

FORM A: GACP ACCOMPLISHMENT REPORT

Names: K. A. Fuller (PI) / S. M. Kreidenweis (Co-PI) D. W. Mackowski (Co-PI)
Institutions: Colorado State University Auburn

MODELED AEROSOL OPTICAL PROPERTIES FROM MEASUREMENT-BASED MIXTURES OF CHEMICAL SPECIES—ASSESSING THE IMPACTS OF PARTICLE MORPHOLOGY AND ABSORPTION.

ABSTRACT: The research proposed here addresses the need for studies of radiative properties of mixed-composition aerosols, constrained by field observations, with a focus on the role of absorbing and nonspherical components. We have assembled a team of researchers with skills uniquely suited to the tasks at hand. Specifically, we will perform modeling studies that are guided by field measurements of size-resolved chemical speciation, hygroscopicity, dry aerosol size distributions, and aerosol scattering and absorption properties, from the summertime SouthEastern Aerosol and Visibility Study (SEAVS), in which one of us (SMK) participated as a principal investigator. We will extend this work using aerosol data from the IMPROVE (Interagency Monitoring of Protected Visual Environments) network, with which we have worked in the past. A radiative model that can accurately represent the effects of externally and internally mixed soot and of light absorbing mineral components of the aerosol will be completed, and will be used to test the effects of the mixing assumptions for absorbing and nonabsorbing species. By comparison with field data, we hope to be able to recommend appropriate treatments of the radiative properties of such mixtures under various conditions. As a step toward constraining uncertainties in radiative forcing and sensitivities of satellite-based retrievals, we will use the optical models to explore changes in column forcing and clear-sky reflectance with measured changes in aerosol composition and ambient conditions.

GOALS: Provision of a model continental aerosol based on point measurements of particle chemistry and (where possible) population statistics that can be used by the Earth system science community for climate diagnostics.

OBJECTIVES: .

1. To develop optical models for aerosols that are specific to measured chemical and physical particle properties at selected geographic locations.
2. To provide the aerosol data and optical models to the GACP community for use in testing retrieval algorithms and aerosol transport models.
3. Provide extensive calculations for polarized light scattering by compounded spherical particles for
 - (a) use in aerosol optical models
 - (b) the simulation of the optical properties of mineral dust
 - (c) development and testing of effective medium theories and other approximations for light scattering by nonspherical particles
 - (d) potential spin-offs in for example biosensor, combustion diagnostic, ceramics processing, and pharmaceutical applications (for example).

APPROACH: .

The effects of soot on the radiative properties of the atmospheric aerosol are being modeled with a light scattering theory that is an extension of the theory for homogeneous or concentrically stratified spheres. Soot is considered to be an agglomeration of carbonaceous spherules and these aggregates may, in turn, be internally mixed with sulfate and organic particles.

Data collected for aerosol properties is being ingested into models that output aerosol refractive index and size distribution parameters as functions of RH, and that can then be processed by an optical model that produces optical parameters for use in radiative transfer routines.

TASKS COMPLETED:

1. Contributed the chapter 'Electromagnetic Scattering by Compounded Spherical Particles' to the book *Light Scattering by Nonspherical Particles: Theory, Measurement, and Applications*, Academic Press, 1999.
2. Measurements at Great Smoky Mountains (GSM) National Park, made in summer/95, of aerosol chemistry and population statistics have been used to provide humidity-dependent datasets that detail changes in material properties and total aerosol mass with water uptake for an atmospheric aerosol typical of the southeastern United States.
3. Computer code for calculation of optical properties of aggregates of large numbers (100-600) of spheres, with each sphere having a size parameter of 5–20, was developed and tested.

4. An effective-medium approximation for coherent wave propagation in a semi-infinite matrix of spheres has been developed to calculate the scattering and absorption properties of aggregated particles. The method can accurately predict the scattering and absorption cross sections and scattering asymmetry parameter of clusters containing a relatively large number of inclusions.

FUTURE PLANS: (Plans for the immediate future have anticipated completion dates in parentheses)

- Posting of sphere aggregate codes to GACP web page. (8/30/99)
- Posting of codes for homogeneous and concentrically stratified spheres to GACP web page. (8/30/99)
- Calculation of column forcing of the aerosol over GSM during the 1995 observation period. (9/30/99)
- Calculation of clear-sky satellite radiances and their sensitivity to changes in humidity for the above GSM aerosol. (9/30/99)
- Posting of GSM aerosol data to GACP web page. (9/30/99)
- Single scattering optical parameters for compounded particles comprised of a few subspheres. (9/30/99)
- Averages of these properties over spectral bands that are compatible with those used in forcing calculations, or with radiometer channels of specific satellites.
- Compare radiative properties for externally mixed particles, homogeneous internally mixed particles, and composite internally mixed particles.

RESULTS: Incorporation of the physical data collected at GSM into our scattering calculations are providing results, the dated items above, for discussion at the GACP Science Team meeting at the end of Sept.

FORM B: GACP SIGNIFICANT HIGHLIGHTS (PART I)

Names: Kirk A. Fuller (PI) / Sonia M. Kreidenweis (Co-PI) Daniel W. Mackowski (Co-PI)
Institution: Colorado State University Auburn

SIGNIFICANT HIGHLIGHTS:

Beginning with Lorenz-Mie (LM) theory for light scattering by single spheres and the addition theorem for vector spherical harmonics, we review advances made in the study of the optical properties of systems of compound spheres. For organizational purposes, these systems will be divided into two categories: (1) Aggregates of two or more spherical particles and (2) one or more spherical inclusions located arbitrarily within a spherical host. As will be discussed, these two categories differ only by certain details at the level of the addition theorem, and are essentially described by a single theory which encompasses clusters of included spheres. We also discuss comparisons of theory and measurement, as well as applications in climatology, visibility modeling, atmospheric remote sensing, planetary astronomy, stellar evolution, fractal optics, nonlinear optics, photonics, sensors, quality control and process diagnostics.

A version of this chapter will be posted on the web in .pdf format. This version will be revised as errors in the original manuscript come to light, and the reference list will be updated periodically. This chapter will provide the foundation upon which our light scattering contributions to the GACP are based, and will provide a reference for use with the computer programs which we post.

FORM B: GACP SIGNIFICANT HIGHLIGHTS (PART II)

The aerosol chemical compositions for each day of the GSM study were used to compute the (volume averaged) refractive index and material density of the aerosol particles, assuming only sulfates were hygroscopic. The water amount at 15% RH was computed for each day based upon the ammonium to sulfate ratio for that day (so more acidic days had more water per unit aerosol mass than those days with more dust-like components). The 15% RH condition was assumed to be the “dry” aerosol, although even then there was some water associated with it that went into the refractive index and density calculation for “dry”. The same ammonium to sulfate ratio was then used to compute the water contents at other RHs. That water was included in the RH-dependent refractive index and density calculations.

A lognormal was fit to dry OPC data, so the reported dry diameter is that at 15% RH. Using the density and total mass at every other RH, a new aerosol diameter was calculated. The ratios are reported as $D(\text{RH})/D_0(\text{RH}=15\%)$. It is important to note that the lognormal applies only to the accumulation mode, whereas the chemical composition is from PM 2.5 data. Because of this the number of accumulation mode particles is also provided in the tabulated data. This can be used to get the total volume of the accumulation mode which should be less than deduced from total PM2.5 mass and density. The difference is coarse mode (which the OPC did not measure).

The water uptakes for the daily measured aerosol chemistry, along with certain meteorological events are displayed in Figure 1.

FORM B: GACP SIGNIFICANT HIGHLIGHTS (PART III)

As part of a collaboration with Dr. Richard Chang of Yale University, the scattering properties of large-scale clusters of latex spheres were calculated by our group (using the theory for compounded spherical particles) and compared to experimental measurements, performed at Yale for two-dimensional light scattering from clusters that are held in an acoustic trap. Preliminary results indicate a good comparison between the theoretical and experimental values of S_{11} (scattering phase function) which, for the most part, is dependent primarily on the overall size parameter of the cluster. Additional calculations have indicated that measurements of polarized light scattering by the cluster – especially the degree of linear polarization S_{12}/S_{11} – could yield information on the monomer (i.e., individual sphere) size.

A sample of the calculation results are shown in Figure 2. Plotted are planar projections of S_{12}/S_{11} as a function of polar and azimuth scattering angles ϑ and φ , into the back scattering hemisphere for three dense-packed sphere cluster configurations. Polar and azimuth angles correspond to latitude and longitude, respectively, on the map, with the backwards scattering direction ($\vartheta = 180^\circ$) at the center (pole). The number of spheres in the cluster are $N_S = 13, 57, \text{ and } 159$, with respective sphere size parameters of 21.67, 13, and 9.29. The refractive index of the spheres is $1.5 + 0.01i$. Each cluster is roughly spherical in shape, and the overall size parameters of the cluster (based on the circumscribing sphere) are the same and equal to 65. The results for each map correspond to a fixed angle of incidence of the incident beam with respect to the cluster—which simulate the conditions of the experiment.

The two-dimensional variation in S_{12}/S_{11} that is seen in the maps is due entirely to multiple scattering of waves among the spheres in the cluster—note that the Rayleigh-Gans approximation (which would model only far-field wave mixing) would give results of S_{12}/S_{11} identically equal to that of a sphere (i.e., no azimuthal variation). However, ‘rings’ in the pattern—centered about the pole—can be discerned among the random-like ‘noise’, and the ϑ location of the rings corresponds closely to that for the individual, isolated sphere in each cluster. Although the work is preliminary, the results do suggest that polarized light scattering could be used to probe the structure of aggregated particles.

FORM C: FUTURE PLANS

Names: Kirk A. Fuller (PI) / Sonia M. Kreidenweis (Co-PI) Daniel W. Mackowski (Co-PI)
Institution: Colorado State University Auburn

- Carbonaceous Particles Mineral Dust
 - Calculate orientation-averaged optical properties of droplets with many inclusions
 - Calculate radiative properties of internally mixed soot fractals
 - DDA-Multipole hybrid calculations for mineral dust
- Extend model calculations to include data from IMPROVE project using assumed size distribution parameters suggested by SEAVS
- Estimate regional forcing by dust component
- Begin long wave forcing and UV-B flux calculations
- Develop recommendations for GCM modeling
- Deliverables
 - Paper on short wave forcing
 - CSU technical report describing IMPROVE data utilized in radiative transfer modeling study.
 - Updates to web site

FORM D: GACP BIBLIOGRAPHY

Names: Kirk A. Fuller (PI) / Sonia M. Kreidenweis (Co-PI) Daniel W. Mackowski (Co-PI)
Institution: Colorado State University Auburn

BIBLIOGRAPHY:

Papers, reports, and presentations

a. List of publications (including books, book chapters, and refereed papers).

- ‘Electromagnetic Scattering by Compounded Spherical Particles,’ in *Light Scattering by Nonspherical Particles: Theory, Measurements and Applications*, M. I. Mishchenko and L. D. Travis, Eds., Academic Press, New York, 1999.
- ‘Effects of mixing on extinction by carbonaceous particles: II possible artifacts associated with particles packed on a filter,’ K. A. Fuller, S. M. Kreidenweis, and W. C. Malm, *in preparation* for submittal to J. Geophys. Res.
- ‘Issues in Radiometry and Absorption by Filter Samples,’ K. A. Fuller, S. M. Kreidenweis, and W. C. Malm, *in preparation* for submittal to Appl. Opt.
- ‘Mueller matrices for complex aggregates: Implication for satellite-based retrieval of dust loadings,’ K. A. Fuller, D. W. Mackowski, and S. M. Kreidenweis, *in preparation* for JGR Special Issue on Atmospheric Dust.
- ‘Effective medium formulation for scattering and absorption of sphere aggregates,’ D. W. Mackowski, *in preparation* for submission to J. Opt. Soc. Am. A
- CSU-Auburn atmospheric aerosol model, Parts I & II. *planned*.

c. Presentations

- ‘Calculation of scattering by sphere clusters,’ invited presentation, Applied Physics Department, Yale University, April 1999.
- ‘Regge, Clebsch-Gordan, and Mie: Theory and Applications of Light Scattering by Compounded Spherical Particles,’ invited presentation to the Department of Physics and Astronomy, Vanderbilt University, July 1999.
- ‘Applied Microparticle Optics and Radiometry,’ presented to the Global Hydrology and Climate Center, University of Alabama in Huntsville, July 1999

FORM E: GACP METRICS

Names: Kirk A. Fuller (PI) / Sonia M. Kreidenweis (Co-PI) Daniel W. Mackowski (Co-PI)
Institution: Colorado State University Auburn

METRICS (not including those in progress or planned):

- a. Number of publications (including books, book chapters, and refereed papers).
1
- b. Number of printed technical reports and non-refereed papers
- c. Number of oral or poster presentations at professional society meetings and conferences. 3
- d. Number of advance degree students:
 - i. current
 - ii. graduated
- e. Number of post-doctorates.
- f. Number and names of proposals:
 - i. submitted
 - ii. accepted
- g. Number of proposals and/or papers that you reviewed. 6
- h. Number of committees served on (outside your organization).
- i. Number of patents (granted and applications).
- j. Number and names of honors and awards.