

This manuscript communicates a new optical instrument that helps address the ever-important need to derive complex refractive indices from biomass burning aerosol, a key parameter in satellite retrieval algorithms and climate modeling. The authors also describe its deployment to the 2016 FIREX experiments and the results from several biomass burning experiments. It is unsurprising that light scattering from fractal BC aggregates cannot be computed from Mie theory, which follows immediately from the requirement of spherical particles. It is even less surprising that the authors conclude that the retrievals are quite sensitive to the size distribution measurement, as errors in broad size distribution measurements will likely dominate the errors propagated from their optical instruments.

I recommend that this manuscript be published pending some minor technical corrections and the satisfactory response to a few minor questions and comments I have below. This manuscript is quite well-written, clearly organized, and follows a logical path from introduction to conclusion. I thank the authors for the opportunity to review their work.

Comments and questions

I appreciate the discussion of the difficulty in ascertaining the error in refractive index, this is something that the community seems to be in disagreement over, though this leads me to a larger point of how we discuss the refractive index in aerosol optics. Fundamentally, it is inappropriate to report the refractive index as $m = (n \pm \sigma_n) + (k \pm \sigma_k)$, since n and k aren't independent of one another, and are rather functions of each other through the Kramers-Kronig relations. The refractive index is a single, complex quantity. However, given the way you conducted your experiments, I believe the way you represented your errors is appropriate. However, in section 4.3, you refer to n as the “scattering component” and k as the “absorbing component”. Both n and k contribute to the overall optical behavior. I suggest re-writing this section to remove the notion that n and k are purely responsible for scattering and absorption, respectively. Using “real part” and “imaginary part” is appropriate.

How fast was your SMPS scan? I'm curious about the width of the transfer function.

In the second paragraph in section 2.5, you assert that aerosol RI is unaffected by wall losses. While this may be practically true, this is not universally true, especially for the reason you state. It certainly is for a homogeneous substance such as pure ammonium sulfate aerosol or PSL spheres. For a complex mixture such as biomass burning, aerosol RI is rather the “effective aerosol RI”, where the particle behaves as if it has a single RI. In turn, the entire aerosol population being sampled will behave as if it has a single effective RI. However, getting down to the losses of particles of certain sizes, I would expect that the volatility of different compounds may preferentially distribute them to smaller or larger particles, and therefore losses above or below a particular size range may impact your measurements. I would certainly expect the effect to be slight, and it in fact may be negligible within the accuracy of your measurements. If this is the case, this is no more than an extremely minor semantics quibble, however, I believe this

section would benefit from a more accurate statement of exactly why you assume wall losses to be negligible on the optical properties of the population as a whole. You do address this in the first paragraph of section 3.3 where you state that you assume the RI does not vary systematically with size. This statement largely satisfies this comment.

Minor comments and technical corrections

Line 104 – “The very broadband...” language is vague and subjective. Consider removing “very”.

Line 277 – suggest changing “2× and 3× greater” to “two and three times greater”.

Line 284 – typo, change “existing” to “exiting”.

Line 297 – Consider moving “Theoretically” to the beginning of the sentence: “Theoretical particle losses between the DMA...”

In section 3.3.2, you state Mie theory is valid when the size parameter x is approximately 1. Mie theory is always valid, despite the size parameter. At lower size parameters, the Rayleigh *approximation* is perfectly fine, and at higher size parameters, geometric optics is a more useful approximation, since the number of terms you need for an accurate Mie theory calculation grows as $x + 4x^{1/3} + 2$ (Wiscombe 1980). Furthermore, B&H’s original FORTRAN algorithm is only valid up to $x \approx 10^4$ (Wolf and Voshchinnikov 2004). Since you use an adaptation of this code, I would rather state “The Mie theory algorithm we used here is valid when $x \approx 1$.” This is a minor suggestion, and the authors may take or leave it.

Line 304 – “It is a series approximation that allows a mathematical representation of light with spheres...” is slightly awkward usage. Consider “It is a truncated infinite series representation of the electromagnetic field scattered from spheres...” or something similar.

Line 411 – Remove comma after “function”.

Throughout – when the imaginary component of m is zero, write “0.00i” to be consistent with your measurement and retrieval precision.

Figure comments

In general, the figures are excellent. The instrument diagrams are fully informative without being cluttered, and I do appreciate that. The graphs, however, are coming across in the pdf as quite low resolution upon zooming in. This may be an artefact of the proof pdf, but do make sure that the journal has access to the highest quality figures you can produce. I would also like to caution that in some figures (notably 3b and 4), the legends dominate the figure area and distract from objective interpretation of the data. Consider moving the legends outside the figures.

In figure 6, it is quite difficult to take in all that data at once. Is it necessary to have the calculated cross sections in each panel? If so, you have ten traces to keep track of. I suggest

making the figure span the whole page and see if a logarithmic y-axis helps separate the data. As it is, it's extremely cluttered.

For all figures where you present k , consider a logarithmic scale. Since k is highly sensitive, far more so than n , a logarithmic scale will better convey the spectral functionality at lower values.

Finally, carefully consider the color schemes you use for data-dense graphs. When printed in grayscale or proofed for colorblindness, many traces are indistinguishable from one another.

References

Wiscombe, W. J., Improved Mie scattering algorithms. *Appl. Optics*. **1980**, 19 (9), 1505.

Wolf, S., and Voshchinnikov, N. V., Mie Scattering by Ensembles of Particles with Very Large Size Parameters. *Comput. Physics Commun.* **2004**, 162 (2), 113-123.