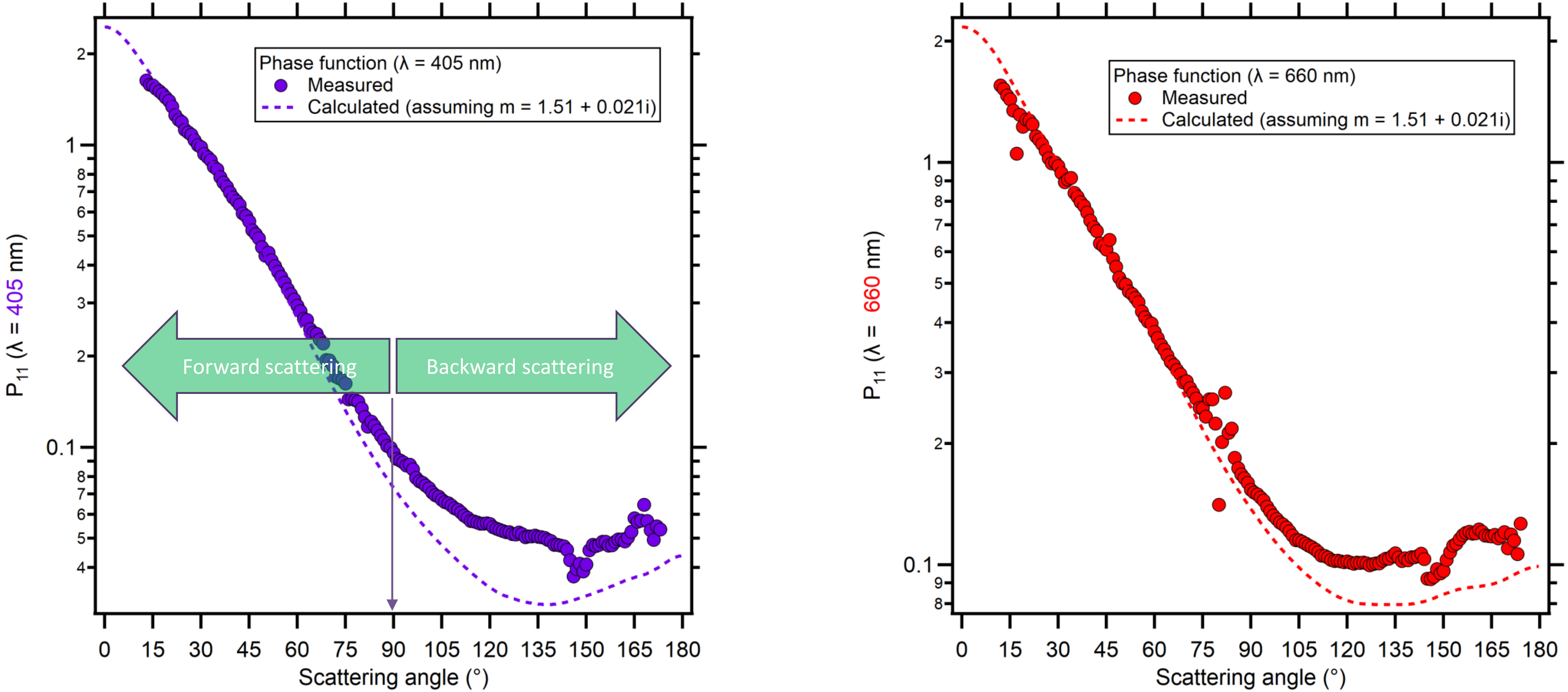
- Requirements for calculating SSA and phase function using Mie theory:
  - Number and size of particles – from optical particle size (Laser Aerosol Spectrometer)
  - Shape – Composition measurements suggest black carbon, the most likely cause of asphericity in soot, is less than 10% by mass during FIREX-AQ.
  - Refractive index – based on AERONET measurements, the RI commonly used in chemical transport models with radiative transfer modules.
    - Although a large span of RI are observed, we only use one in this model.

Biomass burning	African savanna, Zambia (1995–2000)
Number of measurements (total)	2000
Number of measurements (for ω_0, n, k)	700 (Aug–Nov)
Range of optical thickness; $\langle\tau\rangle$	$0.1 \leq \tau(440) \leq 1.5$; $\langle\tau(440)\rangle = 0.38$
Range of Ångström parameter	$1.4 \leq \alpha \leq 2.2$
$\langle g\rangle$ (440/670/870/1020)	0.64/0.53/0.48/0.47 $\pm$ 0.06
n; k	1.51 $\pm$ 0.01; 0.021 $\pm$ 0.004
ω_0(440/670/870/1020)	0.88/0.84/0.80/0.78 $\pm$ 0.015
r_{Vf} (μm); σ_f	$0.12 + 0.025\tau(440) \pm 0.01$; 0.40 ± 0.01
r_{Vc} (μm); σ_c	$3.22 + 0.71\tau(440) \pm 0.43$; 0.73 ± 0.03
C_{Vf} ($\mu m^3/\mu m^2$)	$0.12\ \tau(440) \pm 0.04$
C_{Vc} ($\mu m^3/\mu m^2$)	$0.09\ \tau(440) \pm 0.02$

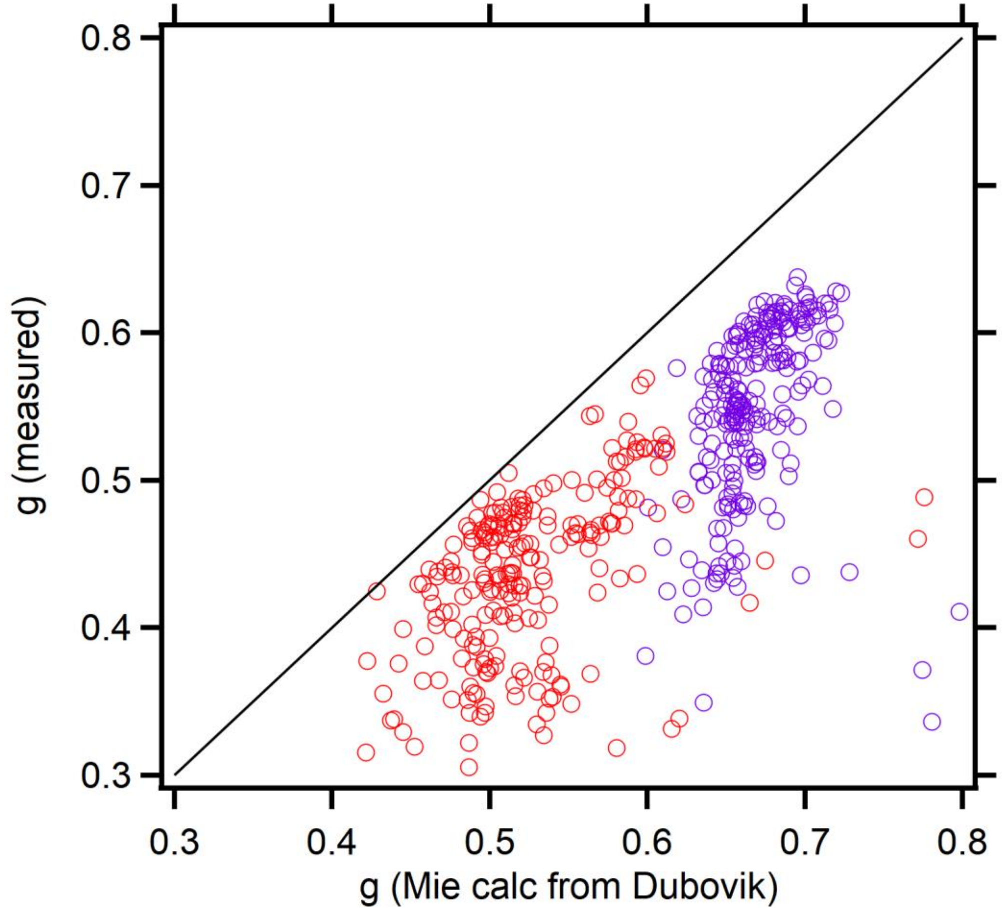
Dubovik, O., et al. (2002). Journal of the Atmospheric Sciences

Results

Case study: Measured phase functions show more back scattering than predicted by Mie.

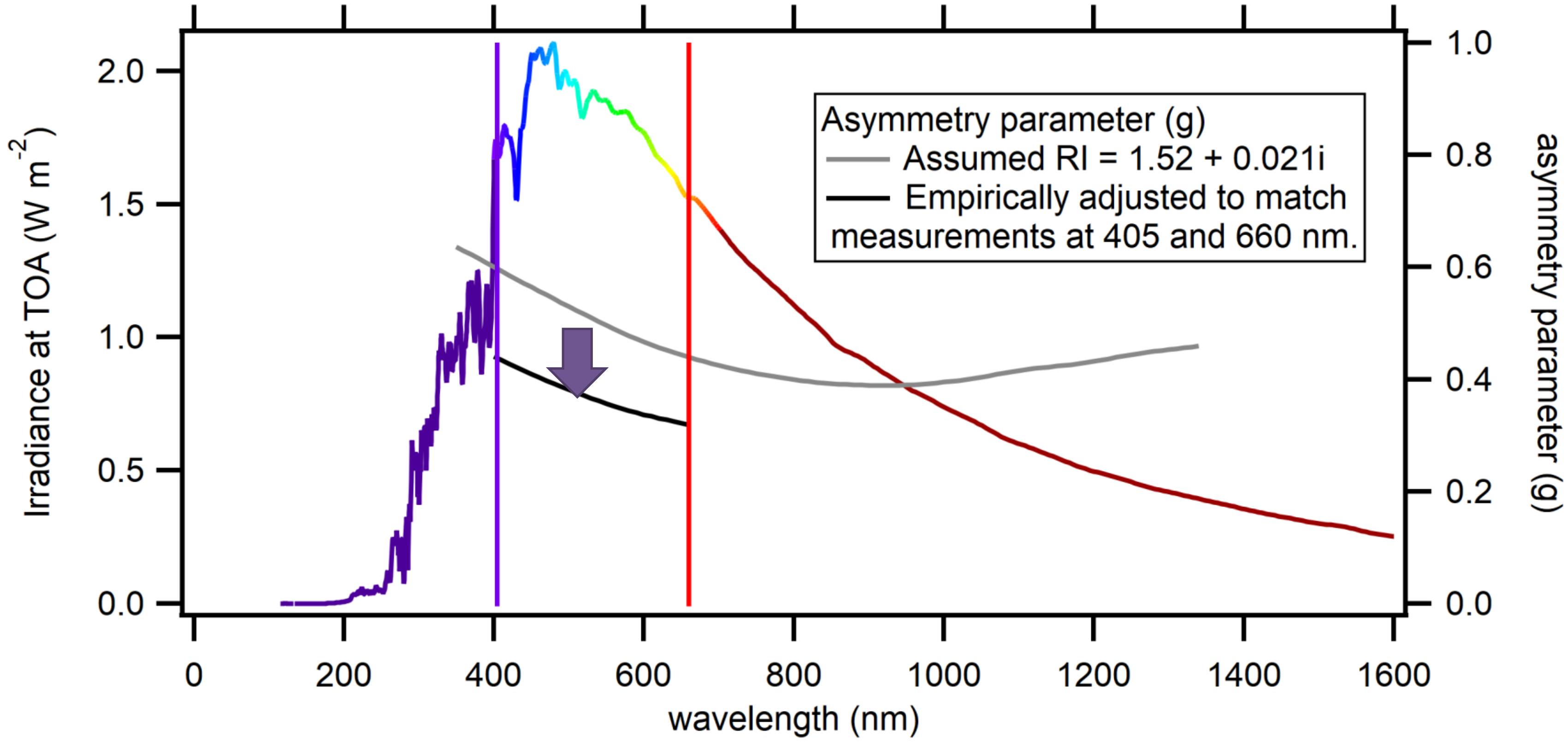


Measurements of g are lower than predicted using Mie theory and literature refractive indices.



Each data point is a plume transect from the FIREX-AQ western US campaign.

We use an empirical relationship to force Mie calculations to match measurements.



Repeat Mie calculation for each plume transect and visible wavelength.

From SSA and g, we can estimate how smoke will effect the global radiative balance.

- A good approximation of how smoke will change the radiative balance of the planet is the aerosol forcing efficiency, W*m^{-2} per optical thickness.

Defines radiation available to interact with aerosol

Aerosol scattering

Aerosol absorption

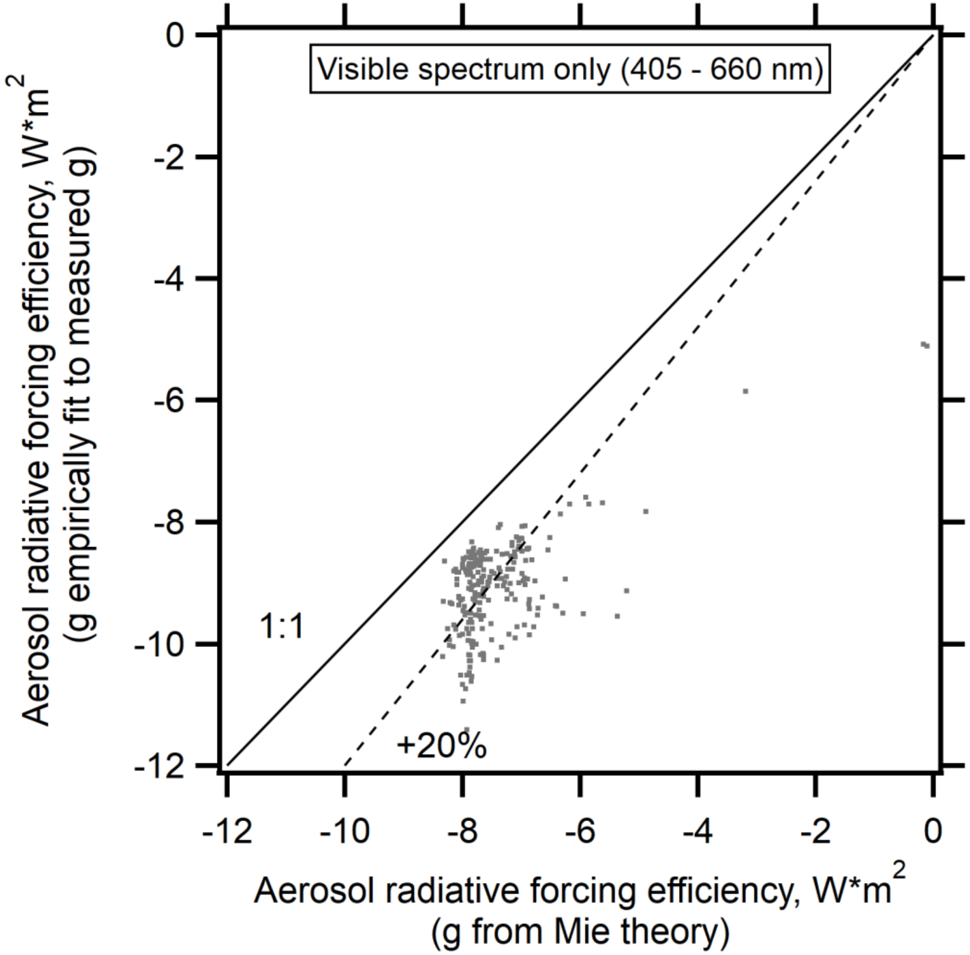
$$\frac{\Delta F_{aer}}{\tau} = \boxed{-\frac{S_0}{2} T_{atm}^2 (1 - A_{cld})} \boxed{\left[\bar{\beta}\,\omega (1 - R_{surf})^2\right.} \boxed{\left. - 2(1-\omega) R_{surf}\right]}$$

SSA = from Mie theory
How much is absorbed vs. scattered?

β = average upscattering fraction, empirically related to g
If light is scattered, does it go back into space?

Chýlek, P.; Wong, J. (1995) Geophys. Res. Lett.

Calculated cooling by smoke is 20% more when calculating direct radiative forcing efficiency using a measured asymmetry parameter.



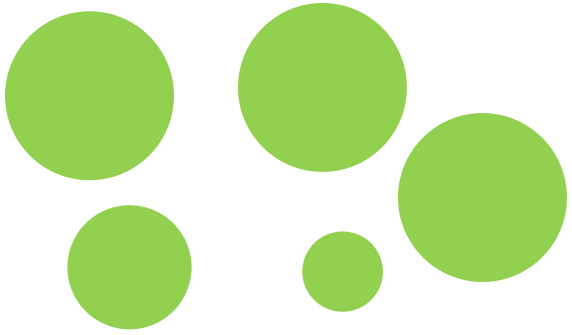
Aerosol radiative forcing efficiency is the direct radiative forcing normalized by aerosol optical thickness which is unit-less.

$$\frac{\Delta F_{aer}}{\tau}$$

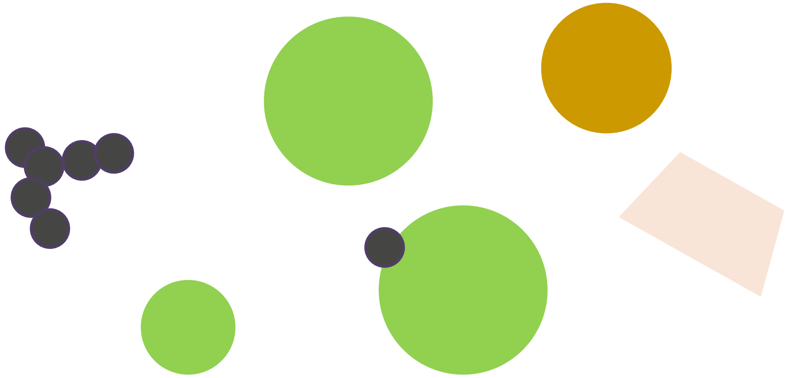
Future work

Possible explanations for why Mie theory doesn't predict phase function.

- All smoke particles have the same composition BUT:
  - Using the wrong refractive index
    - Likely, but unrealistic RI required to match measured g
  - Particles are non-spherical
    - Unlikely, black carbon (cause of asphericity in most soot) was on average less than 1% of aerosol by mass during campaign.

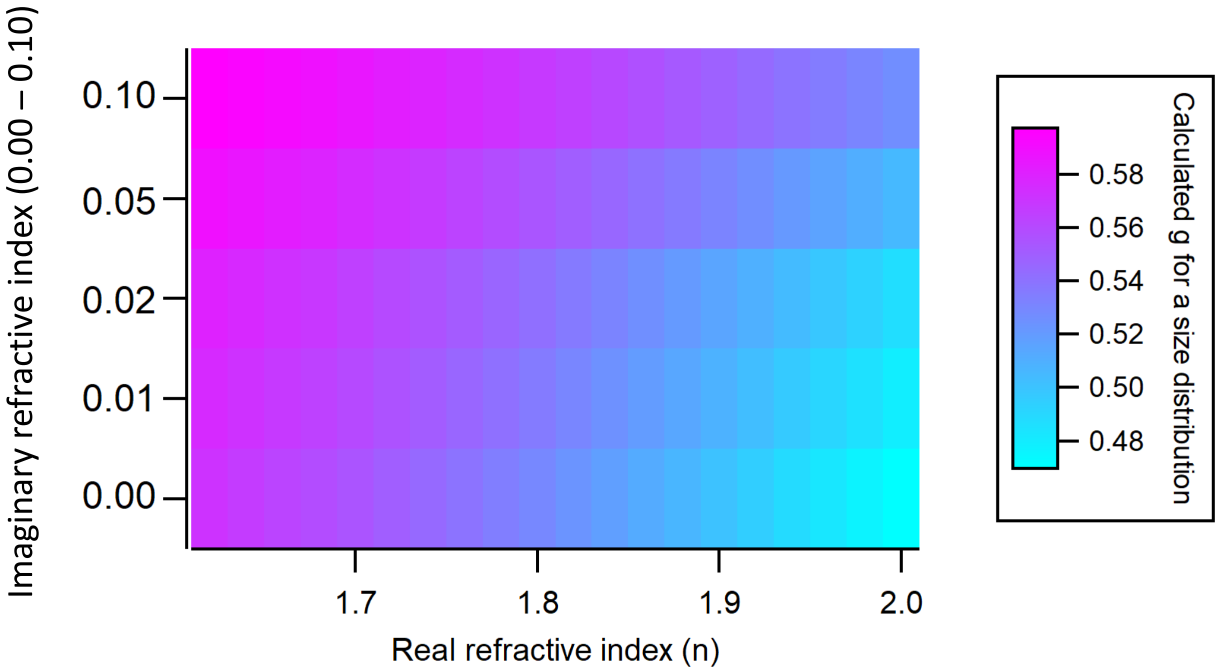


- Smoke particles have varying composition AND:
  - Using the wrong refractive indices
  - Some particles have complex morphology
    - Salt inclusions
    - Core-shell
    - Some "pure" black carbon and some pure organics

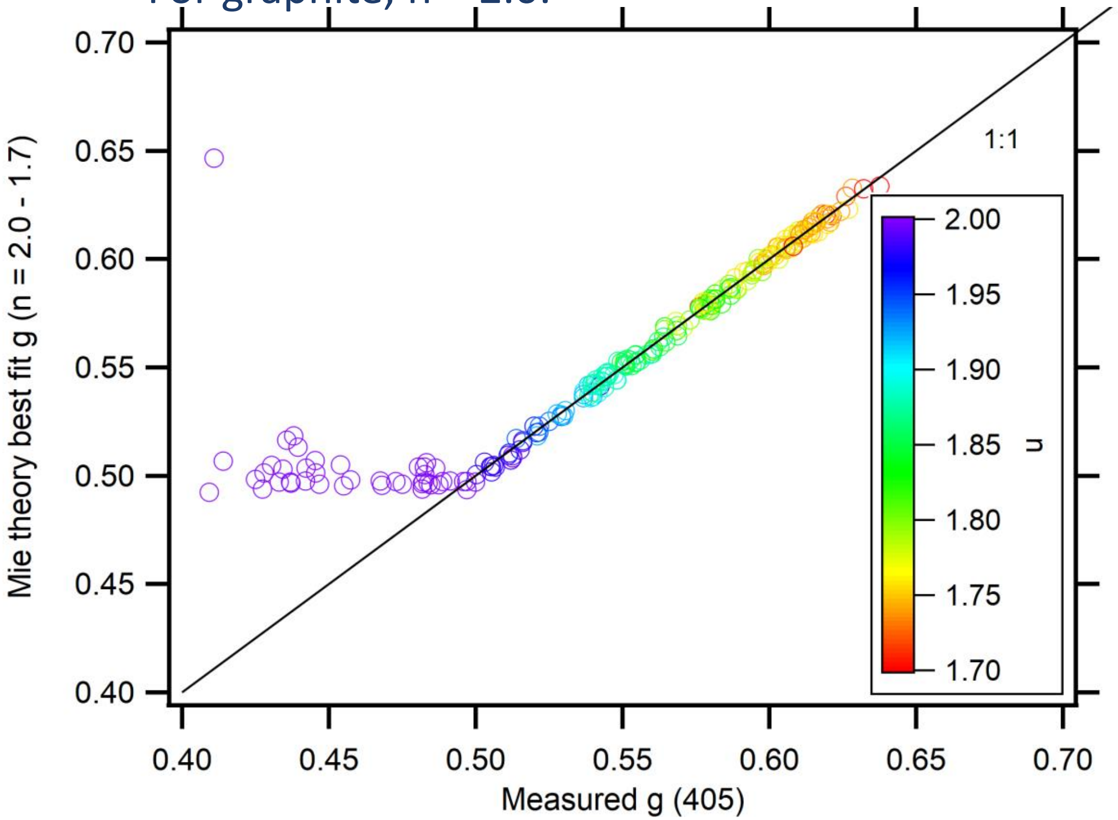


Changes in refractive index cannot reasonably explain observed asymmetry parameters.

For each size distribution, search a matrix of m = n + k*i to find the best fit for g.

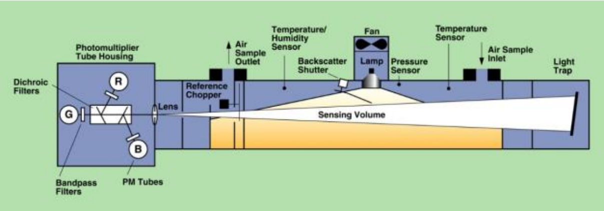


Measured g is so low that it forces n to unrealistically high values.
For ambient organic aerosols, n = ~1.51
For graphite, n = 2.0.



We measured significant deviation from the commonly applied Henyey-Greenstein approximation for relating backscatter fraction to the asymmetry parameter.

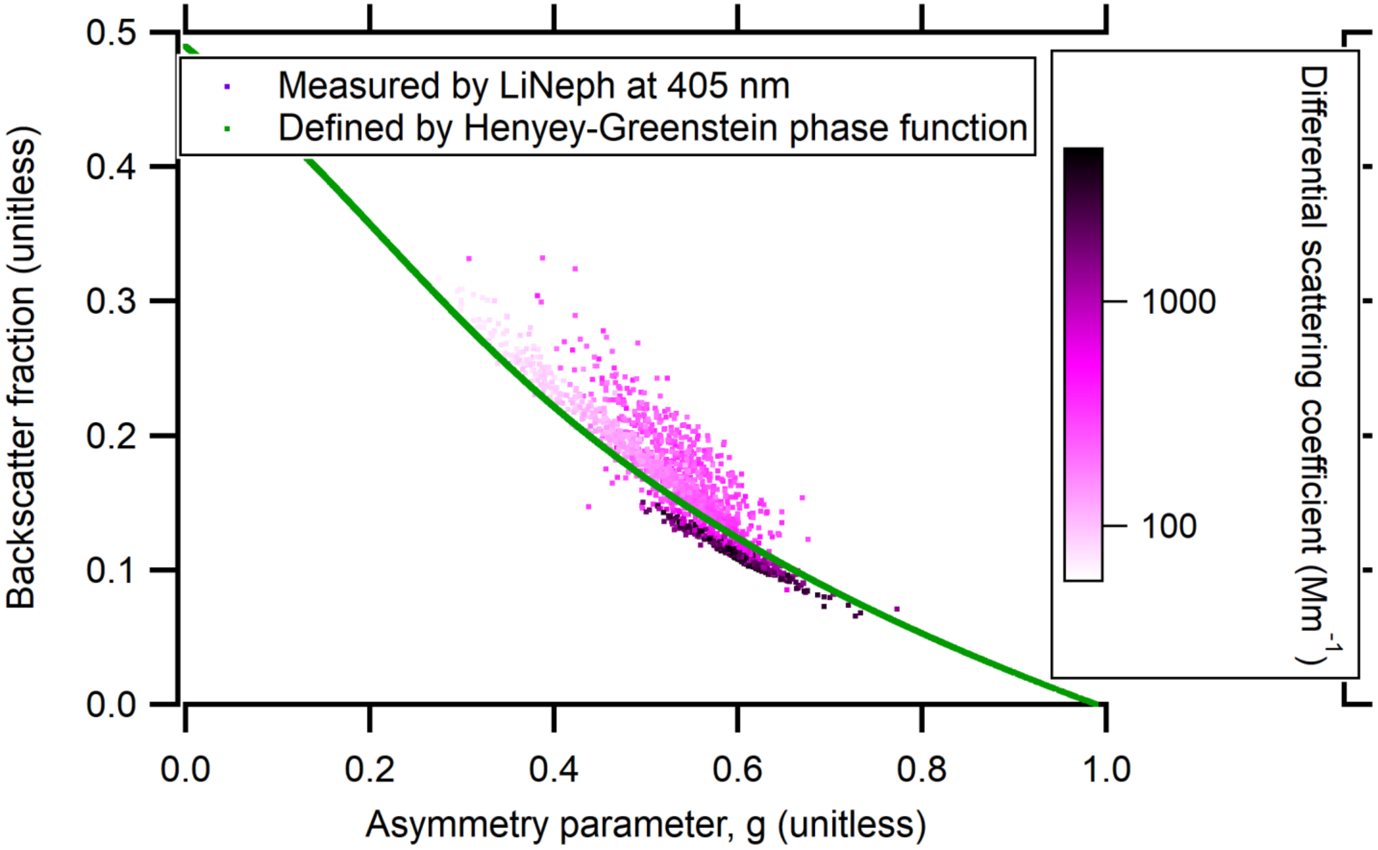
The most common ambient measurement of directional scattering is the **backscatter fraction**, as made by the TSI Integrating Nephelometer.



However**, the asymmetry parameter** is the more useful quantity for radiative transfer programs.

With measurements of the whole phase function, we can measure both quantities directly, and evaluate the accuracy of the approximation.

At the heart of the smoke plumes (darkest points), the approximation leads to a derived asymmetry parameter ~7% larger than the measured value.



This data shows individual measurements from a single research flight.

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