

3. Aerosols and Radiation

3.1. AEROSOL MONITORING

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3.1.1. SCIENTIFIC BACKGROUND

Aerosol particles affect the radiative balance of Earth both directly, by scattering and absorbing solar and terrestrial radiation, and indirectly, through their action as cloud condensation nuclei (CCN) with subsequent effects on the microphysical and optical properties of clouds. Evaluation of the climate forcing by aerosols, defined here as the perturbation of the Earth's radiation budget induced by the presence of airborne particles, requires knowledge of the spatial distribution of the particles, their optical and cloud-nucleating properties, and suitable models of radiative transfer and cloud physics. Obtaining a predictive relationship between the aerosol forcing and the physical and chemical sources of the particles additionally requires knowledge of regional and global-scale chemical processes, physical transformation, and transport models for calculating the spatial distributions of the major chemical species that control the optical and cloud-nucleating properties of the particles. Developing and validating these various models require a diverse suite of in situ and remote observations of the aerosol particles on a wide range of spatial and temporal scales.

Aerosol measurements began at the CMDL baseline observatories in the mid-1970s as part of the Geophysical Monitoring for Climatic Change (GMCC) program. The objective of these "baseline" measurements was to detect a response, or lack of response, of atmospheric aerosols to changing conditions on a global scale. Since the inception of the program, scientific understanding of the behavior of atmospheric aerosols has improved considerably. One of the lessons learned is that residence times of tropospheric aerosols are generally less than 1 week, and another is that human activities primarily influence aerosols on regional and continental scales rather than global scales. In response to this increased understanding, and to more recent findings that anthropogenic aerosols create a significant perturbation in the Earth's radiative balance on regional scales [Charlson *et al.*, 1992; National Research Council, 1996], CMDL expanded its aerosol research program to include regional aerosol monitoring stations. The goals of this regional-scale monitoring program are: (1) to characterize means, variabilities, and trends of climate-forcing properties of different types of aerosols and (2) to understand the factors that control these properties.

A primary hypothesis to be tested by NOAA's aerosol research program is that the climate forcing by anthropogenic sulfate will change in response to future changes in sulfur emissions. The forcing is expected to decrease in and downwind of the United States as a result of emission controls mandated by the Clean Air Act, while continued economic development in China and other developing countries is expected to lead to an increased forcing in and downwind of those areas. Testing this hypothesis will require a coordinated research program involving modeling, monitoring, process, and closure studies. This report describes the observations that CMDL is conducting towards this goal.

No single approach to observing the atmospheric aerosol can provide the necessary data for monitoring all the relevant dimensions and spatial/temporal scales necessary to evaluate climate forcing by anthropogenic aerosols. In situ observations from surface sites, ships, balloons, and aircraft can provide very detailed characterizations of the atmospheric aerosol but on limited spatial scales. Remote sensing methods from satellites, aircraft, or the surface can determine a limited set of aerosol properties from local to global spatial scales, but they cannot provide the chemical information needed for linkage with global chemical models. Fixed ground stations are suitable for continuous observations over extended time periods but lack vertical resolution. Aircraft and balloons can provide the vertical dimension but not continuously. Only when systematically combined can these various types of observations produce a data set where point measurements can be extrapolated with models to large geographical scales, where satellite measurements can be compared to the results of large-scale models, and where process studies have a context for drawing general conclusions from experiments conducted under specific conditions.

Measurements of atmospheric aerosols are used in three fundamentally different ways for aerosol/climate research: algorithm development for models and remote-sensing retrievals, parameter characterization, and model validation. Laboratory and field process studies guide the development of parameterization schemes and the choice of parameter values for chemical transport models that describe the relationship between emissions and concentration fields of aerosol species. Systematic surveys and monitoring programs provide characteristic values of aerosol properties that are used in radiative transfer models for calculating the radiative effects of the aerosols and for retrieving aerosol properties from satellites and other remote sensing platforms. And finally, monitoring programs provide spatial and temporal distributions of aerosol properties that are compared to model results to validate the models. Each of these three modes of interaction between applications and measurements requires different types of data and entail different measurement strategies. Ogren [1995] applied the thermodynamic concept of "intensive" and "extensive" properties of a system to emphasize the relationship between measurement approach and applications of aerosol observations.

Intensive properties do not depend on the amount of aerosol present and are used as parameters in chemical transport and radiative transfer models (e.g., atmospheric residence time, single-scattering albedo). Extensive properties vary strongly in response to mixing and removal processes and are most commonly used for model validation (e.g., mass concentration, optical depth). Intensive properties are more difficult and expensive to measure than extensive properties because they generally are defined as the ratio of two extensive properties. As a result, different measurement strategies are needed for meeting the data needs of the various applications. Measurements of a few carefully chosen extensive properties, of which aerosol optical depth and species mass concentrations are prime candidates, are needed in many locations to test the ability of the models to predict spatial and temporal variations on regional to global scales and to detect changes in aerosol concentrations resulting from changes in aerosol sources. The higher cost of

determining intensive properties suggests a strategy of using a limited number of highly instrumented sites to characterize means and variabilities of intensive properties for different regions or aerosol types, supplemented with surveys by aircraft and ships to characterize the spatial variability of these parameters. CMDL's regional aerosol monitoring program is primarily focused on characterizing intensive properties.

CMDL's measurements provide ground truth for satellite measurements and global models, as well as key aerosol parameters for global-scale models (e.g., scattering efficiency of sulfate particles and hemispheric backscattering fraction). An important aspect of this strategy is that the chemical measurements are linked to the physical measurements through simultaneous, size-selective sampling that allows the observed aerosol properties to be connected to the atmospheric cycles of specific chemical species.