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Seasonal cycle and composition of background fine particles along the west coast of the US

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Abstract

We used aerosol data from 4 sites along the west coast of the U.S. to evaluate the role of transport, seasonal pattern, chemical composition and possible trends in the marine background aerosol for the Pacific Northwest. For the Crater Lake samples, the data have been segregated using 10 day back isentropic trajectories to evaluate the role of transport. Our analysis of the segregated data indicates that the trajectories can successfully separate "locally influenced" from "marine background" aerosol, but are not able to identify a significant difference in the mean concentrations during marine vs Asian transport pathways.

The background marine aerosol has an annual mean and median concentrations of 2.0 and 1.5 $\mu g\,m^{-3}$, respectively, for particles less than 2.5 μm diameter. There is a seasonal pattern in all components of the aerosol mass, with a summer maximum and winter minimum. This pattern is most likely due to the strong seasonal pattern in precipitation, which peaks in winter, combined with enhanced sources in summer. The Crater Lake marine aerosol composition is dominated by organics (~40% by mass), with smaller contributions from sulfates, mineral dust and elemental carbon. Analysis of the background marine aerosol found no apparent trend since 1988. This is in contrast to results reported by Prospero et al. (J. Geophys. Res. 108 (2003) 4019) for nss-SO_4^{2-} samples from Midway Island in the North Pacific. Comparison of the mean concentrations for each site shows that the Midway samples are significantly more influenced by Asian industrial sources of sulfur, compared to Crater Lake, which implies a substantial loss of nss-SO_4^{2-} from Asian sources that occurs during transit across the Pacific to Crater Lake due to precipitation scavenging.

Keywords: Aerosols; Asian pollutants; Long-range transport

1. Introduction

Aerosols are produced by a variety of processes, both natural and anthropogenic. Since it is known that fine aerosols (diameter less than 2.5 μm) have significant health, climate, and visibility effects it is important to understand the sources and sinks of these particles. In the United States, the Environmental Protection Agency (EPA) is developing rules so as to achieve natural visibility conditions in U.S. National Parks by the mid-2lst century. This requires that we quantify the natural aerosol concentration and composition (U.S. EPA, 2001). In densely populated regions, most fine particle mass comes from anthropogenic sources, e.g., fossil fuel combustion or other industrial sources. But natural sources and long-range transport can also make important contributions under some circumstances.

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Gases and aerosols can be transported long distances. In the Pacific, Duce et al. (1980) and Shaw (1980) have shown that Asian desert dust can be transported to the mid-Pacific. A number of researchers have identified episodes of trans-Pacific transport of pollutants for a variety of gas and aerosols compounds (Andreae et al., 1988; Parrish et al., 1992; Jaffe et al., 1999, 2003a). A large episode of intercontinental transport of mineral dust took place in April 1998, when a major dust storm took place in northern China (Husar et al., 2001). The dust was observed on satellite imagery, crossed the Pacific in the free troposphere in ~5 days and was brought to surface sites in North America by large-scale subsidence and orographic effects (McKendry et al., 2001). A second, even larger, episode of trans-Pacific dust transport took place in April 2001 (Thulasiraman et al., 2002). In this event, Asian dust (PM10) concentrations reached 30–40 $\mu g\,m^{-3}$ at a large number of rural sites in the U.S. and contributed to even larger concentrations at some urban locations (Jaffe et al., 2003b). Surprisingly, the dust was seen at similar concentrations in the west and southeastern U.S. However, these large dust events appear to be relatively rare. Jaffe et al. (2003b) were able to identify only 2 large events in 15 years of aerosol observations.

While the large dust events are not common, we now know that trans-Pacific transport of gases and aerosols from Eurasia to North America is relatively common. Numerous publications have identified this transport in spring (e.g., Parrish et al., 1992; Kotchenruther et al., 2001; Jaffe et al., 2003a; Price et al., 2003). VanCuren and Cahill (2002) examined data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network and found that Asian mineral dust can be seen at all of the western U.S. sites throughout the year, with a broad maximum between March and October. They calculated that Asian dust contributes a monthly average of approximately 0.5 $\mu g\,m^{-3}$ to the PM2.5 mass during the peak months at elevated sites in the western U.S. (above about 400 m). This is an important result in that it shows that trans-Pacific transport occurs all year, not just in spring when most previous observations have been made. Jaffe et al. (2003a) identified numerous trans-Pacific episodes using aircraft, satellite and ground-based data from Washington, Oregon and California. This includes several episodes containing varying degrees of dust, sulfate and organic aerosol that were identified in IMPROVE data from Crater Lake, Oregon.

In the past two decades emissions of NO_x and SO_2 have risen by 4–6% from East Asia per year (Akimoto and Narita, 1994; Streets et al., 2001). In addition, aerosol optical depth over China has also increased over the past several decades (Luo et al., 2001). Prospero et al. (2003) have reported that the anthropogenic component of aerosol SO_4^{2-} at Midway Island (roughly half of the total SO_4^{2-}) increased significantly between 1981 and the mid-1990s due to increasing Asian emissions. At present it is not clear if this increase extends to the continental U.S.

Natural sources are also important contributors to fine particle concentrations. This can include non-sea salt sulfate, from marine biogenic emissions of dimethyl sulfide (Bonsang et al., 1980) and organic aerosols (Novakov and Penner, 1993). Sea salt aerosol contributes to the PM2.5 mass, but most of its contribution will be in larger particle sizes (Murphy et al., 1998; Anderson et al., 1999). A review of marine aerosol data by Quinn et al. (2000) found that a substantial fraction of sub-micron marine aerosol mass is not accounted for by measured sulfate and other inorganic ions. For example in the Pacific, between 20° and 60°N latitude, 29–45% of the sub-micron aerosol mass was not accounted for by sulfate, other inorganic ions, or associated water. The authors suggest that a large part of this deficit could be due to elemental and/or organic carbon, although there is little data from remote Pacific locations to evaluate this hypothesis.

In this paper we report on an analysis of data from Crater Lake Oregon, one of the cleanest sites in the IMPROVE network. The focus of our analysis is to understand the chemical composition of fine particles in background air entering the western U.S. from the Pacific. By "background" we mean aerosols in regions not affected by anthropogenic sources within the past 3 days. In this analysis, we aim to address the following questions: What are the concentrations and seasonal cycle of the PM2.5 aerosol at Crater Lake? Can we use meteorological data (trajectories) to separate the Crater Lake data into marine and continental components? Can back trajectories be used to separate the Crater Lake data into those samples most strongly influenced by Asian sources? What is the chemical composition for the Crater Lake aerosol and how does this differ for the marine and continental samples? Is there evidence of changing background aerosol concentrations at Crater Lake, associated with changing sources?

2. Methods

The IMPROVE network consists of more than 100 sites, primarily in U.S. National Parks (Malm et al., 1994; Eldred and Cahill, 1994; Eldred et al., 1997). At each site, aerosols are collected on several filters in two size bins: less than 2.5 μm aerodynamic diameter and between 2.5 and 10 μm aerodynamic diameter at ambient humidity. A fairly complete chemical analysis is made on the fine particle samples whereas only mass is measured on the coarse particle fraction. In this paper we will use only the fine particle data. Samples are collected for 24 h (midnight to midnight, local time). For