

Modeling the Time-Dependent Optical Properties of the Multicomponent Aerosols in the Marine Boundary Layer

PI: Prof. Irina N. Sokolik
Program in Atmospheric and Oceanic Sciences (PAOS)
University of Colorado at Boulder
Campus Box 392
Boulder

Phone: 303-492-5724 fax: 303-492-6946 e-mail: irina.sokolik@colorado.edu

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<http://irina.colorado.edu>

LONG-TERM GOALS

The ultimate goal of this project is to improve the predictive understanding of the time-dependent, frequency-dependent, radiative properties of multicomponent aerosols containing mineral dust in the marine environment.

OBJECTIVES

Specific objectives of our research are:

- (1) elucidate the links between dust particles morphology (shape and size), composition, optical properties and related radiative effects;
- (2) relate the properties of dust to its source and investigate the evolution of dust properties during transport in the marine boundary layer, focusing on the comparative analysis of the Asian, African, Southwestern U.S., and Saudi Arabian types of dust.
- (3) improve algorithms for prediction of frequency-dependent optical properties of mineral dust accounting for its mineralogical composition, life cycle, and interaction with other atmospheric aerosols in the clean and polluted marine environment.

APPROACH

Our approach combines an integrated analysis of the empirical data on dust microphysical, optical, and radiative properties and advanced numerical modeling techniques. During FY2003, we were focusing on the development and testing of the new spectral optical models of internally mixed aerosols containing dust in the near-IR and the IR.

WORK COMPLETED

We have compiled an extensive data set of the refractive indices from the UV to the infrared of the most common tropospheric aerosols components (such as minerals, black carbon, sulfates, sea-salt, and nitrates). All data were incorporated into a library called the Library of Atmospheric Aerosol Refractive Indices (LAARI). In this database we preserved the original data, but we also attempted to construct continuous spectra for individual species that cover the wavelength region from 0.2 to 40 nm.

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The continuous datasets that we have found or constructed have been interpolated to a spectral resolution of 10 cm^{-1} . This database will provide new capabilities for aerosol modeling and retrieval algorithms. Using the LAARI data, we performed an extensive study of the impact that various computational simplifications/assumptions have on the resulting volume optical properties, e.g. effective refractive index mixing rules, mixed particle morphology, and internal vs. external mixtures. Several methods (volume averaged refractive index, volume averaged dielectric constant, volume averaged cube root of the dielectric constant, Bruggeman, and Maxwell-Garnett approximations) to generate an effective refractive index for internally mixed particles were tested and compared (Boer and Sokolik, 2003a, b). Analysis of the sensitivity of near-IR and IR radiances to the composition and vertical distribution of internally mixed aerosols under various atmospheric conditions was carried out. We demonstrated that the presence of atmospheric aerosols strongly affects the IR emission and hence must be taken into account in the retrieval algorithms. Several recommendations for aerosol correction algorithms were suggested. We also continued our work on the improvement of dust models for remote sensing in the visible. Based on a new technique developed in FY 2001-2002, we proposed dust optical models for the Saharan dust analog and Asian dust analog for remote sensing at solar wavelengths. These models were further advanced by incorporating new data on the dust composition and shapes (Kalashnikova and Sokolik, 2003). Using these models, we investigated the ability of multi-angle, multi-spectral measurements to distinguish between different mineral dust types. These models are currently tested in the MISR aerosol retrieval algorithms (Kalashnikova et al., 2003). Overall, we successfully completed all tasks planned for FY2001-2003.

RESULTS

Calculations of the optical properties of aerosols that are needed for the interpretation of remote sensing data require knowledge of the microphysical properties of the aerosols such as particle sizes and spectral refractive indices which are controlled by particle composition. Since aerosol particulates often occur as a complex multicomponent and multiphase mixture of various individual species, methods to handle internally mixed aerosols are also needed. To this end, we have compiled a new library of spectral refractive indices (LAARI) of main individual aerosol species and performed detailed modeling of internally mixed aerosols. Several effective medium approximations (EMA) to generate an effective refractive index for internally mixed particles were tested and compared. Our analysis revealed the significant differences in the spectral effective refractive index depending on the components and the mixture method that was chosen. Detailed analysis of the sensitivity of near-IR and IR radiances to the composition and vertical distribution of internally mixed aerosols was performed and implications for remote sensing of aerosols, trace gases (CO and O_3) and sea surface temperature were evaluated.

Relative humidity affects the size and composition of soluble atmospheric aerosols. As the relative humidity varies, either more water vapor condenses onto the aerosol from the atmosphere, or more liquid water from the aerosol moves to the vapor phase. The change in liquid water amount results in a change in the refractive index and the size of the particles. Both changes alter the optical properties of the aerosols. Thus, accounting for relative humidity in a consistent way has important consequences on the size and composition of aerosols with soluble components and hence on the optics. There are a number of ways to include relative humidity effects in the computation of aerosol optics, e.g. empirically scaling the volume or particle optical properties, numerically solving the full thermodynamics equation, or using measurement data of water activities. Different techniques can give significantly different results. To address this problem, we developed a new technique for computation of aerosol optical properties at different relative humidity ranging from high RH in the

marine boundary layer for low RH in the free troposphere. Our approach is to use water activity data (e.g., *Tang et al.*) to determine the weight concentration of the resulting aqueous solution. Then the refractive index of the solution is determined as a volume average of the refractive index of water and solute. This technique was used to calculate the optical properties of dust particles coated by salt aqueous solutions at varying RH. As a result, we were able to investigate the role of the dust vertical distribution (dust in the marine boundary layer vs. dust in free troposphere) on the IR radiances. High spectral resolution near-IR (3.0 – 5.0 μm) radiances are important to key applications, including trace gas retrievals (CO and CH₄) and atmospheric sounding. Considerable efforts have been invested in developing the methods to account for the effects of aerosols in the UV/VIS spectral region. Similarly, efforts are currently underway for the thermal infrared region. However, the impact of aerosols on top of atmosphere radiances measured in the near-IR spectral region has been largely overlooked. Using dust models developed under this grant, we performed detailed forward modeling studies to explore the sensitivity of high-resolution near-IR spectral radiances to various aerosol characteristics, including aerosol composition, size spectra, vertical distribution and loading. We demonstrate that the presence of atmospheric aerosols can strongly affect absorption/emission and scattering in the near-IR. Models for aerosol corrections in retrieval algorithms were proposed. In summary, new techniques and new optical models for mineral dust and its internal mixtures with other aerosol species developed under this grant provide new capability for aerosol retrievals from passive remote sensing from the UV to the IR, as well as for aerosol corrections. The models also aid to the interpretation of electromagnetic wave propagation in the marine boundary layer.

IMPACT/APPLICATIONS

New techniques and the models of the spectral optical properties of multicomponent aerosol containing dust developed under this grant can be employed in various remote sensing applications and in aerosol chemical transport models.

TRANSITIONS

Our main results were published in peer-reviewed journals and presented at numerous scientific meetings.

RELATED PROJECTS

We are funded by MURI/University of Wisconsin to develop new dust models for hyperspectral remote sensing in the IR. Overall, our work on both projects will provide a framework for the development of the new generation of aerosol models required for remote sensing from the UV to the IR. PUBLICATIONS in 2003 Boer G. and I.N. Sokolik, Developing the high spectral resolution aerosol models for remote sensing in the thermal IR, IGARSS, IEEE International Geoscience and Remote Sensing Symposium, July 21-25, Toulouse, France, 2003a [published, refereed]

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