Final Report

Title: Characterization of Mineral Dust Aerosols to Improve Predictions of Their Impact on the Radiative Balance of the Atmosphere

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Project Summary

Atmospheric dust aerosols can affect the Earth's climate system via a number of complex processes: by altering the radiative budget at the top-of the atmosphere and surface, causing radiative heating or cooling within the dust layer, affecting the photochemistry of the atmosphere, and changing the cloud properties and precipitation, among others. Up to now, all climate models as well as remote sensing retrievals consider dust as a single generic species. Growing evidence demonstrates that distinct regional differences of dust properties are the key factors that control dust impacts on the climate system. Therefore, there is an urgent need to improve the treatments of dust in GCMs to make their predictions more realistic. This provides the motivation for this proposal.

This project conducted a comprehensive study of physicochemical properties of mineral dust representative of main dust production regions: its source-dependent composition, light absorption, CCN activity, and interactions with water clouds. The main goal is to provide critical insights on dust properties and develop specific recommendations on dust treatments in the climate models required for improved assessments of dust impacts on the radiative balance of the atmosphere. The approach integrates laboratory experiments, *in situ* ground-based and satellite data, and several numerical models. The specific objectives are to:

1) perform new measurements of physicochemical properties of mineral dust samples representative of major individual dust sources, including dust sources in the U.S., to provide the critical data required for improved predictions of dust direct radiative impacts;

2) investigate how the distinct regional properties of dust affect its interactions with water clouds by performing laboratory measurements and numerical modeling in conjunction with analysis of satellite observations of mixed dust-cloud scenes;

3) investigate the role of regional differences in dust properties in affecting the radiative balance of the atmosphere directly and indirectly (via clouds) and develop recommendations for improved dust treatments in global climate models.

Summary of Accomplishments:

For this project we have developed a new dust system for generation of mineral aerosol from parent soils and individual minerals. The uniqueness of the Georgia Tech dust system is that it includes a soil-to-aerosol generator which reproduces (via "shaking") the processes governing dust emission in natural environments. This "dry" generation of dust aerosol in the lab is more realistic compared to "wet" generation types (e.g., via atomization processes or fluidized bed generation) that were used in the majority of past studies. Generated dust aerosols as well as some atmospheric dust samples were used for measuring the physiochemical properties of particles (size, composition, shape, and soluble ions) and CCN activation (see 1.1 below). Measurements were performed for dust samples representative of the major dust source regions (Northern Africa, East Asia, and US). The data set provide for the first time a comprehensive compilation of dust properties enabling development of improved aerosol optical models and new dust CCN activation parameterizations (see 1.2 and 1.3).

1.1 Measurements of properties and CCN activity of dust aerosol: main results:

The dust generation system was incorporated into a laboratory setup to enable on-line measurements of dust CCN activation using a Continuous-Flow Streamwise Thermal Gradient CCN Counter (CFSTGC). The setup also includes particle size selection and measurements using a Differential Mobility Analyzer (DMA) and Optical Particle Counter (OPC) (Kumar et al., 2010a). CCN measurements were performed with aerosols generated from soil samples representative of the U.S. (white sand), Northern Africa (Niger soil), and East Asia dust sources (totally about 30 different samples). Furthermore, CCN activities of various individual minerals such as clays (kaolinite, illite, and montmorillonite), carbonates, and quartz were measured to facilitate interpretation of regional mineral aerosol CCN activities in terms of mineralogical composition of dust samples. In addition, a series of experiments exploring how the wetting affects the activation characteristics of dust aerosols was performed, including measurements of soluble ions (Kumar et al., 2010b). Measurements of dust particle morphology (3D shapes) (Sokolik et al. 2010) help to perform a correction for the dust non-sphericity effect in dust CCN activation measurements.

One key finding was that Köhler theory that commonly being used for modeling dust CCN activation could not reproduce the measurements. To overcome this problem, we proposed a new framework based on the absorption activation theory that showed excellent agreement with experimental data. Another important finding was that dust aerosols displayed the retarded activation kinetics compared to soluble $(NH_4)_2SO_4$ aerosols. Comprehensive simulations of activation and growth in the CCN instrument suggest that the delays are equivalent to a reduction of the water vapor uptake coefficient (relative to that for $(NH_4)_2SO_4$) by 20-80% (Kumar et al., 2010a). These results suggest that dust particles do not require deliquescent material to exhibit hygroscopicity.

1.2 Parameterizations of dust CCN activation for regional and global models.

Motivated by observational evidence favoring the absorption activation theory over Köhler theory, we have developed a new physically based parameterization for dust particle CNN activation using the multilayer Frankel-Halsey-Hill (FHH) adsorption isotherm, which was modified to account for particle curvature (Kumar et al., 2009a, b). The new activation parameterization is based on the cloud parcel framework that considers cloud droplet formation within an ascending air parcel containing hydrophilic insoluble particles. Formulations were developed for sectional and lognormal representation of aerosol size distribution so they can be incorporated into various regional or global models. We also tested another approach based on a multilayer adsorption isotherm model (Brunauer-Emmett-Teller, BET) that can also describe adsorption activation of dust into cloud droplets (Kumar et al., 2009b). We examined κ-Köhler theory (KT) and FHH adsorption activation theory (AT) for insoluble mineral dust aerosol using our new dust-CCN measurements along with data reported by other groups. We demonstrate that observed dust CCN activity suggests that the critical supersaturation scales with dry diameter are substantially different from -3/2, typically associated with KT. Adsorption activation theory is better suited to describe activation of freshly emitted mineral dust aerosol. When consistent κ-and adsorption parameters are applied to the predictions of CCN concentrations, KT predicts up to tenfold higher CCN than AT. Even though KT particles require more water vapor than AT to activate, the higher CCN concentrations associated with KT result in larger droplet concentrations. Furthermore, KT cannot describe dust hygroscopic growth in both sub- and supersaturated conditions.

1.3 Assessments of dust impacts on atmospheric chemistry and Earth's radiative energy budget

The new data on physicochemical properties of regional dust samples obtained under this project along with data from recent field campaigns and satellite aerosol products made it possible to investigate how different regional dust types affect the shortwave (SW) and longwave (LW) components of the radiative budget at the surfaces and the top-of-the-atmosphere (TOA) (i.e., radiative forcings), as well as examine the dust impact on atmospheric chemistry through both photochemistry and heterogeneous chemistry processes.

We developed a set of spectral optical characteristics of dust taking into account size-resolved composition data (Jeong and Sokolik, 2007; Xin and Sokolik, 2010). The dust optical models were incorporated into the National Center for Atmospheric Research tropospheric ultraviolet-visible radiation transfer code (TUV) to examine the effect of regional dust composition on the atmospheric photochemistry (Jeong and Sokolik, 2007) and into the SBDART radiative transfer code that was used in assessments of dust regional radiative forcing (Xin and Sokolik, 2010).

Regarding the photochemistry, we analyzed (13) photolysis reactions that were classified into three groups according to their photolytic wavelengths and the vertical profile of J values in the aerosol-free atmosphere. The photolysis reactions of O3(O1D), NO2, and NO3(NO) were selected as representative of groups I, II, and III, respectively. We found that depending on its properties, dust causes either a decrease or an increase in the spectral actinic fluxes relative to the aerosol-free condition. The wavelength range in which the changes in actinic fluxes are negative becomes broader as the amount of dust load increases, a dust size distribution is shifted to coarse size mode, and the iron oxide content in dust aggregates increases. Changes in actinic fluxes also depend on the sun position (time of the day) and an altitude considered. As a result, dust exerts the differing impact on J values of the three photolytic groups. The diurnal cycle of dust-affected J values of a given group is similar among the differing size distribution and dust compositions, but changes in J values vary by a factor of 1.5–2. For a given content of iron oxide, the largest changes are caused by the size distributions that are shifted to the fine size mode. A change in J values of groups I and II caused by the varying amount of iron oxide in dust aggregates (from 1% to 10%) is negative in and below the dust layer. In contrast, J values of group III increase in the presence of low absorbing dust (with 1% of iron oxide), but they decrease with increasing dust absorption. Our results indicate that the dust composition not only will be needed to accurately model a decrease in J values of groups I and II but also to determine a correct sign and value of changes of J values of group III. In the case of dust mixed with pollution, we found that the external mixing of dust and black carbon causes somewhat larger changes in J values compared to the internal mixing of these species (Jeong and Sokolik, 2007). Furthermore, we examined the heterogeneous chemistry occurring on dust particle surfaces and its relative importance compared to the dust-affected photochemistry. Both mechanisms strongly depend on the size and composition of dust aerosols. Our study was the first to examine these mechanisms by introducing a consistent representation of the size-resolved mineralogical composition of dust in the modeling actinic fluxes and heterogeneous loss rates (Jeong and Sokolik, 2010). We demonstrate that region-specific differences in microphysical and chemical properties of mineral dust lead to important changes in spectral optical properties, photolysis rates, and heterogeneous loss rates, stressing the need for improved treatments of dust in regional and global chemical transport models. It was also shown that non-linear relationships of photochemical species with two mechanisms result in various changes in the photochemical oxidant fields and that the most important controlling factors are the dust size distribution, followed by the amount of mineral dust species with high uptake coefficients, and the amount of iron oxide-clay aggregates.

Assessments of dust radiative forcings at the surface and TOA were done by performing the radiative transfer modeling with SBDART (Xin and Sokolik, 2010) in conjunction with analysis of satellite aerosol products in dust-laden conditions.

Analysis of satellite data was focused on the regional specific of Asian dust properties. In collaboration with the MISR aerosol science team, we analyzed 10-year Terra Multi-angle Imaging SpectroRadiometer (MISR), 7-year Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) Deep Blue, and 5-year Aura Ozone Monitoring Instrument (OMI) (Kalashnikova et al., 2008, 2009, 2010; Sokolik et al. 2008, 2009). Multi-year mean spatial patterns, seasonal cycle, and inter-annual springtime variability/anomalies of aerosol optical thickness (AOT) and OMI aerosol index (AI) were examined to determine common features and differences among the sensors and implications to dust radiative forcing assessments. Based on this analysis and radiative transfer modeling, we assessed the range and efficiency of SW, LW and net radiative forcing of Asian dust aerosol (Xi and Sokolik, 2010). Our work demonstrated that the presence of dust leads to significant reduction in surface SW radiation and surface radiative balance. For the moderate loading, the dust SW surface forcing ranges from -38.9 Wm^{-2} to -287.2 Wm^{-2}. The percentage of SW forcing compensated by the longwave forcing ranges from 6.1% to 34.4%. Our study demonstrated that the important role of the dynamics of dust particle size distribution and spectral surface albedo in assessments of dust radiative forcing.

Peer-reviewed publications prepared with total or partial support from this grant:

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