Chapter 1

INTRODUCTION

> "'I used to think of clouds as the Gordian knot of the problem,' says cloud specialist V. Ramanathan of Scripps. 'Now I think it's the aerosols. We are arguing about everything.'"
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> Richard A. Kerr, *Science*, 1997

In 1896 Svante Arrhenius asked, "Is the mean temperature of the ground in any way influenced by the presence of heat-absorbing gases in the atmosphere?" (Arrhenius, 1896), and opened the question of global warming. More than 100 years have passed, and, at last, we have the answer to this question with some degree of certainty. According to the most recent report of the Intergovernmental Panel on Climate Change (IPCC, 2001), various indicators "illustrate a collective picture of a warming world." While the change toward warmer climate is virtually certain, uncertainty remains about the reasons for this change. The IPCC report (2001) recognizes that natural climate variability alone is not enough to explain the observed trend, and points to human activity as another likely reason. Large uncertainties in observations and model estimates preclude reliable identification of the relative contribution of natural and anthropogenic climate agents to the global changes. Whatever the reasons though, natural or anthropogenic, it is necessary to predict climate changes timely and accurately in order to prevent or mitigate possible adverse consequences.

Climate change predictions employ both direct observations and numerical models. Ground and satellite measurements help to better understand and parameterize the physical, geophysical, chemical, and biological processes involved in the climate system. Climate models simulate the responses of the climate system to perturbations, such as the increase of greenhouse gas concentrations in the atmosphere.

The most challenging problem in climate modeling is the correct representation of various feedback mechanisms between the basic climate elements—the atmosphere and its composition (gases and aerosols), clouds, precipitation, and surface type (ocean, land, or ice). While there is clarity and agreement between different models for some mechanisms, e.g., the positive feedback of water vapor, there are difficulties about others, e.g., the net result of the interplay between the positive and negative feedbacks provided by the clouds (Wielicki et al., 1995). How the climate change would alter the net cloud forcing further complicates the problem.

The uncertainty in climate models due to cloud feedbacks initiated many programs for measuring the properties and global pattern of clouds (Wielicki et al., 1995, 1996; Durkee et al., 2000). The findings of these initiatives gradually revealed that aerosols (microscopic airborne particles or droplets) significantly affect cloud properties and lifetime. Thus, the aerosols became the Gordian knot of the climate studies and the emphasis shifted to documenting the properties, mapping the distributions, and estimating the radiative forcing of various types of aerosols.

Natural and anthropogenic aerosols affect the climate in two distinct ways: directly via increasing the planetary albedo, and indirectly via modifying cloud optical properties and lifetime (Charlson et al., 1992).

The mechanisms of direct forcing are well understood. Aerosol particles either absorb or reflect the shortwave solar radiation back to space. In most cases the reflection cools the Earth-atmosphere system as less heat reaches the planet's surface (Coakley and Cess, 1983, 1985; Charlson et al., 1987, 1991; Kiehl and Briegleb, 1993; Chuang et al., 1997; Tegen et al., 2000). In other cases, the absorption adds to the warming by greenhouse gases (Jacobson, 2001b).

The mechanisms of indirect forcing relate to aerosol action as cloud condensation nuclei (CCN) on which water vapors condense and form cloud droplets (Twomey, 1991; Baker, 1997; Taylor et al., 2000). Aerosols exert two distinct indirect effects on clouds. As aerosol concentration increases, the number of CCN increases too, leading to the formation of more cloud droplets (Chuang and Penner, 1995; Menon and Saxena, 1998; Lohmann et al., 1999). The presence of more cloud droplets provides more surface for reflecting the incoming sunlight. The clouds become brighter and their albedo increases (Stephens, 1978b; Coakley et al., 1987; Slingo, 1990; Chuang et al., 1997). This is referred to as the first indirect effect. The second indirect effect of the aerosols refers to the inhibition of droplets' growth in the presence of more cloud droplets, delaying formation of rain drops (Ferek et al., 2000). In this way cloud lifetime is prolonged; the cloud eventually evaporates without producing rain (Andreae, 1995). As a result, cloud coverage and water vapor distribution in the atmosphere are perturbed and with that their climate forcing.

The basis for the direct and indirect effects of aerosols on climate is atmospheric chemistry. Aerosol particles provide surface and volume for heterogeneous chemical reactions (Andreae and Crutzen, 1997), which involve reagents in different phase states (gaseous, liquid, or solid). Consequences of these reactions are changes in the state, chemical composition, and concentration of the atmospheric constituents in a manner and to a degree sufficient to alter the radiative properties of the atmosphere.

Uncertainties in assessing the direct and indirect aerosol forcing come from the intricate interactions between different types of aerosols (O'Dowd et al., 1997; Li-Jones and Prospero, 1998), incomplete knowledge of the properties and distribution of specific aerosol types (IPCC, 2001), and inadequate understanding of important chemical reactions in the atmosphere (Andreae and Crutzen, 1997; Keene et al., 1998).

The inclusion of aerosol radiative forcing in climate models significantly improves model results (IPCC, 2001). The effects of anthropogenic aerosols, such as sulfate aerosols, biomass-burning aerosols, fossil fuel black carbon (soot), and organic carbon aerosols are of particular importance since collectively these man-made aerosols provide a negative forcing (cooling) as large in magnitude as the warming from the greenhouse gases (Andreae, 1995; IPCC, 2001). Aerosol radiative forcing is defined as the total radiative effect due to all types of aerosols minus the radiative effect due to background aerosols (Bates et al., 1998b). Thus, to assess correctly the radiative forcing of anthropogenic aerosols, an accurate baseline, reflecting the condition of a clean atmosphere affected only by natural background aerosols, is required (Bates et al., 1998a, 1998b; Whittlestone et al., 1998).

Ocean-produced sea-salt aerosols are the most numerous naturally emitted aerosols. In clean marine air, their physical, chemical, and optical properties control the direct and indirect radiative effects of the total aerosol load. In polluted air, sea-salt aerosol chemistry affects the concentration and distribution of anthropogenic aerosols.

Thus, the characteristics and effects of sea-salt aerosols must be correctly parameterized and simulated in climate models.

The generation of sea-salt aerosols by the ocean is the first of many processes included in climate models. A reliable generation function for sea-salt aerosols exists, but it does not explain completely variations in atmospheric sea-salt concentration and distribution. In addition to the two major dependencies on wind speed and particle size, sea-salt aerosol generation is affected by various meteorological and environmental factors, which are not accounted for in the currently available sea-salt generation function. These additional factors could be included in the sea-salt generation function through the whitecap coverage, W, which represents the ocean area actively producing sea-salt aerosols. The existing database of W measurements does not cover a wide range of meteorological and environmental variables and is not adequate for this purpose. To compile a comprehensive database of whitecap measurements under various conditions, a new method for retrieving W on a global scale should be developed.

The importance of sea-salt aerosols for the radiative forcing of the climate system motivates this research. Sea-salt aerosols and their progenitors, whitecaps, are the subjects of this investigation. The main goals of this study are:
1) Development of a method for estimating whitecap coverage globally; 2) Compilation of a whitecap coverage database; 3) Parameterization of water temperature and salinity effects on whitecap coverage; 4) Modification of the existing generation function of sea-salt aerosols; 5) Evaluation of the contribution of sea-salt aerosols to various climate processes.

The current work is divided into six chapters. **Chapter 2** reviews what is known about sea-salt aerosols. First, their formation (§2.1) and properties (§2.2) are summarized. A list of processes involving sea-salt aerosols delineates their role in the climate system (§2.3). The generation function of sea-salt aerosols is introduced in §2.4, and its current status quo is analyzed. A brief account of *in situ* measurements of whitecaps establishes the necessity to develop a method retrieving whitecap coverage on a global scale and identifies the remotely sensed variable appropriate for this purpose (§2.5). On the basis of this review, the scientific objectives of the dissertation research are formulated in §2.6. **Chapter 3** describes the development of a new method for estimating whitecap coverage on a global scale. The concept and implementation of the method are explained in §§3.1 and 3.2. Method results and validation are reported in §§3.3 and 3.4. Possible improvements of the method are suggested in §3.5. **Chapter 4** describes the organization of the whitecap coverage database (§4.1), the analysis and implications of global whitecap coverage (§4.2), and the search of appropriate methods and procedures to parameterize the effects of additional factors on whitecap coverage (§4.3). **Chapter 5** documents the modification of the current sea-salt generation function (§5.1), the results on sea-salt aerosol production (§5.2), and their implications for climate (§5.3). **Chapter 6** presents the conclusions of this research and suggestions for future work.