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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 most of the data record, samples were taken two days per week, but this has recently been changed so that currently a sample is now taken every third day. No measurements are made on the other days. The fine particle samples are analyzed for total mass as well as a wide array of chemical species including major ions, metals, organic and elemental carbon. Based on the recommendation in the IMPROVE data guide (UCD, 1995) we use the X-ray fluorescence sulfur data, rather than the ion chromatography sulfate data. Below detection limit data are replaced with half the detection limit. Non-seasalt sulfur (nss-S) is calculated based on the S/Na ratio found in seasalt (Harte, 1988). Several composite variables have been calculated per the recommendations in the IMPROVE data guide (UCD, 1995), including "Soil" ($2.20{*}Al + 2.49{*}Si + 1.63{*}Ca + 2.42{*}Fe + 1.94{*}Ti$) and "Organic Mass by Carbon (OMC)" (total organic $C{*}1.4$). Other composite variables are defined in the UCD 1995 report. For this report, we will use the term total elemental carbon, which is also called "light absorbing carbon". IMPROVE data are available on-line at: http://www2.nature.nps.gov.

In addition, we used aerosol data from the Cheeka Peak Observatory (Jaffe et al., 2001), which is located directly adjacent to the Washington state coastline. Samples were collected using two sequential dichotomous samplers (Model 2025, Rupprecht and Patashnick, Albany N.Y.), which collected two 24 h samples daily for coarse (2.5–10 μm) and fine (<2.5 μm) aerosol mass and chemical composition. For aerosol mass, Teflon filters were weighed before and after sampling at controlled temperature and humidity, in accordance with standard EPA procedures.

Table 1 gives the coordinates for each site, a summary of key aerosol measurements and nearby population for the three sites. Fig. 1 shows a map giving the locations of the 4 sites. What is apparent from Table 1 is that while none of these sites are very polluted, the Cheeka Peak site has the lowest aerosol concentration of these four sites and is in one of the least populated regions of the U.S. west coast. Crater Lake is close to Cheeka Peak, both in population and aerosol concentrations, however, it is inland from the coast by 180 km, so inevitably, these samples pick up some degree of local pollution. Unlike Cheeka Peak, however, Crater Lake has an IMPROVE aerosol record that extends back to 1988.

The Crater Lake N.P. sampling site is located at the park headquarters, on a road that is open year round to the public, so while the site is generally quite clean, one of the lowest in the entire IMPROVE network, some anomalies are possible. Local meteorological data for the site are not available for most of the period of the IMPROVE aerosol record, so for this reason we used atmospheric back-trajectories to segregate the dataset (described below). The Crater Lake sampling site is located 200 m below and to the south of the crater that

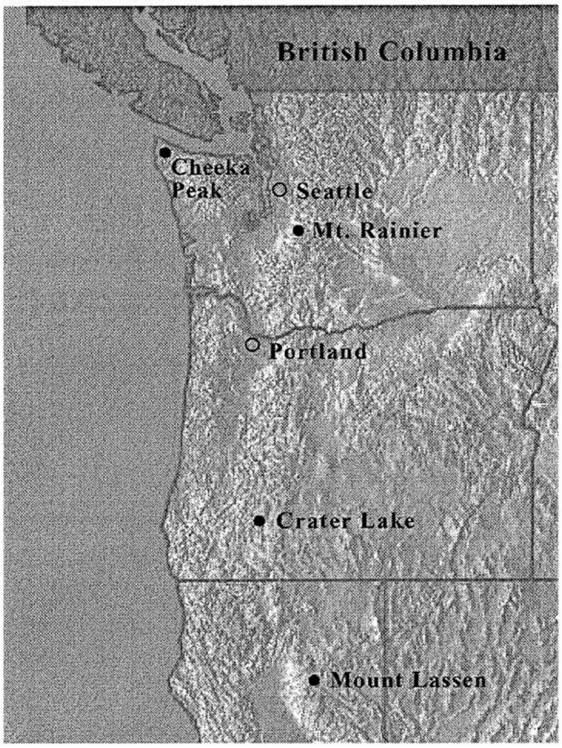

Fig. 1. Map of the Pacific Northwest showing the 4 sampling locations.

Table 1
Geographic and fine particle data for 4 sites

Site	Coordinates, elevation	Fine mass mean/median ($\mu g\,m^{-3}$)	Fine S mean/median ($\mu g\,m^{-3}$)	Nearby population in millions
Crater Lake N.P.	42.88 N 122.13 W, 1982 m	2.65/1.91	0.082/0.063	0.3
Mt. Rainier N.P.	46.75 N 122.12 W, 436 m	3.91/3.18	0.28/0.21	2-3
Mt. Lassen N.P.	40.53 N 121.57 W, 1799 m	3.59/2.72	0.15/0.13	0.7
Cheeka Peak	48.30 N 124.69 W, 480 m	2.04/1.57	No data	0.2

Data from the three IMPROVE sites (Crater Lake, Mt. Rainier and Mt. Lassen) are for the period 1999–2001. Data for Cheeka Peak is from March 2001–May 2002.

forms Crater Lake. The local wind directions tend to follow a shallow, local valley and the wind speeds are generally lower than winds at an equivalent height in the free troposphere, based on radiosonde data from Medford, Oregon. Therefore, we believe the Crater Lake IMPROVE site samples a mix of boundary layer and free tropospheric air. VanCuren and Cahill (2002) found that for both the Mt. Lassen and Crater Lake sites, there was much greater influence from Asian dust compared with west coast sea-level sites.

Ten-day Isentropic back-trajectories were calculated twice daily (at 0 and 12 UTC) for Crater Lake using data from the European Centre for Medium range Weather Forecasts (ECMWF) and the methods described by Harris and Kahl (1994). While our previous work with isentropic trajectories has given us much useful information regarding long range transport (e.g., Jaffe et al., 1999, 2003a), segregation by trajectories does not always give the expected result (e.g., Jaffe et al., 2001). There are a number of possible sources of error associated with isentropic trajectories, including low resolution meteorological data, assumption of dry adiabatic flow and/or sub-grid scale vertical motions. Possible trajectory errors have been discussed in several publications (e.g., Kahl, 1996; Stohl, 1998).

Since a long-term record of local meteorological data is not available for the Crater Lake site, we used the isentropic trajectories to segregate the dataset. While most trajectories arriving to Crater Lake come from the Pacific marine environment, we assumed that the longer an airmass spent over North America before reaching Crater Lake, the greater was the probability that this airmass picked up North American emissions and lost its marine character. So each trajectory was classified as "local", "marine" or "Asian". A trajectory was considered local if it spent more than 24 h east of longitude 125°W, in other words over the continental U.S. The 24 h period was necessary since Crater Lake is about 180 km inland from the Pacific coast. A trajectory was classified as Asian if it crossed over the region defined by 0–50°N and 100–150°E within 10 days. While we have used this method previously with reasonable success (e.g., Jaffe et al., 2001, 2003c) it should be pointed out that the separation between these categories is not perfect. In other words it is probable that a sample classified as "marine" will still include some local contribution and one identified as "local" will still have some Asian contribution.

3. Results

For this analysis, we used data for the period March 1988 (beginning of IMPROVE measurements) through October 2001. Table 2 shows the results of the trajectory classification for this time period (2346 trajectories). Winter is defined as the months of December, January and February and the other seasons are defined accordingly. For the entire year, 57% of the Crater Lake trajectories are arriving within 24 h from the marine environment.

Since each filter sample spans a 24 h period, it is possible that more than one type of airmass has been sampled. To account for this possibility, we categorize each filter sample based on the two trajectories that were calculated for that period. This leads to four possible classifications for each filter sample, as shown in Table 3.

For this study, our working hypothesis is that the "local" samples will most clearly reflect western U.S. emission sources and "Asian" samples will most clearly reflect Asian sources. To evaluate this hypothesis, we segregated the filter data based on the trajectory classification. Table 4 shows the mean, median, standard deviation and count (n) for each sample type, by season, for PM2.5 mass.

For all seasons, the local samples show the highest concentrations of fine particle mass, compared to the other categories. We used a t-test to evaluate whether the difference is statistically significant. However, since the data are not normally distributed, we did the t-test on both the original, and log-transformed data. Grouping the data by season and using the t-tests, we find that this local-marine difference is statistically significant at a >99% confidence level for all seasons. We conclude

Table 2
Classification of Crater Lake 10 day back-trajectories by season (%)

	Winter	Spring	Summer	Fall
Local	42.2	37.7	46.0	44.3
Marine	33.5	42.2	46.7	42.7
Asian	24.3	20.2	7.2	13.0

Table 3
Classification of filter samples based on two trajectories in 24 h period

Category	Description	% of all days
Local	Both trajectories local	32.8
Mixed	1 trajectory is marine or Asian the other is local	19.6
Marine	Both trajectories are marine (may include 1 Asian)	42.0
Asian	Both trajectories are classified as Asian	5.7

Table 4
Summary of PM2.5 mass concentration data (μg m^{-3}) by sample type and season

	Winter	Spring	Summer	Fall
Local (mean/median)	1.72/1.50	4.11/3.25	4.96/4.50	4.13/3.76
S.D. (*N*)	0.96 (80)	3.02 (80)	2.59 (102)	2.42 (85)
Mixed (mean/median)	1.33/0.99	2.74/2.02	4.56/3.82	2.76/2.18
S.D. (*N*)	.98 (38)	1.83 (45)	2.59 (79)	1.77 (42)
Marine (mean/median)	1.22/0.99	2.14/1.62	2.98/2.33	1.72/1.52
S.D. (*N*)	0.89 (88)	1.58 (111)	2.55 (112)	.94 (102)
Asian (mean/median)	1.01/0.83	2.57/2.38	2.73/2.54	2.28/1.54
S.D. (*N*)	.59 (25)	1.80 (19)	.90 (6)	2.06 (9)

from this that the trajectories are capable of segregating local from marine airmasses. Comparing the marine and Asian categories we find that there is not a statistically significant difference between these for any season. This indicates that the isentropic trajectories, by themselves, are not adequate to segregate out those samples with an obvious Asian signal, when compared with the marine samples. Very likely those samples classified as both marine and Asian, have some contribution to the aerosol mass from Asian sources. Therefore, for the remainder of our analysis, we will group the marine and Asian samples into a single category called "Combined Marine/Asian" (CMA). As expected, the "mixed" samples have mass concentrations that are between the marine and local values.

The Crater Lake CMA samples have an annual mean and median fine mass concentration of 2.05 and 1.54 μg m^{-3}, respectively. These values are very close to the values for Cheeka Peak (see Table 1). So while it is possible that the Crater Lake samples have somewhat higher mineral dust concentrations, due to its higher elevation (VanCuren and Cahill, 2002), the similarity of concentrations argues that any local contribution in these "marine" samples must be quite small. Thus our trajectory classification method appears to be largely successful in identifying marine samples, with very little local contribution to the aerosol mass.

Fig. 2a and b show the fine particle and total organic carbon concentrations by month for the three sample classifications; local, mixed and CMA. The maximum concentrations occur in late summer, which is true regardless of the trajectory regime. For fine particle mass there is also a secondary peak in April. Fig. 2a also shows the monthly average precipitation, which is discussed in a later section of this paper.

Having used the trajectories to segregate the dataset, we wish to examine the chemical composition within each sample type. Table 5 shows the percentage of the total fine mass that is accounted for by each aerosol type for the local and the CMA categories. In general, the totals sum to less than 100%, presumably due to other unmeasured compounds.

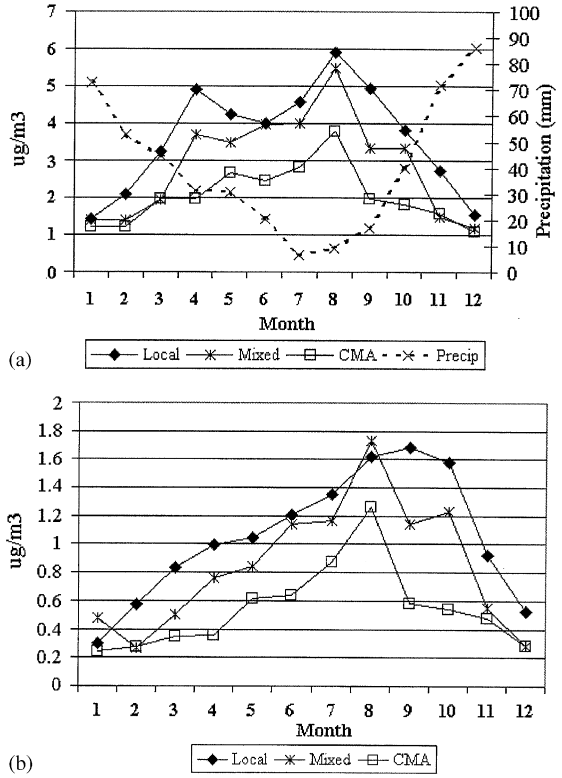

Fig. 2. Mean concentration vs month for local, mixed and CMA sample types as defined in text for (a) fine particle mass (μg m^{-3}) and (b) organic carbon (μgC m^{-3}). Fig. 2a also shows the climatological monthly mean precipitation from Medford Oregon.

In all seasons and categories, organic compounds dominate the fine mass. While the seasonal and category differences are small, there is some useful information. The organic fraction is slightly higher in summer and fall. Sulfur and soil mass fractions have modest peaks in spring, while elemental carbon and seasalt reach their maximum mass fractions in winter. The higher average seasalt fraction in the CMA samples, compared with the

Table 5
Mean percent of total fine particle mass for local and CMA samples

	Organic mass by carbon	Sulfur (as $(NH_4)_2SO_4$)	Soil	Total elemental carbon	NaCl	NO_3^-	Total
Local class							
Winter	39.6	20.5	11.3	14.8	2.3	4.3	92.8
Spring	36.2	21.0	18.1	5.5	2.1	4.6	87.5
Summer	40.6	17.6	11.9	5.2	2.5	2.2	80.0
Fall	51.1	15.3	13.0	7.4	2.1	2.7	91.6
CMA class							
Winter	39.7	16.6	11.4	19.1	10.2	3.7	100.7
Spring	34.2	21.2	18.4	7.3	6.0	4.0	91.1
Summer	42.9	18.6	14.3	5.9	4.5	3.2	89.4
Fall	42.4	19.7	14.8	8.5	5.0	3.3	93.7

local samples, adds further support to our trajectory classification used to segregate these samples. For seasalt, the much higher wind speeds in winter are most likely responsible for the higher seasalt fraction. For elemental carbon, it should be noted that there were 11 samples in the database where total elemental carbon concentrations (TEC) were more than 50% of the total PM2.5 mass (out of 1023 samples with TEC data), in a few cases TEC was more than 80% of the fine particle mass. We considered excluding these 11 samples from the averages in Table 5, however, these changed the percentages and overall pattern very little. The higher TEC fraction in winter, may reflect the much higher prevalence of precipitation in winter, and the relatively lower precipitation scavenging compared to other aerosol components.

3.1. *Trend in CMA and local/continental aerosol*

We evaluated the local and CMA category samples for trends in fine particle mass and major chemical components. Because of the large variability in aerosol concentrations, it is necessary to do monthly or seasonal averaging before looking for any possible trends. We used the seasonal medians vs year in a linear regression model to examine possible trends. Table 6 shows the results of this analysis and Fig. 3 shows seasonal median concentrations vs year for two of the most significant relationships.

The most striking result in Table 6 (and Fig. 3) is the decline in fine mass during spring for the local samples. Over the 14 year record, the median spring PM2.5 mass has declined by about 4 $\mu g\,m^{-3}$ or 8% $year^{-1}$. The mean spring value has also declined about the same amount. Organic carbon, mineral dust and sulfur also decline during spring in the local samples. For CMA samples, none of the trends are statistically significant. Data from the Mt. Rainier IMPROVE site also shows a statistically significant decrease in fine mass concentration between 1988 and 2003. Regression of the Mt. Rainier annual median fine mass concentration vs year yields a line with a slope of $-0.19\,\mu g\,m^{-3}$ year and an R^2 of 0.76. Since the Mt. Rainier site is much more strongly influenced by regional aerosol sources, compared with Crater Lake, this explains why the Crater Lake data only show a trend in the local classified samples, and not the CMA samples. The decline in the Mt Rainier fine mass concentration and the Crater Lake local samples are consistent with changes in particulate concentrations for urban areas in the western U.S., as reported by the EPA. For example in EPA regions IX and X, PM10 concentrations have declined by an average of 10% and 31%, respectively, between 1992 and 2001 for monitoring locations within these regions (U.S. EPA, 2003). So while the trend in local concentration is not surprising, given the change in emissions within these regions, it is important to note that there is no evidence for changes in the background marine aerosol concentrations.

3.2. *Correlations*

Table 7 shows the correlation coefficient between the major aerosol components for local and CMA samples. With the large number of samples in each category (347 for local, 472 for CMA) all of these species correlations are significant at a confidence level of 99% or better. Not surprisingly, all of these components tend to rise and fall simultaneously. In general, the correlations are slightly better in the CMA samples, compared to the local ones. To some extent these correlations may reflect the similarity in seasonal cycles, however, the correlation coefficients generally increase if the correlations are calculated for one season. This indicates that the

Table 6
Trend analysis for fine mass and major components using seasonal medians for local and CMA samples

	PM2.5 mass		Organic C		Mineral dust		Non-seasalt sulfur	
	Slope	R^2	Slope	R^2	Slope	R^2	Slope	R^2
Local								
Winter	−.053	.22	**−.026**	**.34**	−.006	.11	−.002	.17
Spring	**−.289**	**.46**	−.063	.24	**−.038**	**.42**	**−.010**	**.40**
Summer	−.091	.07	**−.052**	**.42**	−.019	.11	−.004	.10
Fall	−.060	.05	+.010	.007	−.020	.18	−.005	.22
CMA								
Winter	−.011	.04	−.008	.18	−.003	.03	−.002	.14
Spring	−.006	.001	−.033	.13	−.007	.04	−.0001	.00
Summer	−.018	.007	−.023	.23	−.003	.03	−.003	.25
Fall	−.048	.12	+.004	.05	−.0002	.006	−.002	.10

Slopes are given in $\mu g\,m^{-3}\,year^{-1}$. Values in bold are statistically significant at a 95% confidence level.

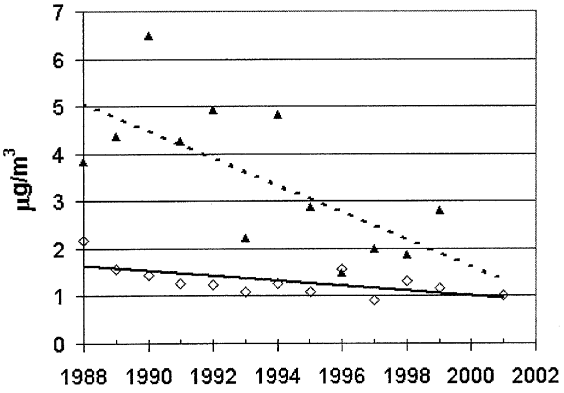

Fig. 3. Trend in median spring PM2.5 (solid triangles) and summer organic carbon (open diamonds) for two parameters in the local dataset. Parameters for the linear regressions are shown in Table 6.

Table 7
Top right side of table shows values correlation coefficient (R) for local samples and bottom left side of table shows values from CMA samples

	Organic carbon	Mineral dust	Non-seasalt sulfur	Nitrate
Organic carbon		.236	.416	.179
Mineral dust	.232		.371	.144
Non-seasalt sulfur	.340	.483		.358
Nitrate	.288	.281	.406	

day-to-day variations in these components tend to track each other which is due, at least in part, to precipitation removal.

4. Discussion

This analysis indicates that isentropic back-trajectories can be used to segregate out samples that have been influenced from nearby processes (emissions, wet deposition, etc.) from those that have not. At the same time, the similarity in concentrations, seasonal cycle and chemical composition indicates that this segregation is not perfect and that the Crater Lake "marine" aerosol and "local" aerosol are part of two overlapping distributions. Another way of thinking about this is that the CMA aerosol is the starting material for the local aerosol and so it should not come as a surprise that the two show similarities. Regarding the use of trajectories to characterize long-range transport, Table 4 demonstrates that trajectories alone are insufficient to identify sources that are 5–10 days upwind (e.g., Asia). In other words, for the Crater Lake samples, a day with Asian trajectories is no more likely to have enhanced concentrations than any other marine sample.

This analysis has revealed a number of patterns, including the summer maximum in background marine aerosol, the dominance of organic carbon in the fine aerosol mass and the absence of a positive trend in the CMA aerosol samples due to increasing upstream (Asian) emissions. Here we will try to develop plausible explanations of these results.

The summer maximum-winter minimum is present in both the local and CMA samples. Park et al. (2003) discuss this seasonal pattern for elemental and organic carbon in the IMPROVE samples and conclude that the seasonality is mostly due to the strong increase in continental sources during summer. While this is certainly correct, we also feel that the strong seasonality in precipitation in the Pacific generally and the Pacific Northwest in particular must also play a role. Long-term observations from Medford, Oregon (approximately 120 km from Crater Lake) give a monthly mean precipitation amount of 74 mm in January, compared with just 9 mm in August. Overall, 44% of the annual precipitation at Medford falls in winter, compared with 8% in summer. As shown in Fig. 2a, the monthly pattern of rainfall is inversely related to the PM2.5 concentrations.

It is possible that rainfall also explains some of the local-marine differences as well. This is possible since on-shore, marine flow is more likely to be associated with precipitation, compared to a continental/high pressure situation. We examined the relationship between trajectory type and daily precipitation using 3 years of data from Medford. During winter, days with CMA trajectories have, on average, about 3 times more rainfall than days with local trajectories. However in summer, this pattern is reversed. This likely reflects the fact that summer rainfall often occurs in relatively large amounts on a small number of days. Likely these days are associated with strong convective activity, which would not necessarily be associated with marine flow.

The dominance of organic compounds in the CMA background aerosol is consistent with the results reported by Quinn et al. (2000), who found that for samples from 20–60°N latitude an average of 29–45% of the sub-micron aerosol was not accounted for by sulfur, seasalt or other inorganic elements. For the Crater Lake CMA samples, we calculate an average organic + elemental carbon fraction of 40–60% of the PM2.5 mass. The difference could be due to the fact that Quinn et al. accounted for associated water, which was not considered in our analysis, and did not include mineral dust in their analyses, which makes up 15% of the Crater Lake mass, on average. In any case, it is clear that a large fraction of the CMA aerosol is due to organic carbon. Whether these compounds are principally natural or anthropogenic is not clear at this point.

Park et al. (2003) used a global model simulation to evaluate the sources of elemental and organic carbon to the U.S. From model runs with all U.S. sources turned off, they estimate that the background organic aerosol has an annual average of 0.43 $\mu g\,m^{-3}$ in the western U.S. This can be compared to our annual average from the CMA trajectories of 0.73 $\mu g\,m^{-3}$. More importantly, Park et al., conclude that this background organic aerosol is almost entirely due to Canadian and Mexican sources, and very little due to trans-Pacific sources. Our analysis, and that of VanCuren (2003), suggests otherwise. The location of Crater Lake and the selection of marine only trajectories would tend to minimize contributions from Canada and/or Mexico on this background aerosol. Also, recall that the aerosol concentrations for the Crater Lake "Asian" and "marine" classifications are indistinguishable, suggesting that these groups of samples are being impacted by the same sources. However, our analysis cannot distinguish between natural marine and trans-Pacific sources. Thus the background marine aerosol at Crater Lake is likely due to some combination of contributions from Eurasian anthropogenic sources, natural marine emissions, Eurasian vegetation and/or Eurasian biomass burning. VanCuren (2003) reaches a very similar conclusion.

The Crater Lake aerosol contains 11–18% mineral dust throughout the year. Given the numerous reports on long-range transport of Asian dust to North America (and specifically Crater Lake), we believe this dust is mostly from Asia. This is supported by the analysis of VanCuren and Cahill (2002) and VanCuren (2003). This translates into a spring–summer mean value of 0.3–0.4 $\mu g\,m^{-3}$ of Asian dust at Crater Lake (PM2.5 only) in the CMA samples, or 0.4–0.7 $\mu g\,m^{-3}$ if all samples are considered. For comparison, VanCuren and Cahill (2002) using a cluster analysis method, calculate that Crater Lake and other high elevation IMPROVE sites in the western U.S. receive an average of 0.4–0.6 $\mu g\,m^{-3}$ of Asian dust in spring and summer months. VanCuren (2003) extends this analysis to show that organics are nearly always associated with the Asian dust and make up a comparable, or an even larger fraction of the total fine mass. Our analysis suggests that the Asian dust component is greatest in the local samples, compared to CMA. Greater precipitation on days with CMA trajectories would explain this result.

The question arises as to why the Crater Lake CMA samples show essentially no trends, while data from Midway Island show a positive trend in aerosol sulfate, presumably due to increasing Asian emissions (Prospero et al., 2003). The most likely cause for this difference is that the springtime Midway Island samples have significantly more aerosol SO_4^{2-} compared to samples from Crater Lake. The mean springtime nss-SO_4^{2-} at Midway is 0.94 $\mu g\,m^{-3}$, compared with just 0.3 $\mu g\,m^{-3}$ at Crater Lake, however, it must be mentioned that the Midway samples are for total particulate, not just fine particles. However, since sulfur is mostly on sub-micron aerosols, this sampling difference is probably not important. Thus Midway receives a much larger amount of Asian pollution during spring, compared with Crater Lake, and is therefore much more likely to detect changes in the Asian emissions. The lower values at Crater Lake reflect the longer transit time to the Pacific Northwest, compared to Midway Island, and the much greater chance for precipitation scavenging during transit across the Pacific. This is in contrast to low solubility gaseous pollutants, such as carbon monoxide, which show much smaller concentration gradients across the Pacific.

In summary, we have used the IMPROVE aerosol record at Crater Lake, Oregon to evaluate the role of transport, seasonal patterns, chemical composition and possible trends in both local/continental and marine background aerosol for the Pacific Northwest. This analysis has revealed patterns that can be attributed to transport and relationship with precipitation. The most important findings from this work are the summer maximum in background marine aerosol, the large fraction of organic compounds that make up the background marine aerosol, the lack of any significant trends in the background concentrations, and the substantial loss and removal of nss-SO_4^{2-} from Asian sources that occurs during transit across the Pacific.

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