Estimating the direct radiative forcing of wildfire smoke using aerosol optical properties measured during FIREX-AQ.

Adam T. Ahern[1,2], Nicholas L. Wagner[1,2], Charles A. Brock[1], Ming Lyu[3], Richard H. Moore[4], Elizabeth B. Wiggins[4], Edward Winstead[4], Claire E. Robinson[4], Daniel M. Murphy[1]

[1] NOAA Chemical Sciences Laboratory (CSL), 325 Broadway, Boulder, Colorado 80305
[2] Cooperative Institute for Research in Environmental Sciences (CIRES), University of Colorado, Boulder, Colorado 80309
[3] University of Alberta, Dept. Of Chemistry, Edmonton, Canada T6G 2R3
[4] NASA Langley Research Center, Hampton, Virginia 23666

NOAA CHEMICAL SCIENCES LABORATORY

University of Colorado Boulder

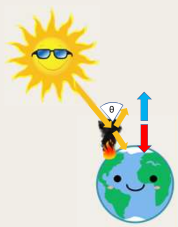
CIRES

Before we start, here's what you need to know:

- Global warming is caused by the imbalance of energy being absorbed by and emitted from Earth.
- Greenhouse gases contribute to warming by causing more energy to be absorbed by the atmosphere.
- Particles in the atmosphere, like smoke, can reflect energy back into space before it can be absorbed
  - This is called the direct radiative effect, and it can mask global warming caused by greenhouse gases.
- The contribution of smoke particles to the direct radiative effect is uncertain in part because of their complicated optical properties.
- In this work, we measured what direction smoke particles scattered light and compared it to what direction is typically assumed by models. Then we calculated what effect this would have on the direct radiative effect.

When sunlight hits particles in the atmosphere, the light scatters in many directions.
If it scatters upwards, away from Earth, this can cause global cooling.
If it scatters downwards, then there is no cooling effect.
Climate models use the asymmetry parameter, g, to describe which direction scattered light will go.

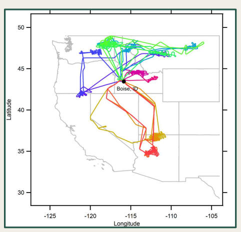
$$g = \frac{1}{2}\int_0^{\pi} \cos\theta\, P(\theta)\, \sin\theta\, d\theta$$

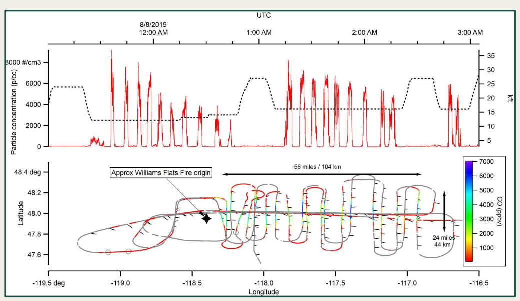


Where θ is the scattering angle and P(θ) is the probability light will scatter in that direction.
g = 1 → all scattered light continues forward (towards earth, depending on solar angle)
g = 0 → scattered light is evenly split between forward and backward

Aerosol Microphysical and Optical Measurements during FIREX-AQ

- FIREX-AQ was an airborne mission to study wildfire smoke in the western USA in the summer of 2019.
- The NASA DC-8 Airborne Laboratory (top right) was equipped with instruments to measure aerosol optical and microphysical properties.
- 12 wildfires in the western US were sampled in a pseudo Lagrangian pattern (middle right show flight tracks.)
- Bottom-right plot shows the time series where the aircraft transects the plume very close to the fire, then travels down wind for subsequent transects, resulting sets of measurements with smoke roughly the same age.





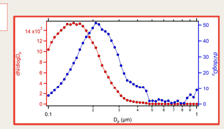
- NASA Langley Aerosol Research Group measured particle size distributions using the LAS (Moore et al., 2021) as well as total and hemispheric backscatter using a TSI Integrating Nephelometer.
- NOAA Chemical Sciences Lab measured aerosol optical properties using the NOAA Laser Imaging Nephelometer and Aerosol Optical Properties instrument suite, consisting of a cavity ringdown spectrometer (measuring extinction) and a photoacoustic (measuring absorption.)

Three ways to determine g:

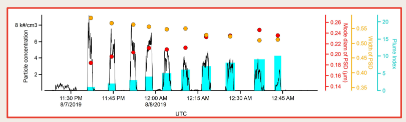
1) Calculate using **Mie theory** (requires assumptions about particle shape, particle size distribution, and refractive index.)
   - BUT smoke composition varies with source and time
2) Measure a similar value, e.g. **hemispheric backscatter**, *b*, and use an empirical relationship to derive g (Henyey and Greenstein, 1941.)
   - BUT this relationship has been shown to be inaccurate using Mie calculations for small particles, like smoke (Marshall et al., 1995).
3) Measure it directly using the **NOAA Laser Imaging Nephelometer**
   - Precise measurements with a relative standard deviation of 3%*, but only at two wavelengths.

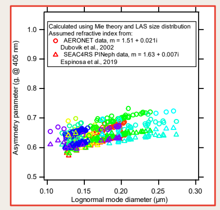
*Ahern et al., AMT 2022

1) Calculating the phase function from Mie theory



- An example of the size distributions measures is shown in the top right. The number size distribution is dominated by the fine mode, which we can fit with a lognormal function.
- The plot on the bottom shows the evolution of the size distribution as it travels downwind; the lognormal mode size grows and the distribution narrows with aging.
- The bottom right plot shows the g that is calculated from each distribution for multiple wildfires (colors). The circles show the result of using a refractive index from AERONET retrievals to estimate g, and the triangles show the results using a refractive index from previous in situ measurements of the phase function.

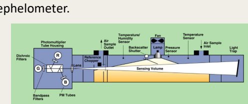




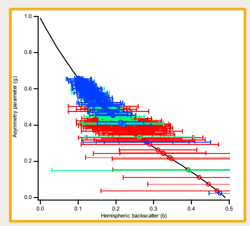
2) Inferring g from backscatter measurements

- Hemispheric backscatter (b) is the ratio of all backwards scattering light to total light, routinely measured globally by the TSI Integrating Nephelometer.

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

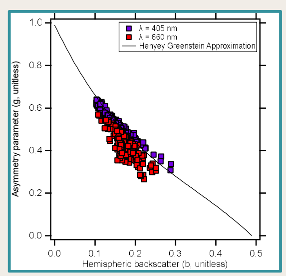


- Conversion from b to g is typically done using the Henyey Greenstein phase function, shown on the right. (Henyey and Greenstein, 1941)



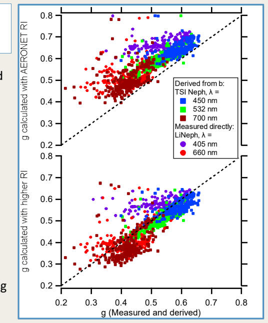
3) Measuring g directly with the LiNeph

- The NOAA Laser Imaging Nephelometer (LiNeph) directly measures the intensity and direction of scattered light by aerosol particles.
- From this measurement, we can directly calculate both the asymmetry parameter, g, and the hemispheric backscatter, b.
- The Henyey Greenstein approximation works well for scattering of 405 nm light, deviating on average by 1%.
- For 660 nm, the g predicted from the Henyey Greenstein approximation results in an average 14% overestimation compared to the measurements.
- **This is consistent with theoretical studies that show that for particle size distributions dominated by small particles, the Henyey-Greenstein phase function tends to overpredict g (Marshall et al., 1995)**



Measured g shows more backscatter than RI values from remote retrievals would predict

- We used Mie theory to calculate g at five wavelengths based on the size distribution measured by the optical particle counter.
- The top panel of figure shows the g calculated using the mean refractive index from AERONET retrievals, where the refractive index is found to be 1.51 + 0.021i for all wavelengths (Dubovik et al., 2002.)
- However, more recent in situ studies have found higher and wavelength dependent refractive indices (Espinosa et al., 2019, Womack et al., 2021.).
- Bottom panel shows g calculated with the higher refractive indices from Espinosa et al.
- This higher assumed refractive index also affects the interpretation of the size distribution measurements, shifting the volume mode of the size distribution ~20% smaller.



Estimating how smoke will effect the global radiative balance

The equation shown here calculates the **aerosol radiative forcing efficiency** ($\Delta F_{aer}/\tau$) caused by smoke. This is a scalar that can be used in a radiative transfer model to simulate the effect of wildfires on global and regional climate and atmospheric stability.

Defines radiation available to interact with aerosol | Aerosol scattering | Aerosol absorption

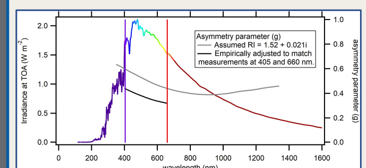
$$\frac{\Delta F_{aer}}{\tau} = -\frac{S_0}{2}T_{atm}^2\left(1-A_{cld}\right)\left[\bar{\beta}\,\omega\left(1-R_{surf}\right)^2 - 2\left(1-\omega\right)R_{surf}\right]$$

S_0 = Solar irradiance
T_{atm} = Transmission through gases
A_{cld} = Fraction of sky with clouds
R_{surf} = Surface albedo

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

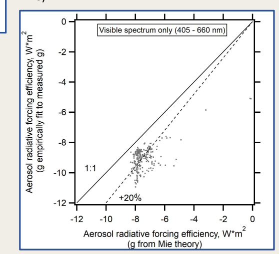
β = average upscattering fraction, empirically related to g
If light is scattered, does it go back into space?

From Chýlek, P.; Wong, J. (1995)



For this work, we will integrate across the solar spectrum, shown to the left. A reference case is evaluated using Mie theory and a commonly assumed refractive index to calculate g (grey line) and SSA as a function of wavelength. Then, a linear empirical adjustment is made to g to force the Mie-calculated g to match the measured g (black line).

- **Our measurements applied to the radiative transfer model results in 20% more cooling than the model based on literature values.**
- This calculation takes into account only the shift in the direction of the scattered light, and neglects any change in total amount of light scattered.
- Increasing the assumed refractive index of the particles will also change the measured optical size of the particles.
- Both changes will result in a smaller modelled g, and therefore better agreement with the measurements.



Conclusions and future directions

- For there to be agreement between the modelled and measured g, one must use a higher refractive index than the remote retrieval literature for both the particle size measurements and the Mie calculations.
- Direct measurements of the asymmetry parameter show models may be underestimating the cooling effect of smoke by 20%.
- We will use the GRASP retrieval algorithm to find the optimal solution for the refractive index of the smoke.

Acknowledgements

This work was supported by the NOAA Cooperative Agreement with CIRES, NA17OAR4320101.

References

Ahern, A. T., et al. (2022). "Laser imaging nephelometer for aircraft deployment." Atmos. Meas. Tech. 15(5): 1093-1105.
Chylek, P. and J. Wong (1995). "Effect of absorbing aerosols on global radiation budget." Geophysical Research Letters 22(8): 929-931.
Dubovik, O., et al. (2002). "Variability of Absorption and Optical Properties of Key Aerosol Types Observed in Worldwide Locations." Journal of the Atmospheric Sciences 59(3): 590-608.
Espinosa, W. R., et al. (2019). "Retrievals of Aerosol Size Distribution, Spherical Fraction, and Complex Refractive Index From Airborne In Situ Angular Light Scattering and Absorption Measurements." Journal of Geophysical Research-Atmospheres 124(14): 7997-8024.
Henyey, L. G. and J. L. Greenstein (1941). "Diffuse radiation in the galaxy." The Astrophysical Journal 93: 70-83.
Marshall, S. F., et al. (1995). "Relationship between asymmetry parameter and hemispheric backscatter ratio: Implications for climate forcing by aerosols." Applied Optics 34(27): 6306-6311.
Womack, C. C., et al. (2021). "Complex refractive indices in the ultraviolet and visible spectral region for highly absorbing non-spherical biomass burning aerosol." Atmospheric Chemistry and Physics 21(9): 7235-7252.