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Real-time single particle monitoring of a relative increase in marine aerosol concentration during winter rainstorms

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Abstract. During the months of December 1996 and January 1997, aerosol characterization was performed in real-time at Riverside, CA, an inland location approximately 100 km east of the Los Angeles coast. Analysis was performed on a single particle basis with measurements of both particle size and composition. Data were collected throughout the course of five rainstorms. Single particle mass spectra demonstrate that obvious composition changes occur in the aerosol systems as the marine air mass is transported inland. While precipitation scavenging of aerosols was observed, there was a significant relative increase of sea salt particles in the non-scavenged aerosol. Compositionally-resolved particle size distributions also demonstrate real-time changes that occur in the atmospheric aerosol sample.

Introduction

Oceans are a significant natural source of both primary and secondary atmospheric aerosols [Fitzgerald, 1991; O'Dowd *et al.*, 1997]. Primary marine particles are formed by mechanical disturbances of the ocean surface and are mainly composed of sea salt [Exton *et al.*, 1985; Fairall *et al.*, 1983; Smith *et al.*, 1993; Wu *et al.*, 1984]. Secondary marine aerosols are formed through gas-to-particle conversion processes and are composed of mainly organic and sulfur compounds [Anderson *et al.*, 1991; Kriedenweis *et al.*, 1991].

Several source mechanisms that generate sea salt aerosols have been identified by both field and laboratory experimentation. The primary mechanism for sea salt particle production is the bursting of bubbles in whitecaps, resulting in the formation of either film drops or jet drops [Afeti and Resch, 1990; Resch *et al.*, 1991]. An increase in sea salt particle production is also observed with greater wind velocities due to an increase in total whitecap area [Monahan *et al.*, 1983; O'Dowd and Smith, 1993; Smith *et al.*, 1989; Woodcock, 1953]. Another means of sea salt aerosol production is rain impacting on the ocean surface [Marks, 1990; Woodcock, 1950]. During storm conditions, both increased wind velocity and raindrops impacting the ocean surface contribute to an increased production of sea salt aerosols.

This letter discusses measurement of a relative increase in the atmospheric concentration of sea salt particles during winter rainstorms. This increase of sea salt particles is observed experimentally at an inland location by detecting the unique chemical signatures of single sea salt particles. In contrast to filter collection methods of atmospheric aerosols which require months for analyzing chemical information, this letter reports on chemical data acquired in real-time.

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Experimental Methods

Data are obtained in real-time using aerosol time-of-flight mass spectrometry (ATOFMS) [Gard *et al.*, 1997; Liu *et al.*, 1997; Noble and Prather, 1996; 1997; Silva and Prather, 1997] which measures the size and composition of individual particles in polydisperse aerosol systems by coupling time-of-flight aerosol beam spectrometry (TOFABS) [Dahneke, 1978] with time-of-flight mass spectrometry (TOFMS). Although several real-time single particle mass spectrometry (RTSPMS) methods exist [for a review see Johnston and Wexler, 1995], ATOFMS is currently the only one which provides precise particle size and chemical composition for particles in polydisperse samples [Salt *et al.*, 1996].

Particle aerodynamic diameter is determined empirically using a calibration curve created using monodisperse ammonium sulfate particles over the size range of 0.1-10 μm in diameter [Noble and Prather, 1996].

Single particle data were collected during the months of December 1996 and January 1997 at the University of California, Riverside campus, which is approximately 60 km inland to the northeast and 100 km inland to the east of the Pacific Ocean. Over this period, particles were analyzed nearly continuously during five rainstorms with the ATOFMS instrument sampling at a height of 8 m above ground level (*i.e.* below-cloud sampling). The rainstorms of interest occurred in Riverside, California on 6-7, 9-12 and 27 December 1996 and 15 and 20 January 1997.

Results and Discussion

Typically, aerosol particles at Riverside, CA consist mainly of some combination of sea salt, soil and both biogenic and anthropogenic organic particles [Noble and Prather, 1996, 1997]. Also, evidence of heterogeneous reactions is often indicated by the presence of ammonium, sulfate and nitrate ions in the particles.

Prior to most of these rainstorms in Riverside, the majority of particles observed contained organic compounds, with only a relatively low percentage of soil derived particles and sea salt particles. Common mass-to-charge (m/z) peaks found in organic mass spectra of simple organic particles include hydrocarbon envelopes extending out to $^{60}(\text{C}_5^+)$ or $^{72}(\text{C}_6^+)$ in positive ion spectra. Negative ion spectra acquired during the same sampling periods indicate the presence of both sulfur and nitrogen oxides in many of the particles. See Noble and Prather [1996, 1997] for examples of organic particle mass spectra.

At the onset of the rainstorms, below-cloud precipitation scavenging occurs, resulting in an overall decrease in the aerosol concentration. Simultaneously, an increase in the relative concentration due to sea salt particles is observed in Riverside as the coastal air mass is transported inland. This is represented by a relative increase in single sea salt particle mass spectra, examples of which are shown in Figure 1. These spectra are typical of