

Asian continental aerosol persistence above the marine boundary layer over the eastern North Pacific: Continuous aerosol measurements from Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2)

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[1] We report analyses of aerosols collected for the 2002 Intercontinental Transport and Chemical Transformation experiment (ITCT-2K2). Sampling was conducted 15 April to 25 May 2002. Data are from three sites: a sea level site at Trinidad Head, California, a coastal mountain site 1 km altitude (Trinity Alps), and an inland mountain site near 2 km altitude (Mount Lassen). Aerosols were continuously collected in eight size bins (0.09 to 10 micrometers aerodynamic diameter) using eight-stage rotating drum impactors. Samples were analyzed in 3-hour time steps by synchrotron x-ray fluorescence. We find the following. (1) Aerosol chemical composition at Trinidad Head was generally dominated by marine aerosols with varying minor contributions from local sources. Despite the presence of Asian continental aerosol above the marine boundary layer, significant concentrations of Asian aerosols were observed at sea level only during a strong frontal passage between 22 and 25 April. (2) At the elevated sites, aerosol elemental composition was predominantly Asian despite wide swings in concentration. Analysis of soil-forming elements shows that Asian continental dust and associated combustion products overwhelmed local-source aerosols through the first half of the sampling period; in the latter half, Asian aerosols present in the free troposphere were regularly delivered to the mountain sampling sites by nocturnal subsidence. (3) Asian aerosols in the lower free troposphere, although highly variable, were very persistent, not arriving only in discrete “transport events.” We conclude that throughout the experiment the aerosols in the lower free troposphere over the northeastern Pacific Ocean and western North America were dominated by continental outflow from Asia, with little marine or North American continental influence. Viewed in the context of previously published analyses of the long-term aerosol history for Mount Lassen that showed frequent, strong Asian influence throughout spring, summer, and fall, the Asian impact appears likely to be quasi-continuous for much of the year.

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1. Introduction

[2] Gaseous and aerosol combustion products have been observed to accompany Asian dust at several sites within the Pacific basin, including Hawaii, Eniwetok, Midway, and

others [Shaw, 1980; Duce *et al.*, 1980; Jaffe *et al.*, 1997; Perry *et al.*, 1999]. Limited purposeful monitoring on the Pacific coast of North America has detected trans-Pacific transport of gaseous Asian tracers and pollutants, including ozone, carbon monoxide, nitrogen species, organic compounds, and aerosols [LeRouley and Danielsen, 1990; Parrish *et al.*, 1992; Jaffe *et al.*, 1999; Tratt *et al.*, 2001; Murayama *et al.*, 2001; Uno *et al.*, 2001; Berntsen *et al.*, 1999; Husar *et al.*, 2000; Jaffe *et al.*, 2001].

[3] Analysis of long-term aerosol sampling data has shown Asian dust to be a frequent component of the lower free troposphere over much of North America [VanCuren and Cahill, 2002]. Analysis of a decade of data from Mount Lassen, CA, and Crater Lake, OR, has shown that the dominant aerosol mode over the western cordillera of North America is a “dusty Asian plume” consisting of carbona-

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ceous particles, soil, sulfates, and industrial trace elements, indicating frequent transport of natural and anthropogenic aerosols from Asia to North America [VanCuren, 2003]. These earlier aerosol studies relied on a decade of twice-weekly filter samples, and thus could not answer questions about the synoptic-scale dynamics of trans-Pacific transport, nor could they resolve the question of whether the transport was episodic or continuous. The data reported here shed new light on these questions.

2. Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2)

[4] Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) [<http://www.al.noaa.gov/ITCT/2k2/>] is a major research activity of the International Global Atmospheric Chemistry (IGAC) [<http://www.igac.noaa.gov/>] program addressing the tropospheric chemistry and transport of ozone, fine particles and other chemically active greenhouse compounds. The 2002 field study focused on transport across the North Pacific Ocean to North America, using a heavily instrumented primary ground site at Trinidad Head, CA supplemented by aircraft sampling over coastal North America and adjacent areas of the Pacific Ocean. During the ITCT 2K2 sampling period (mid-April through May, 2002) we conducted a continuous aerosol monitoring program using 8-Stage Davis Rotating Uniform Size-Cut Monitor (8-DRUM) impactors at Trinidad Head, CA, Trinity Alps, CA, and Lassen Volcanic National Park, CA, and fine particle (<2.5 micrometer) 3-stage DRUM samplers at Crater Lake National Park, OR, and White Mountain, CA. This first report covers only the data for Trinidad Head, Trinity Alps, and Mount Lassen.

3. Sampling Sites and Experimental Goals

[5] The ITCT 2K2 experimental plan used a heavily instrumented primary ground site on the Northern California coast at Trinidad Head, CA (Figure 1) supplemented by aircraft missions over the eastern Pacific Ocean. For this aerosol study the Trinidad Head site was supplemented with nearby mountain sites. The mountain monitoring was suggested by the results of published Asian dust analyses which showed that coastal sites in California may miss Asian transport events due to the strong, persistent marine inversion common along the west coast of North America [VanCuren and Cahill, 2002]. The intent of the multisite experiment was to use continuous data to explore three aspects of trans-Pacific aerosol transport:

[6] The first goal was evaluation of the temporal variability of Asian impacts in western North America. Previous aerosol analyses [VanCuren and Cahill, 2002; VanCuren, 2003] showed strong and regular Asian impacts in this region, but the intermittent sampling on which those reports were based could not support analyses of the frequency and duration of individual transport events, whereas continuous measurements permit direct observation of the temporal structure of trans-Pacific aerosol transport.

[7] The second goal was examination of the lower troposphere/marine boundary layer separation of trans-Pacific aerosol transport. Statistical analysis of intermittent sample data [VanCuren and Cahill, 2002] showed a strong

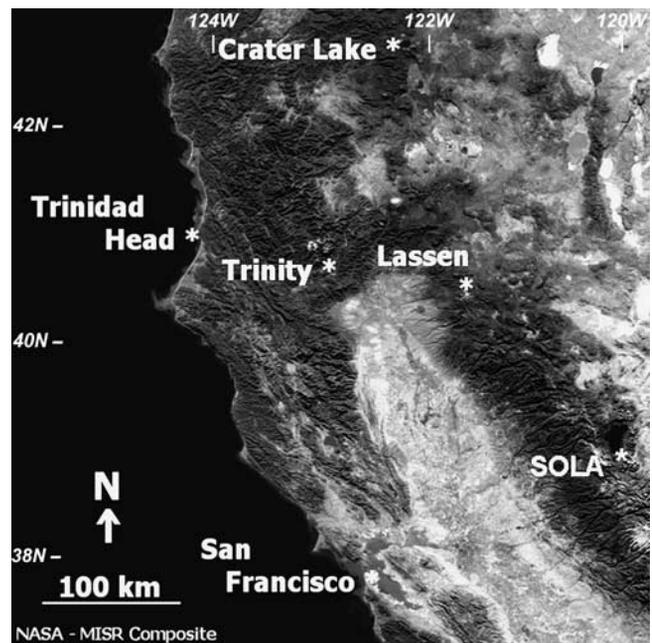


Figure 1. Location map showing ICTC 2K2 DRUM sampling sites at Trinidad Head, Trinity, and Lassen. Data previously collected at SOLA were used to characterize local North American soil dust. San Francisco and Crater Lake shown for reference only. See color version of this figure in the HTML.

vertical gradient of Asian impact along the Pacific coast of North America. Simultaneous monitoring at sea level and elevated sites in the present study permits direct examination of this vertical zonation of Asian aerosols, allowing validation (or refutation) of the impression given by published analysis of twice-weekly samples.

[8] The third goal was determination of the temporal pattern of mixing states (local vs. Asian) of lower free troposphere aerosols over the west coast of North America. Previous work [VanCuren, 2003] showed that the long-term composition of the aerosols at elevated sites in the study region is dominated by Asian materials. Continuous sampling for a period of several weeks provides a means to assess the temporal structure of the relative contributions of Asian and North American sources to the observed aerosol loading, and to determine whether the region's aerosols are generally a mixture of Asian and local materials, or if air masses associated with the Asian and North American aerosols remain distinct within a regime of alternating Asian and local influences.

[9] The mountain sampling sites themselves were chosen because they are part of the IMPROVE network (see <http://vista.cira.colostate.edu/improve/>; Cahill and Wakabayashi [1993]). IMPROVE sites offered power, space, and security to operate the 8-DRUM samplers unattended, as well as overlap with the IMPROVE long-term aerosol records.

[10] The ITCT 2K2 main ground site was at sea level on a rocky coastal promontory, Trinidad Head (41.05N, 124.15W). The IMPROVE Trinity Alps site (hereafter referred to as "Trinity") is located at Pettijohn Mountain in the coast ranges (40.78N, 122.81W), elevation 1007 m, about 100 km east-southeast of Trinidad Head. The

Table 1. Elemental Signal to Noise Ratios (S/N) (Concentration/Uncertainty) and Typical Source Associations for the 14 Elements Used in the Present Analyses

Element	Mean S/N	Mean S/N	Mean S/N	Source Association
Cl	9.7	7.7	4.7	sea salt, green biomass
Na	4.9	2.2	1.1	sea salt, soil, fly ash
Mg	3.9	3.2	2.3	sea salt, soil, fly ash
Ca	11.1	12.6	11.4	sea salt, soil, fly ash
Fe	11.4	13.5	12.7	soil, fly ash
Al	3.8	7.0	5.4	soil, fly ash
Ti	11.1	11.4	10.8	soil, fly ash
Si	8.6	10.5	9.2	soil, fly ash
K	11.7	12.4	10.7	soil, fly ash, biomass
P	9.4	7.6	7.2	soil, heavy oil, lubricants, biomass
Zn	10.2	9.7	9.4	soil, heavy oil, lubricants
V	4.7	2.7	2.6	soil, heavy oil
Ni	4.8	5.1	4.2	soil, heavy oil
S	13.0	12.1	11.2	sea salt, fossil fuel

Lassen Volcanic National Park site (hereafter referred to as “Lassen”) is located at Manzanita Lake on the north side of Mt. Lassen in the southern Cascade range (40.54N, 121.58W), elevation 1755 m, about 200 km east-southeast of Trinidad Head. These sites are marked on a satellite image of Oregon and northern California in Figure 1.

4. Sampling System

[11] The 8-Stage Rotating DRUM Impactor Sampler (8-DRUM) used in this study is a cascade impactor based on the basic design of *Lundgren* [1967], and evolved from the original DRUM impactor as described by *Raabe et al.* [1988]. The sampler as configured for the present work operated at 16.7 liters per minute. The samplers located at the Trinity and Trinidad Head sites were coupled to a 10 μm cutpoint impactor-type (“PM₁₀”) inlet; the sampler at the Lassen site used only a protective rain shield with an undefined (ca. 15 μm) primary cut. While the sampler for this work uses a slightly modified orifice design, impactor theory predicts similar efficiency for particle collection and robust size-cut determination as reported in the work of *Raabe et al.* [1988]. The aerosol sample for each stage is deposited onto a rotating drum faced with a removable greased Mylar impaction surface. As the drum rotates a continuous aerosol sample is laid down along the direction of rotation with density varying along the length of the Mylar strip in proportion to the aerosol collected as the drum rotates. By replacing the circular jets of the original DRUM with slits, the aerosol deposit is made uniform crosswise to the direction of rotation and the total deposit is spread over a known area per unit time [*Bench et al.*, 2002]. With the drums for all stages of the impactor geared together, coincident samples are collected on all eight stages. Analysis using a narrow X-ray beam scanned along each strip produces X-ray fluorescence elemental analyses with time resolution proportional to the ratio of drum surface speed divided by the beam width. For ITCT 2K2, the samplers were configured to provide a 42-day continuous record in 8 size bins (10–5, 5–2.5, 2.5–1.15, 1.15–0.75, 0.75–0.56, 0.56–0.34, 0.34–0.26, 0.26–0.09 micrometers aerodynamic diameter) analyzable in 3-hour time steps.

[12] Samples were analyzed by synchrotron X-ray fluorescence (S-XRF) [*Haller and Knochel*, 1996] using a broad

spectrum X-ray beam generated on beam line 10.3.1 at the Advanced Light Source (ALS) at E. O. Lawrence Laboratory in Berkeley, CA. The ALS S-XRF system is capable of detecting elements from Na to U. As an example of data recovery, Table 1 shows the mean signal-to-noise ratios for the elements used in the present analyses. Standard 8-DRUM units were operated at Trinidad Head, Trinity Alps, and Lassen. A second 8-stage DRUM operated at Trinidad Head was fitted with a humidity triggered heater designed to warm the sampler just enough to avoid fog condensation inside it; the Trinidad Head data presented here are from that sampler. Comparison of total sample from the heated and unheated samplers showed no significant differences between the two units for the elements discussed here [*Perry et al.*, 2004].

5. Data Analysis and Presentation

[13] We present three analyses of the ITCT 8-DRUM samples.

[14] First, a qualitative Principal Component Analysis (PCA) of the chemical composition variation at all three sites. The PCA provides an overview of the types of aerosols contributing to the total sampled at each site and a semiquantitative (correlation analysis) comparison of the relative strengths of the aerosol types.

[15] Second, the quantitative soil analysis uses a mixing model derived in part from previously reported analyses of IMPROVE network samples [*VanCuren*, 2003]. Regression analyses of soil and combustion species yields a rough estimate of the mixing state (Asian vs. local) of the aerosols.

[16] Finally, The Lassen comparison combines the size resolved ITCT data into a single size cut compatible with the historical IMPROVE data, then compares the general composition during the ITCT period with the long-term record.

5.1. Principal Component Analysis

[17] Our primary tool for inspecting DRUM samples is principal component analysis (PCA). This provides a picture of the gross composition and temporal variation of chemical composition in the samples, and permits graphic visualization of the chemical variation within the data sets, both temporally and across sampling sites. For this analysis, the raw elemental data were filtered based on their mean signal-to-noise (S/N) ratios and a reduced set of elements selected for their strong quantification across all three sites. This produced a data set of 14 elements (Cl, Na, Mg, Ca, Fe, Al, Ti, Si, K, P, Zn, V, Ni, S) in eight stages (i.e., 112 variables) for approximately 300 3-hour periods for each site (902 samples represented here). The signal-to-noise statistics for this suite of elements are presented in Table 1.

[18] This application of PCA follows the approach previously applied to the long-term IMPROVE filter-based data set from Lassen and Crater Lake (Figure 1) to detect the Asian component of the regional aerosol [*VanCuren*, 2003]. In this application the PCA results provide a dynamic reconstruction of the changing mixing state of aerosols from diverse origins over the time period represented by the continuous sampling.

[19] The conceptual basis for this analysis is the assumption that ambient aerosol samples are mixtures of material

Table 2. Explained Fractions of Total Variance by Principal Components (PC) Using 14 z -Transformed Elements (Top 120 of 112 PCs), and Correlation With X-Ray Mass (Sum of Reported Elemental Concentrations) by Site^a

PC	Explained Fraction	Cumulative Fraction	Trinidad Head	Trinity	Lassen
			Correlation to X-Ray Mass	Correlation To X-Ray Mass	Correlation To X-Ray Mass
1	0.299	0.299	-0.155	-0.955	-0.929
2	0.112	0.411	-0.674	-0.910	-0.496
3	0.065	0.476	0.471	0.059	0.549
4	0.055	0.532	-0.406	0.167	-0.866
5	0.044	0.575	0.134	0.114	0.373
6	0.029	0.605	0.260	-0.014	0.511
7	0.028	0.633	0.220	-0.117	-0.121
8	0.028	0.660	-0.382	-0.283	0.226
9	0.022	0.683	0.017	-0.158	-0.047
10	0.021	0.703	0.224	0.231	-0.176

^aPCs representing more than 5 percent of variance are in bold. PCs with low explained fraction and poor correlation with aerosol mass are generally dominated by variation due to measurement uncertainty. Composition at Trinidad Head is more diverse than at the mountain sites, as shown by the correlation data: PC-mass relationships are more varied at Trinidad Head; total mass at the mountain sites is largely explained by the first PC.

from different origins, with each aerosol type represented by a characteristic suite of elements; by pooling the data from the three sites it is possible to directly compare relative impacts of particular aerosol types among the sites. PCA deconvolves the bulk aerosol into a suite of distinct size chemistry “components” that are statistical analogs of the real aerosol components. PCA works on the variance matrix in a data set. Since a variance matrix constructed from raw concentration data will be dominated by the most common components in the heaviest size cuts, the raw elemental concentration values were first converted to size-specific z scores (the difference between an observed element/size concentration value and the data set mean for that element/size variable, divided by the data set standard deviation of that element/size variable). This procedure makes the PCA more sensitive to trace elements, which are often diagnostic of aerosol origins. The variance statistics from the transformed data set show what elements vary together within and across size bins without regard to absolute amounts, thus this PCA is primarily a tool for characterizing chemical and size variation, and it is only moderately responsive to total aerosol mass variation.

[20] The 14 selected elements were chosen not only for good quantification, but also for expected aerosol source associations (see Table 1): Cl (sea salt, grass smoke); Na, Mg, Ca (sea salt, soil); Fe, Al, Ti, Si, (soil, these elements plus Ca are sufficient to distinguish Asian dust from local soil [VanCuren and Cahill, 2002] and fly ash); K (soil, biomass smoke); Zn (motor vehicles, biomass, metallurgy); P (lubricants, biomass); V, Ni (fossil fuels, especially heavy oil); S (fossil fuels, sea salt). This data set contains 112 variables, and PCA accordingly produces 112 components. Virtually all the meaningful variation in the data are represented by the first few components (Table 2), so only the most significant PCs are discussed here. PCA results presented here were computed using the MATLAB function “princomp” [MATLAB, 1999].

[21] Our PCA generates two classes of results: The first is a set of correlation matrices that define the covariance structure of the data, statistical analogs of the real chemical

components that compose the total aerosol. The second is a set of time series of PCA “scores” that denote the relative strength of each component at each time step. The scores can be used to correlate changes in aerosol composition with other factors, such as meteorological conditions, or, by regression, to assign aerosol mass concentrations to the statistical components.

[22] In PCA the first component can be thought of as describing the mean state of the sample set, and other components describe sources of deviation from the mean. Components may represent a “real” aerosol, a temporal “resonance” driven by coordinated variation of the influences of “real” aerosols, or an artifact due to measurement or analytical factors. Intuitive analysis is often adequate to associate the suite of elements identified for a major component with a “real” aerosol type, while “resonances”

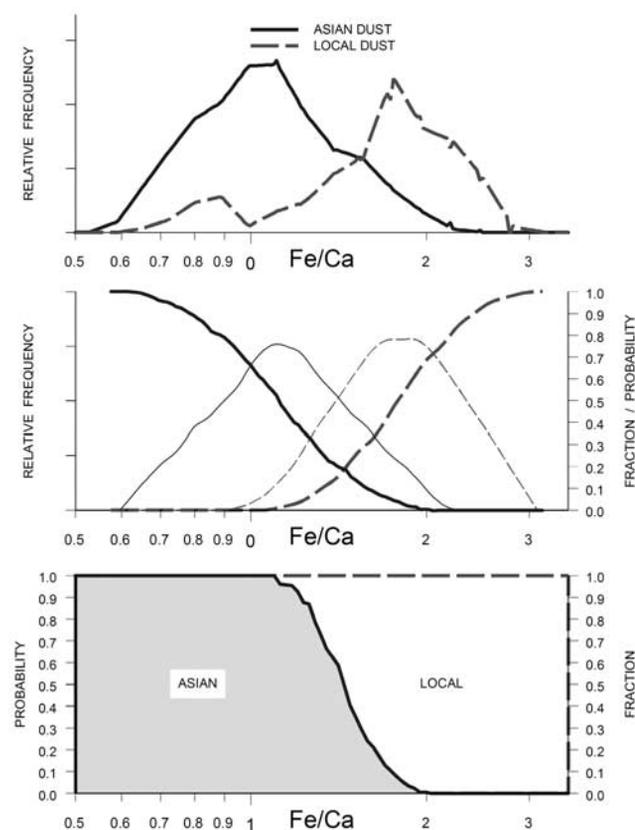


Figure 2. Bayesian mixing model for Asian and local dust. (top) Curves of frequency distributions for Fe/Ca ratios for 501 Asian dust events (solid line) and 100 local dust samples (dashed line); area under both curves equals 100%. Bulge on low end of “local” distribution appears to be due to Asian transport events included in the data set; high side shoulder on “Asian” distribution suggests cross-contamination with local dust. (middle) Frequency distributions smoothed and adjusted for cross-contamination plotted with resulting cumulative frequency curves. (bottom) Bayesian probability that a sample having a particular Fe/Ca ratio is part of the Asian or local distribution (left y axis); the mixing model interprets the relative probabilities at each Fe/Ca value as fractional contributions from the two populations (right y axis). See color version of this figure in the HTML.

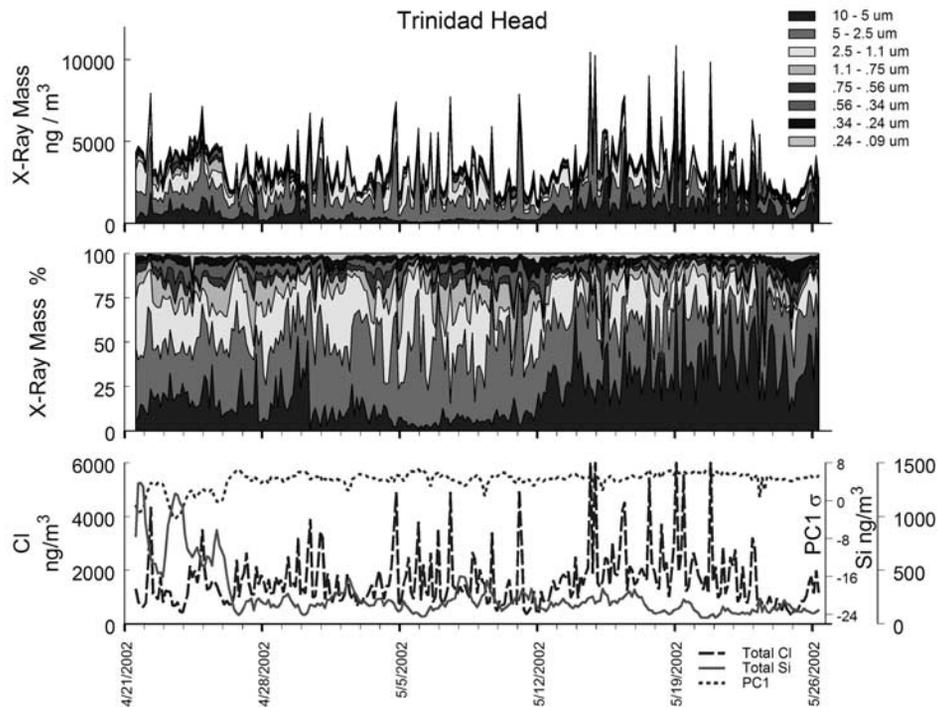


Figure 3. Aerosol summary for Trinidad Head during ITCT 2K2. (top) Aerosol concentration shown as cumulative plot of X-ray mass (sum of measured elements) for 8 stages in 3-hour increments. (middle) Relative mass by stage. (bottom) Total chlorine (long dash), total silicon (solid), and the first principal component (PC1) for the 3-site ITCT 2K2 data set (short dash). Strong diurnal cycling is evident in all three plots. Cl, a marker for sea salt, correlates with the mass spikes in the top plot, showing the episodic nature of onshore/offshore winds and strong salt flux. Silicon, a marker for continental aerosol, peaks at the Cl minima, indicating replacement rather than mixing with marine aerosol during a frontal passage early in the sampling period. PC1 tracks the oscillation between continental and marine aerosols; very low values mark replacement of marine aerosol by continental materials. See color version of this figure in the HTML.

often track meteorological patterns. Artifacts generally appear physically nonsensical (e.g., association with a single element or a “square wave” time trace), and “noise” tends to lack any temporal structure.

[23] In practice, the signs of the species correlations within a single PC are arbitrary in relation to physical reality, since the computation works on total variance (squared differences from means). Physical reality may be represented by either the positive or negative associations in the PC, or by both. Interpreting the PCs as representative of contributing aerosol mass, we generally rely on the fact that the lower limit of any input z value is fixed at negative \bar{x}/σ , while there is no similar limit on maximum z values; thus peak concentrations for most species (elements) tend to have higher absolute value PC scores than minima. Using this as a guide, we invert PCs as needed to force the largest scores to be positive unless otherwise indicated by additional information. When reporting correlation coefficients for inverted PCs, we accordingly change their signs as well.

[24] Our reference for aerosol chemical speciation links to source types is primarily the United States Environmental Protection Agency’s SPECIATE database [U.S. Environmental Protection Agency (U.S. EPA), 2002], while we base identification of the Asian continental material on published analyses from Hawaii [Holmes and Zoller, 1996] and the United States [VanCuren, 2003]. Aerosol aging (growth,

hydration) is inferred from the migration of combustion elements from very fine sizes typical of fresh emissions or young secondary particles (stage 8) into larger size bins (generally stages 7, 6, or 5). Na without Cl is generally interpreted as a sign of nitration of sea salt (Cl displaced by reaction with combustion-derived nitric or sulfuric acid).

5.2. Soil Analysis

[25] Asian soil aerosol collected over the North Pacific and in the continental United States has a distinctive elemental composition which has been used to recognize aerosol samples in which the dust is overwhelmingly Asian [Holmes and Zoller, 1996; VanCuren and Cahill, 2002; VanCuren, 2003]. Here we extend the use of that chemical signature to deconvolve the mixing state of samples containing both Asian and local dust. Our mixing model is based on a Bayesian probability analysis comparing the Fe/Ca ratios of Asian and local dust (Figure 2). The model computes the probability that a sample with a specific Fe/Ca ratio is part of either the Asian or local population of Fe/Ca values, then interprets the probabilities as a mixing ratio. Since the two sample populations have about 20% overlap (Figure 2, middle), the uncertainty of the assignments is about 20% at the 50/50 point (i.e., there is a 20% probability that that data point is actually purely of one type), declining to zero for samples identified as purely Asian or purely local.

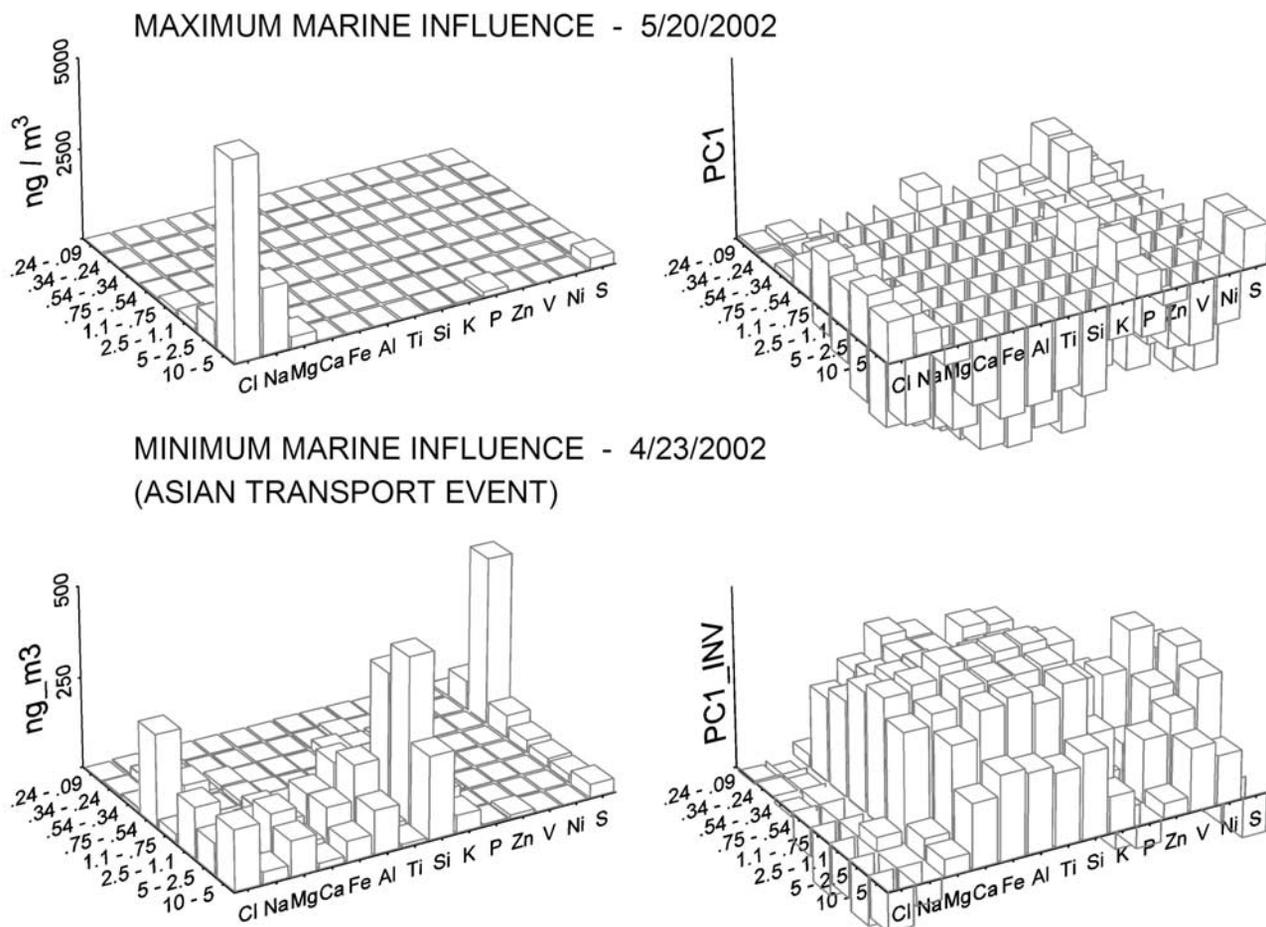


Figure 4. Alternating composition of the Trinidad Head aerosol. (a) Composition at the peak PC1 score (5/20/02); coarse sea salt and sulfur dominate. (b) Composition at the lowest PC1 score (4/23/02) when Asian transport reaches sea level; coarse salt drops, coarse soil appears, and sulfur is concentrated in very small sizes. (c) PC1 and (d) PC1 inverted correlograms demonstrate that the replacement occurs for virtually all elements and sizes.

[26] The Asian Fe/Ca data are from 501 IMPROVE samples collected at Crater Lake and Mount Lassen (Figure 1) that were previously identified as dominated by Asian dust [VanCuren, 2003]. The local dust Fe/Ca data are from 100 winter (low Asian transport season) IMPROVE samples collected at South Lake Tahoe, CA (SOLA in Figure 1). We believe the SOLA samples are representative of local Sierra-Cascade soil dust because the site is located 50m from a heavily traveled highway where road dust is dominated by local soil tracked onto the roadway from unpaved areas, occasionally supplemented by locally obtained volcanic cinder applied for winter traction. On the basis of Sierra-Cascade geology, we assume that the Fe/Ca ratios for these materials are representative of local dust at the ITCT mountain monitoring sites, and the calculated results shown here are consistent with that assumption. We did not apply the model to the Trinidad Head data due to the potential confounding effects of copious sea-salt aerosol.

5.3. Comparison With the Long-Term Record at Mount Lassen

[27] The comparison between the present data and the long-term record for Mount Lassen provides a qualitative

check on how well the ITCT 2K2 sampling period represents long-term conditions in the region. The comparison uses PCA applied to mimic the previously published PCA analyses [VanCuren, 2003], modified to use elements common to both data sets. The results are focused on comparing general aerosol composition during ITCT 2K2 to the decade of data previously reported [VanCuren, 2003]. This section has been kept simple and brief, as a full intercomparison of the two monitoring methods is beyond the scope of this paper.

6. Results

[28] This section discusses each site separately; beginning with a brief overview of the record at the site, followed by discussion of the PCA results, and concluding with the soil analysis results. An overview of the findings follows the site-specific discussions.

6.1. Trinidad Head

6.1.1. Aerosol Chemistry

[29] Figure 3 shows the general structure of the Trinidad Head aerosol during the ITCT 2K2 study period. The

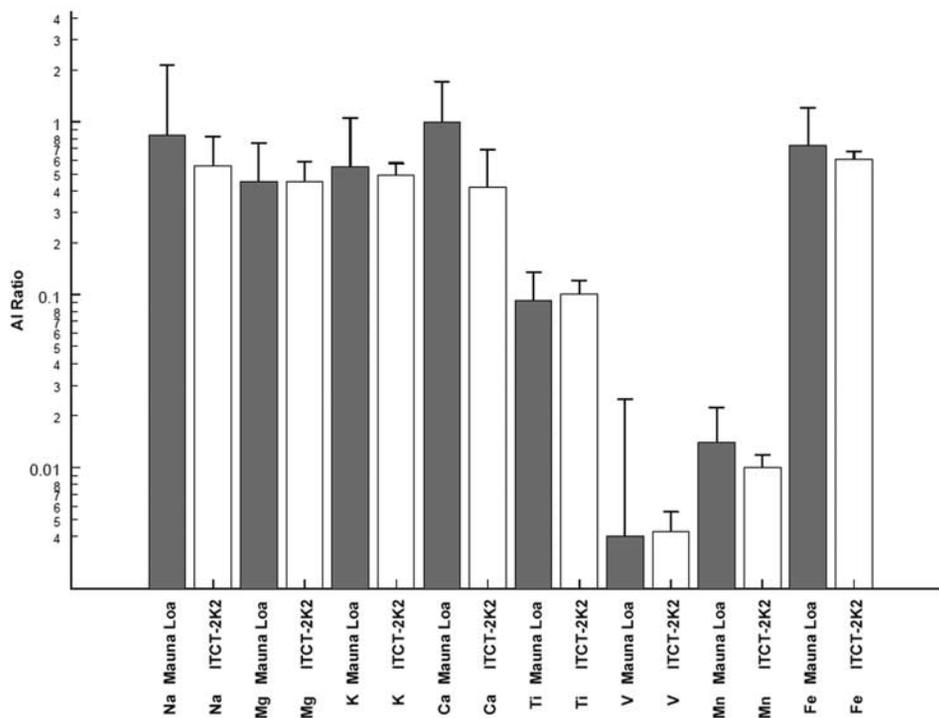


Figure 6. Comparison of ratios between Al and other major elements in the continental aerosol (PC1 Inverted) observed in ITCT 2K2 with published Al ratios for Asian dust collected at Mauna Loa, HI [Holmes and Zoller, 1996].

which would reduce large particles by sedimentation and fines by coagulation.

[32] The continental aerosol represented by low values of PC1 is the Asian continental plume. This was confirmed by: (1) back-trajectories that indicate it came from Asia (Figure 5); (2) comparison with the Al ratios of major soil elements from these periods with published values for Asian dust collected at Mauna Loa, HI [Holmes and Zoller, 1996] (Figure 6); and (3) the fact that the particular pattern of elemental ratios found in the Asian dust has previously been shown to be a definitive marker distinguishing Asian dust from local soil dust in most of North America [VanCuren and Cahill, 2002].

6.1.2. Aerosol Temporal Trends

[33] The Trinidad Head aerosol time series (Figure 3, bottom curves) shows a persistent oceanic aerosol mixed with various amounts of combustion products that is only occasionally supplemented or displaced by a continental aerosol composed of soil and combustion products. There is little relationship between chemical composition and total aerosol concentration at Trinidad Head. The three curves plotted at the bottom of Figure 3 are total chlorine (an index of marine influence), total silicon (an index of continental influence) and the first Principal Component (PC1), which is flat for most of the sample period. Despite considerable variation in trace chemical composition and strong diurnal variation in size distribution, the Trinidad Head aerosol retained its marine character with the exception of 4/20, 4/22, and 4/24 when marine aerosols were exchanged for primarily Asian continental material as a result of downward mixing associated with strong frontal activity. At those times the PC1 score goes strongly

negative as marine aerosol concentrations drop and the ambient aerosol composition is dominated by soil- and combustion-derived species.

[34] The strongest driver of aerosol mass at Trinidad Head is a mixture of sea salt and aged combustion aerosol, represented by PC2. It is anticorrelated with soil, and seems to come from either recirculation of industrial stack emissions from coastal sources or large combustion sources at sea (ships?).

6.2. Trinity

[35] The Trinity site is about 100 km east-southeast of Trinidad Head, and about 1 km above sea level. The site is a local high point, has no nearby population, and is only seldom visited. The air at the site is exposed both to elevated flow off the Pacific Ocean and upslope movement of locally generated pollutants. Unlike Trinidad Head, the aerosols at Trinity were much less varied and were strongly continental in character. Total aerosol mass varied on multi-day scales, rather than diurnally, and there is a period of about a month when a single, monomodal size distribution (Figure 7, center) was observed despite wide variation in total aerosol mass. These gross characteristics suggest that aerosol loading at Trinity was controlled by synoptic-scale processes modulating the concentration of an otherwise fairly uniform aerosol, with only weak influence by local emissions.

6.2.1. Aerosol Chemistry

[36] The aerosols at Trinity were dominated by continental material consisting of soil and combustion products. The general structure of the Trinity data is shown in Figure 7. At first glance it might appear that the

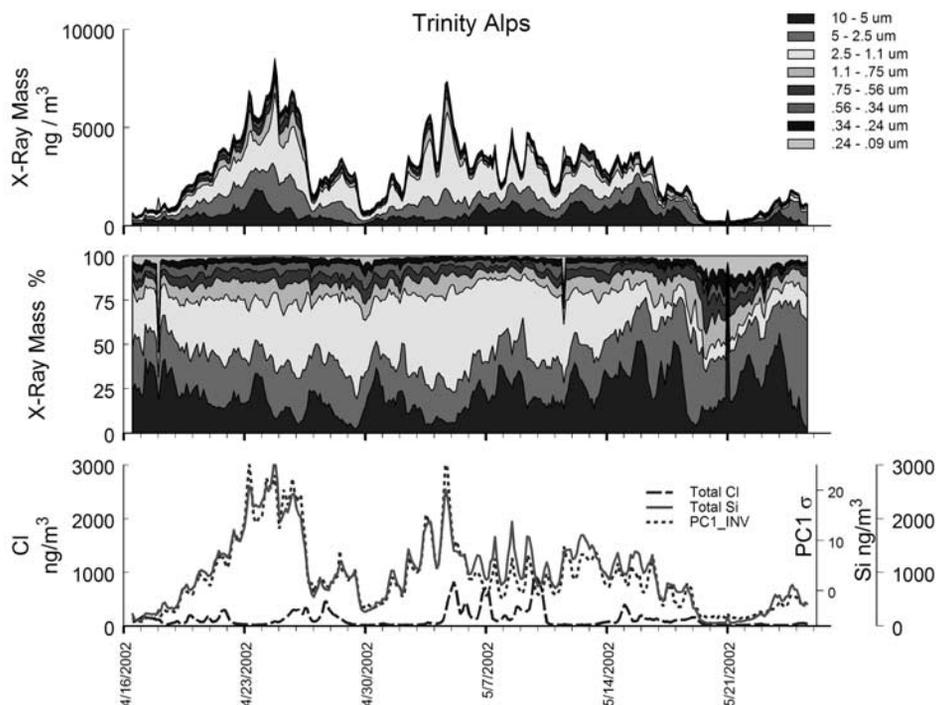


Figure 7. Aerosol summary for Trinity site during ITCT 2K2. (top) Size distribution (8 stages) of X-ray mass (total of S-XRF reported elements) in 3-hour increments. (middle) Cumulative plot of X-ray mass for 8 stages. (bottom) Total chlorine (long dash), total silicon (solid), and first principal component for this data set (short dash). The first principal component is dominated by soil and sulfur, indicating an anthropogenic continental aerosol. Time series of total silicon tracks with total aerosol mass. The strong period of trans-Pacific transport from 4/16 to 5/16 is marked by highly variable aerosol flux (top), but relatively stable aerosol composition (middle). Sea-salt chlorine is suppressed when the continental aerosol peaks (bottom). See color version of this figure in the HTML.

Trinity site experienced local North American continental aerosols.

[37] On closer examination, however, the data are incompatible with that interpretation. The presence of soil elements but persistent dominance of medium diameter particles over the period 4/16–5/16 (Figure 7, middle) suggests long distance transport is a major factor in addition to local sources. The composition of the aerosol is also indicative of Asian origin, with characteristic comparable concentrations of Ca and Fe in stages 2–4 (Figure 8, top). Although visible in the concentration plot (Figure 8), an additional feature of Asian aerosol is more obvious in the PCA correlograms: the combination of elevated Ca and S (PC1, Figure 9), characteristic of the Asian continental plume due to atmospheric reaction of sulfuric acid with mineral calcium carbonate [Xiao *et al.*, 1997; Song and Carmichael, 2001]. The abundance of fine combustion metals, especially P, Zn, and K (PC1, Figure 9) and the monomodal size distribution peaking near 3 micrometers diameter (Figure 7, middle) are also characteristic of the Asian aerosol [VanCuren, 2003].

[38] The Trinity site is not downwind of any local population centers likely to produce such an aerosol, and, moreover, the peak concentration event for this aerosol component at Trinity coincided with the frontal activity that delivered Asian continental aerosols to Trinidad Head (4/21–24). The Asian attribution is further reinforced by the bulk Al ratios at Trinity, which agree with the Mauna Loa

data cited above for the continental aerosol event at Trinidad Head.

6.2.2. Aerosol Temporal Trends

[39] The title for each time series plot in Figure 10 gives our subjective interpretation of the composition associated with that component, and each curve is labeled with the site-specific correlation coefficient between that time series and total aerosol (X-ray mass) concentration. On a bulk aerosol basis, the Asian continental plume controlled aerosol concentration and chemistry at the Trinity site throughout the sampling period (correlation of 0.96). Unlike the Trinidad Head situation, the alternation of marine and Asian influence is dominated by the Asian mode, with marine aerosol accompanying it in the westerly winds, but the marine mode is significantly weaker except during deep mixing associated with frontal passages (4/21–24, 5/2–5). Regression of PCs 1 and 2 against X-ray mass indicate that the “marine combustion” aerosol is only about $1/4$ the strength of the Asian plume at Trinity. Moreover the Asian influence is consistently larger than the marine influence. When marine air (as denoted by sea salt, Figure 9, bottom) does reach this site (5/5–5/10) there is a mixture of Asian continental material and salt in the aerosol and the diurnal concentration variation is enhanced.

[40] Unlike Trinidad Head, PC1 dominates both chemical variance and total aerosol mass, and has significant temporal variation throughout the sampling period. The drop in aerosol mass on 4/26–27 is due to a low pressure system

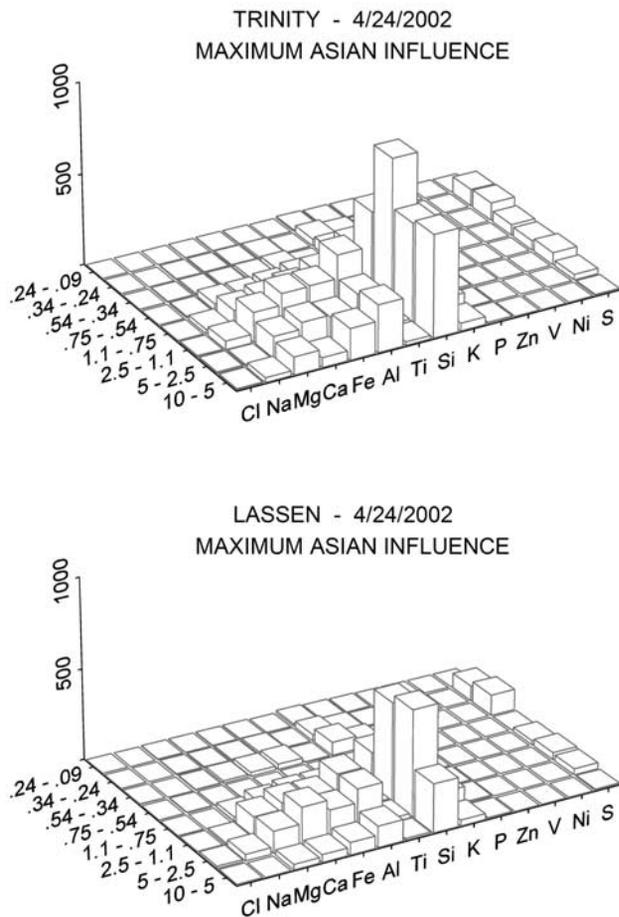


Figure 8. Aerosol composition at the mountain sites during the peak Asian impact on 4/24: (top) Trinity and (bottom) Lassen (concentration in ng/m^3). Composition is essentially the same at both sites. Sea salt is absent, and the Asian soil signature (Ca comparable to or greater than Fe) is present except in stage 1 at both sites. Fine sulfur indicates that although dust is strong in these samples, it is accompanied by combustion products.

passage that pushed air from the SW over the region and brought rainy weather through about 5/1 along most of the U.S. Pacific coast. This wet period also forced PC4 (local soil dust) to low values, with gradual recovery later in the sampling period.

[41] An unusual feature of the Trinity data is the spikes in PCs 3 and 4. These appear to be related to servicing the equipment at the site, and emphasize how isolated this site is from daily human activity.

6.2.3. Soil Analysis

[42] Application of the soil mixing model to the Trinity data (Figure 11) shows that the coarse dust was predominantly local, while the fine dust, especially in the period before 5/13, was overwhelmingly Asian. The pattern of Asian soil dominance in the smaller stages from the beginning of the sampling period up to about 5/8 supports the interpretation of the chemical data that aerosol chemistry over that time was strongly controlled by synoptic patterns, and that these flows contained aerosols derived from mainland Asia. In the later period, after 5/8, local flow became

stronger relative to the synoptic fields, and diurnal effects controlled aerosol composition. During this part of the record daytime upslope flows carried locally generated soil dust to the Trinity site. Nonetheless, the persistent presence of Asian aerosols in the overlying free troposphere is attested by the nightly fumigation of the Trinity site with fine Asian dust.

[43] The model was also used to address the uncertainty that coarse soil is local. When viewed with the two large spikes removed, PC4 lacks the expected dominant diurnal cycle in the PCA score and instead appears to be under synoptic-scale control, challenging the expected pattern for emissions from local sources. Figure 12 shows the Fe/Ca ratio time series for Stage 1 plotted with the PC4 score. It is clear from the plot that periods with high PC4 scores are associated with Fe/Ca ratios that are generally outside the range of Asian dust. This indicates that the local coarse dust sampled at Trinity was strongly modulated by such factors as soil moisture and atmospheric stability. As mentioned above, the Trinity site is rarely visited, thus, unlike populated areas, its coarse dust is not linked to diurnal human activity in the immediate vicinity of the monitoring site.

6.3. Lassen

[44] The Lassen site is about 100 km farther inland than Trinity and at nearly twice the altitude, with a moderate size urban area (Redding CA) and a substantial agricultural region (northern Sacramento Valley) in the lowlands between the Lassen and Trinity sites. Despite these potential local aerosol sources, historical analysis of a decade of aerosol data from Lassen indicates that the dominant aerosol mode at Lassen is tropospheric transport from Asia [VanCuren, 2003]. Following the pattern applied to Trinidad Head and Trinity, aerosol mass, size variations, and soil-salt contrasts are shown in Figure 13.

[45] The pattern at Lassen is similar to that for Trinity, but with significantly greater coarse particle mass. This appears to be a consequence of the combination of road dust in the vicinity of the site (there are permanent National Park facilities nearby) and the lack of the PM10 inlet; consequently there is a higher diurnal cycle imposed on top of the synoptic variation. In the finer size ranges (stage 3 and below) the Lassen data show a generally weaker local signature and a greater impact of Asian air masses.

6.3.1. Aerosol Chemistry

[46] The chemistry of PC1 peak events at Lassen is virtually identical to that for Trinity (Figure 8, bottom), and is clearly the same aerosol, the dusty Asian continental plume. PC2, the marine air that dominates Trinidad Head and appears as a consistent signal at Trinity is virtually absent at Lassen. PCs 3 and 4 are motor vehicle exhaust and road emissions. As at Trinity, PC3 has a “spiked” temporal pattern, reflecting infrequent vehicle traffic near the sampler, while PC4’s road dust is the strongest secondary component after PC1 (correlation coefficient of 0.87). Regression analysis of PC1 and PC4 against total aerosol mass indicates that the road plus exhaust aerosol mass was generally about the same size as the aerosol load from the Asian plume. During low total aerosol periods it exceeded the Asian mass by a factor of 5 or more, but the reverse was true during the peak Asian flux periods. If the local dust size

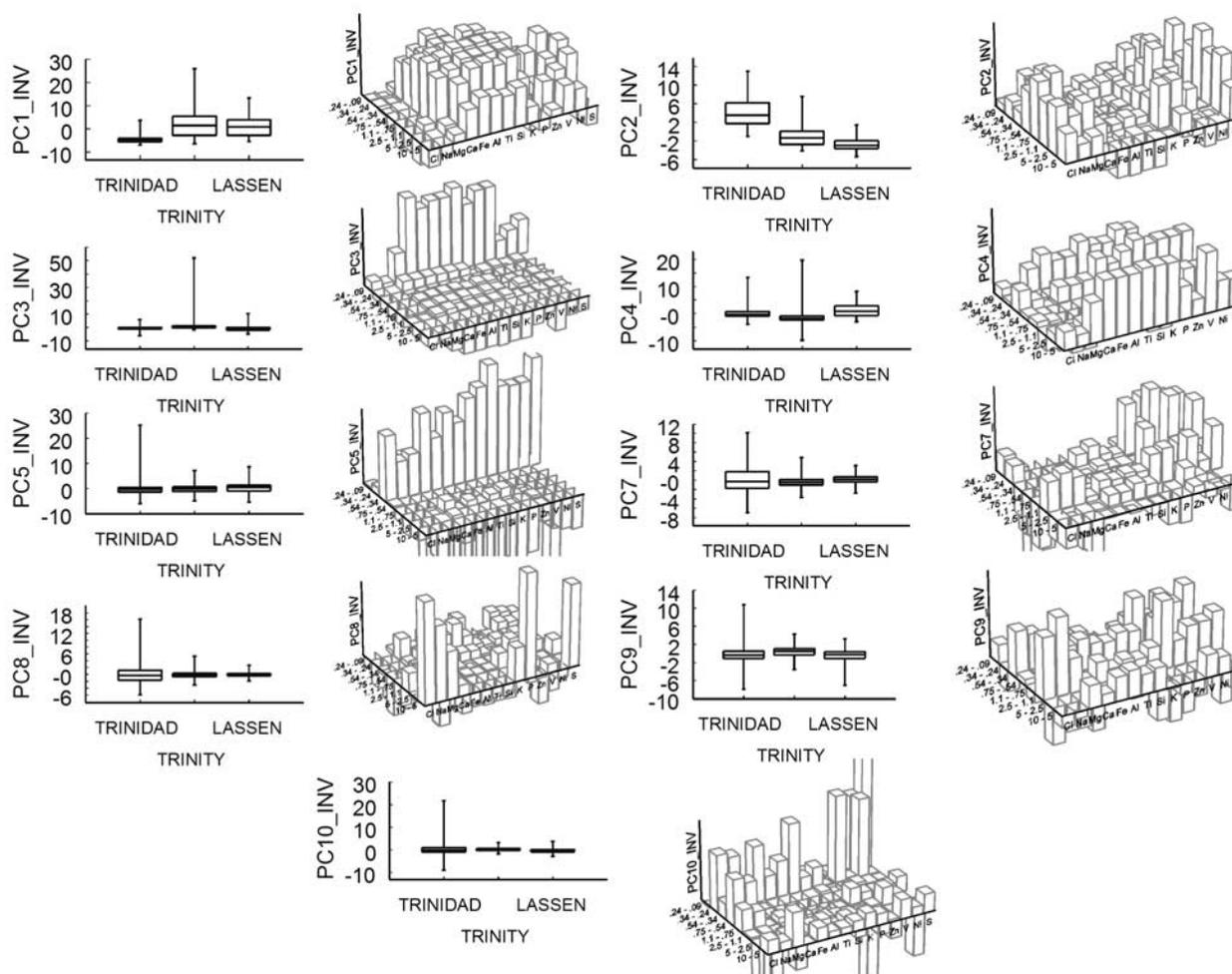


Figure 9. PCA score distributions (box and whisker quartile plots) and correlograms (3-D plots) for major components of the Trinidad Head, Trinity, and Lassen pooled data. PC1 tracks a broad mixture of soil and combustion, anticorrelated with salt (the Asian plume; see text). PC2 is sea salt with aged combustion (ships?) and secondary sulfate, anticorrelated with soil. PC3 is fresh fine combustion such as motor vehicle exhaust. PC4 is aged hydrated combustion mixed with coarse soil, presumably road emissions. PC5 is hydrated fresh combustion alone, possibly a plume from a large utility boiler. PC7 is coarse salt with aged secondary combustion, broad size distribution suggests aged aerosol, and source association is unclear. PC8 is coarse salt, generally weak except at Trinidad Head. PC9 is medium salt, aged combustion, and some soil, perhaps aged coastal air. PC10 is fresh combustion and fine salt, possibly small marine engines; it is generally confined to Trinidad Head. PC6, not shown, is an artifact due to frequent low values of certain species.

mode (stage 1) is excluded from the regression, then the Asian material becomes dominant.

6.3.2. Aerosol Temporal Trends

[47] The temporal pattern for PC1 is much the same as seen at Trinity, including the same general time trend in total aerosol, and the same reactions to precipitation events observed at Trinity. The coarse particle fraction at Lassen is somewhat greater than that at Trinity, as noted above, and the site's closer proximity to regular vehicle traffic is reflected in a stronger diurnal cycle in large particles traced by PC4.

6.3.3. Soil Analysis

[48] The soil model results for Lassen are shown in Figure 14. The strong diurnal cycles for coarse dust are evident. In addition, perhaps because of its higher altitude,

Lassen appears to be more strongly influenced by nighttime fumigation of coarse Asian dust particles. The local fraction of finer size dust may have been reduced early in the period by weak local soil dust emissions due to snow cover, but its persistence into late May suggests it is, at least in part, driven by atmospheric processes. Fine Asian soil is also more persistent late in the sampling period at the Lassen site than at the Trinity site, especially after 5/7, when local circulation appears to have become stronger compared to synoptic fields.

7. Intersite Comparison

[49] There are three salient features in these data. First, the Asian continental aerosol dominated the samples at

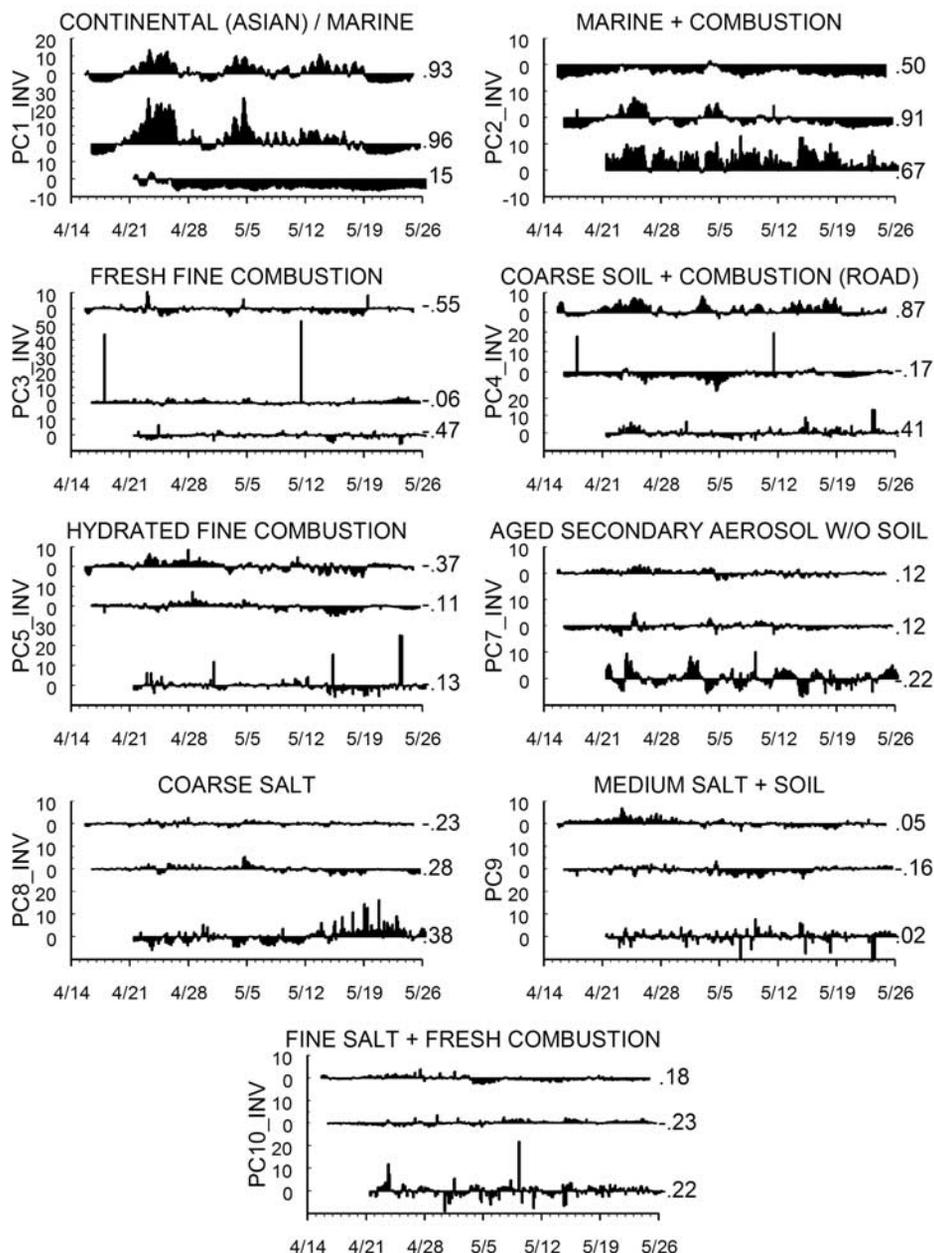


Figure 10. PC score time series for Trinidad Head (bottom curve), Trinity (middle curve), and Lassen (top curve) for the strongest PCs. Labels characterize aerosol represented by each PC. Numbers at right are correlation coefficients with X-ray mass by site. PCs 1 and 2 are the dominant modes associated with prevailing westerly winds: continental aerosol (the Asian plume) anticorrelated with sea salt, and sea salt mixed with combustion (ships?). PCs 3, 4, 5, and 10 are associated with strong local source impacts: fresh combustion such as on-site engines, road emissions, and plume impacts from large combustion sources. PC 7 is very long range transport, while PCs 8, 9, and 10 are diurnal effects originating in the coastal zone.

Trinity and Lassen throughout the sampling period. Second, marine aerosol was dominant at Trinidad Head except for a few brief periods when it was replaced by the Asian continental aerosol due to intense vertical motion in the atmosphere. Third, the data from Trinity and Lassen clearly show that there was Asian material overlying the marine and continental boundary layers more or less continuously during the six weeks of this experiment. The Asian aerosol

was observed only sporadically at Trinidad Head; it was dominant at Trinity, accompanied by small amounts of marine aerosol; it dominated Lassen without significant accompanying marine aerosol. It was strong at both mountain sites due to persistent strong synoptic forcing early in the sampling period. After early May local circulation became more important, but the Asian aerosol continued to regularly appear at night, fumigating down from the free

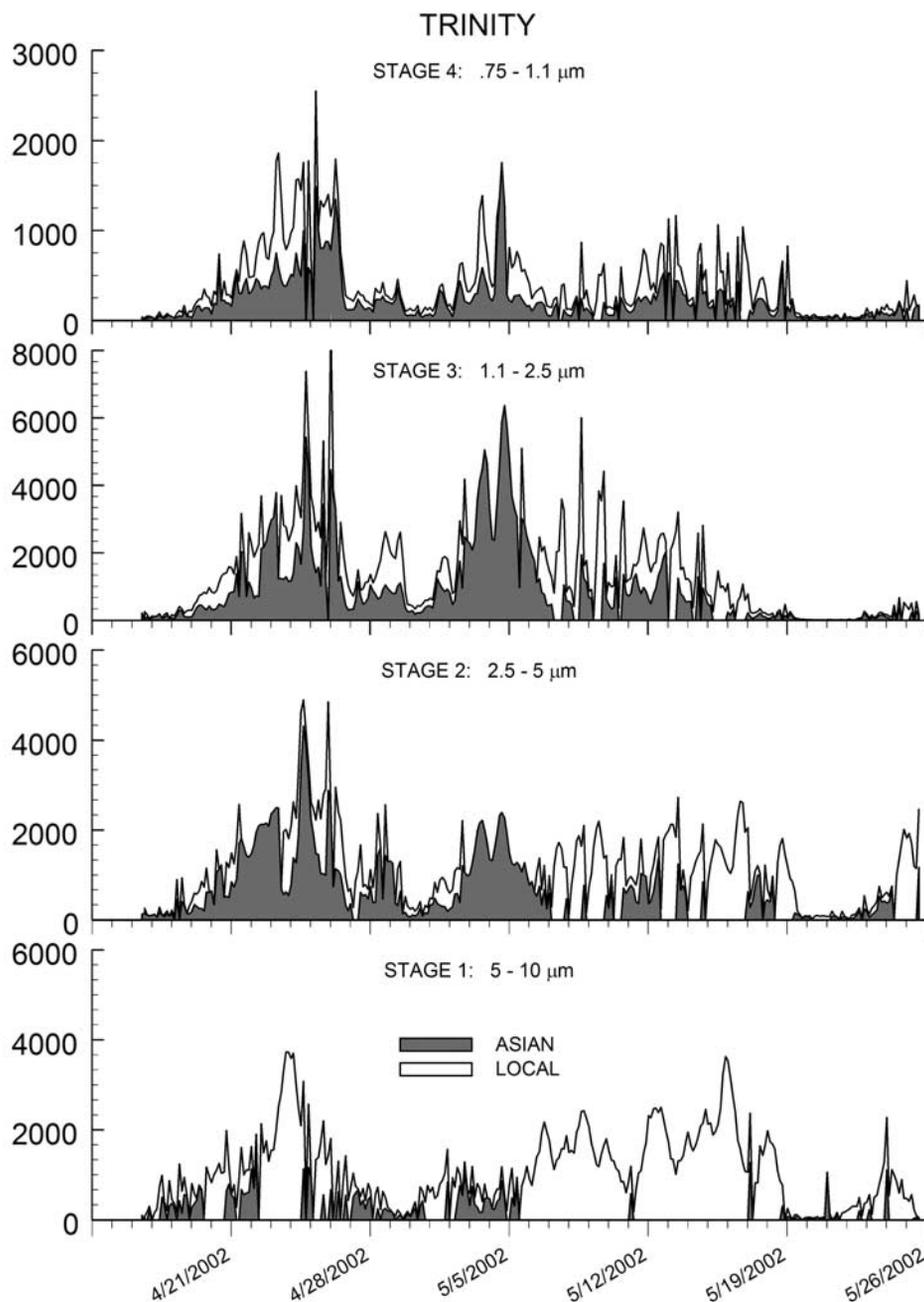


Figure 11. Modeled local and Asian soil for Trinity by stage. Coarse soil is generally dominated by local material except during periods when the local soil was wet from recent rains. Soil in smaller size bins is increasingly Asian. Note that the diurnal peaks in stages 2–4 in the period after 5/1 are out of phase with local upslope transport as tropospheric transported material is delivered to the surface by subsidence. This coincides with the cessation of the synoptic process control period and the emergence of local diurnal controls seen in the mass and chemical data from Trinidad Head.

troposphere, even when there was no strong synoptic driver for rapid trans-Pacific transport.

[50] The time series analyses show that mixing between the marine layer and overlying air was generally poor, with the chlorine-rich marine aerosol infrequently mixing with, and never dominating the Asian aerosol at Trinity or Lassen.

[51] The bases for these general conclusions are evident in the box-whisker plots in Figure 9. The range of mid-quartile (25th to 75th percentiles) PC scores for PC1 (the

Asian plume) are nearly identical for Trinity and Lassen, and both score consistently higher than Trinidad Head. The persistent low values at Trinidad Head are the result of isolation of free troposphere air from the coastal zone by the persistent, stable marine boundary layer; the consistent scores at the mountain sites indicate that they both have about the same exposure to the free troposphere aerosol.

[52] In contrast, PC2, consisting of marine aerosols, shows a classic fall-off with increasing inland distance

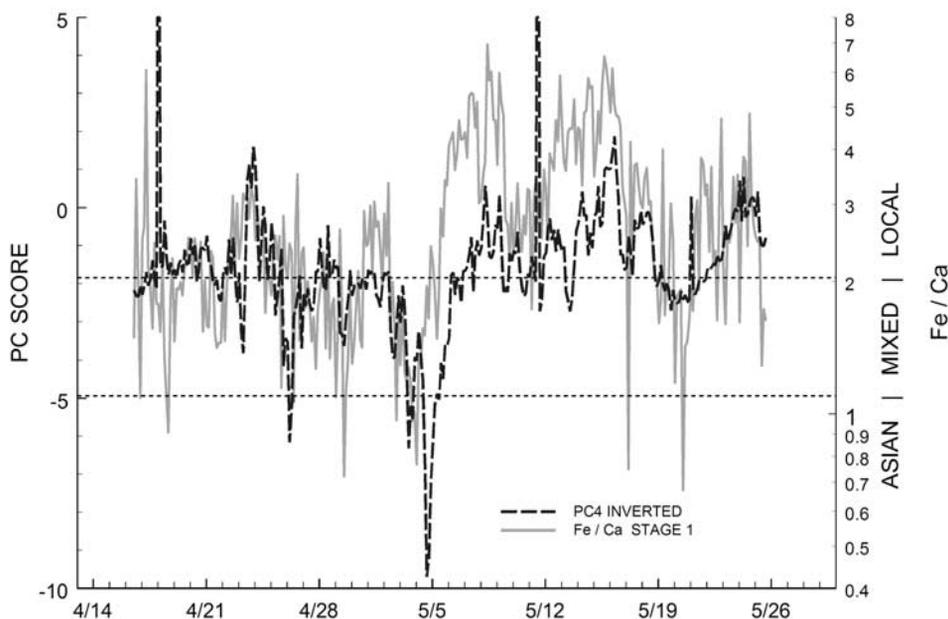


Figure 12. Time series overlay of Trinity PC4 (inverted) and coarse particle (stage 1) Fe/Ca ratios. The horizontal lines are the limits of the mixed composition range from the soil model; bottom of graph is Asian composition, top is local. The latter period of stronger local soil coincides with the cessation of the synoptic process control period and the emergence of local diurnal controls seen in the mass and chemical data from Trinidad Head and Trinity. See color version of this figure in the HTML.

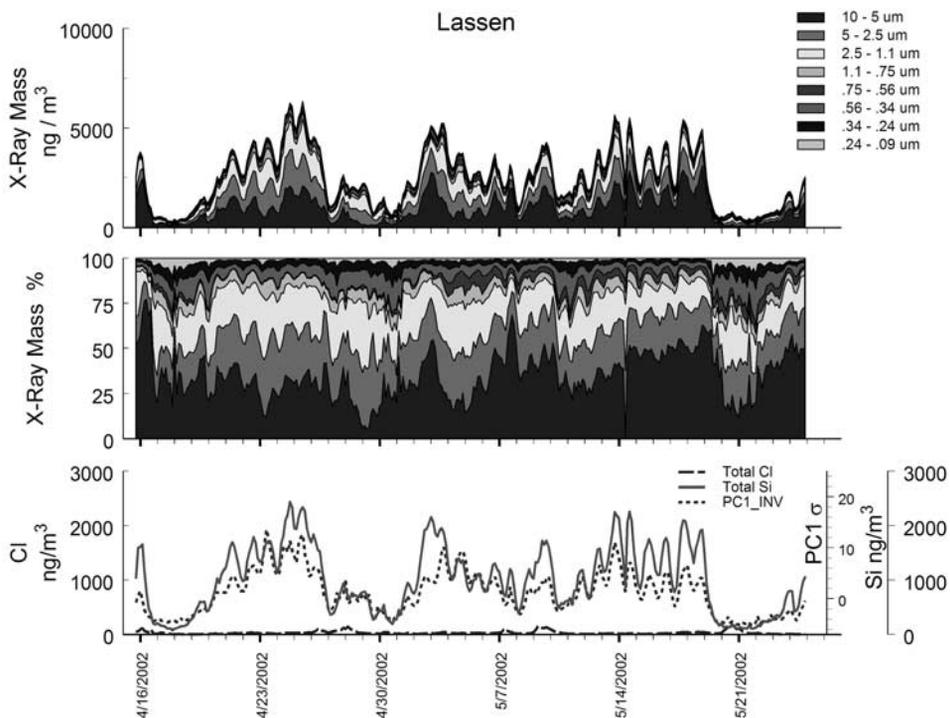


Figure 13. Aerosol summary for Mount Lassen site during ITCT 2K2. (top) Size distribution (8 stages) of X-ray mass (total of S-XRF reported elements) in 3-hour increments. (middle) Cumulative plot of X-ray mass for 8 stages. (bottom) PC1 inverted (short dashed line), total chlorine (long dashed line), and total silicon (solid line). The first principal component, the dusty Asian aerosol, dominates total aerosol mass, with the diurnal effects of local emissions and upslope/downslope winds superimposed. General absence of Cl throughout the period shows that unmodified marine air is rare at Lassen. Note the strong similarity of these plots with those for Trinity (Figure 7). See color version of this figure in the HTML.

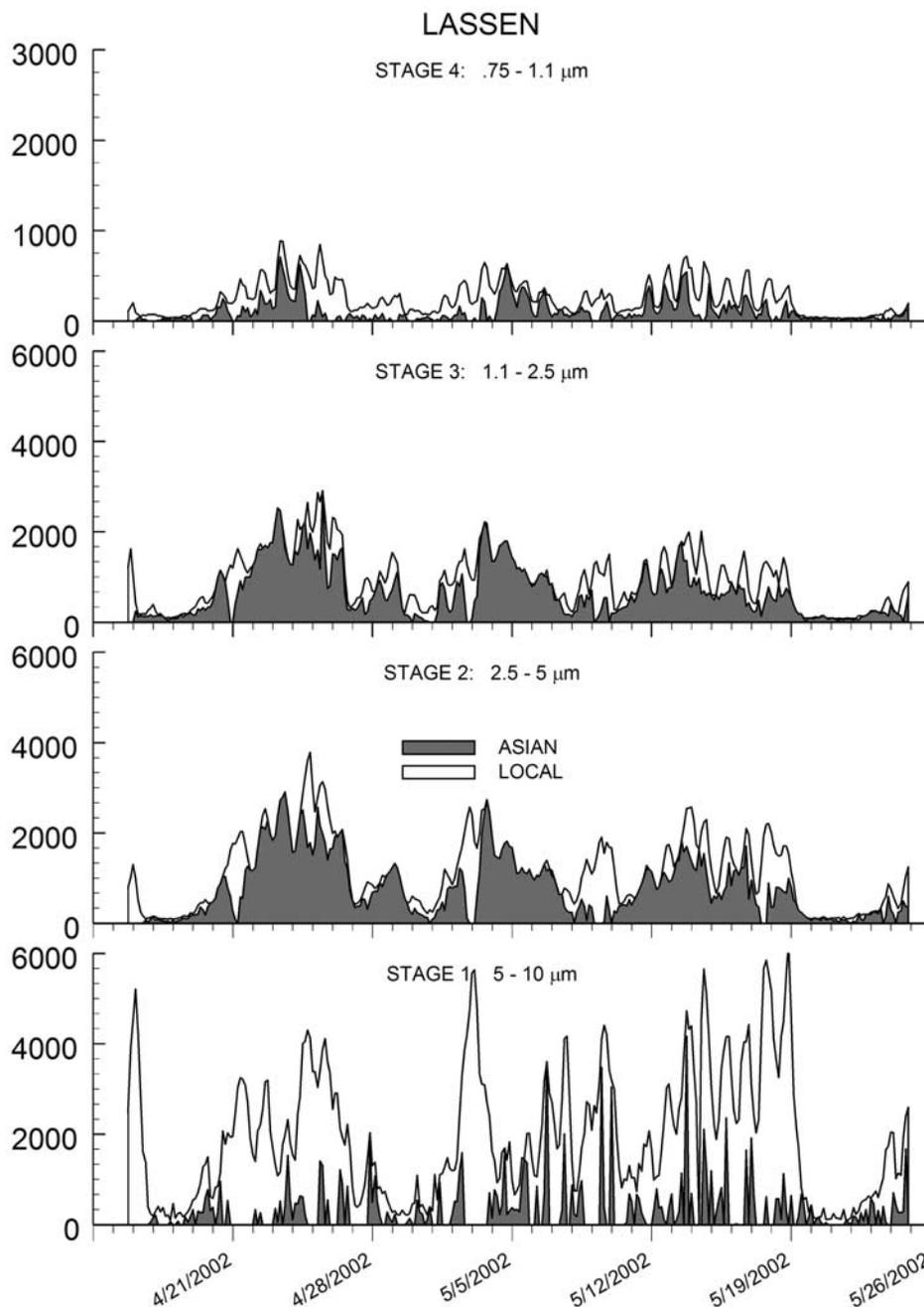


Figure 14. Modeled local and Asian soil for Lassen by stage. Coarse soil is dominated by local material due to daytime upslope transport, but Asian large particles appear almost every night due to subsidence (compare with stage 1 at Trinity in Figure 11). Soil in smaller size bins is strongly Asian throughout the sampling period. Note that stages 2–4 are more strongly Asian than observed at Trinity, probably owing to both increasing altitude and snow cover. Snow cover at this site through the first half of the sampling period significantly reduced local dust production except for roads, which produced abundant coarse aerosol. Coarse dust production at Lassen increased with the cessation of the synoptic process control period and the emergence of local diurnal controls in concert with the pattern at Trinity.

and elevation. The difference between the coast-mountain trends for these two PCs underscores the difference between marine and free troposphere effects on continental aerosol loading; the marine effect is confined to near-coastal areas, while the free troposphere impacts are carried far inland. This is consistent with the continental-scale impacts reported by *VanCuren and Cahill [2002]*.

[53] Regressions between the first component and total aerosol (X-ray mass) for each of the mountain sites (Figure 15, top and center) shows that the Asian aerosol chemical signature explains most of the observed aerosol mass at both Trinity and Lassen. These regressions indicate that there is a single dominant aerosol mode, and site-specific aerosol sources are minor. The chemistry indicates

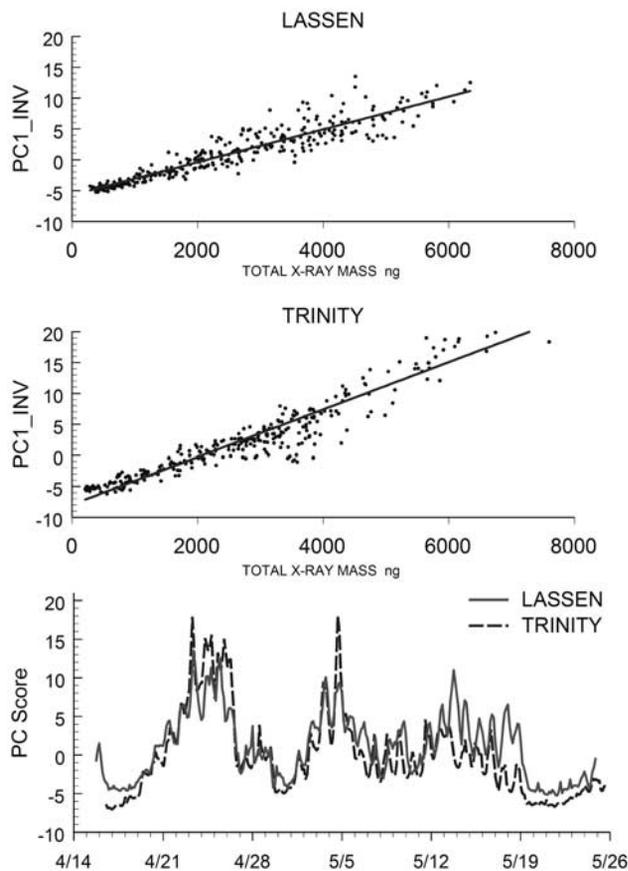


Figure 15. Regressions between analyzed mass (“X-ray mass”) and the first principal component of the aerosol at Trinity and Mount Lassen and comparison of time trends at the two sites. PC1 inverted represents the Asian continental aerosol, which clearly dominates both sites, with its variation in concentration driven by a combination of large-scale circulation and nocturnal fumigation at these mountain sites. See color version of this figure in the HTML.

that it is the Asian continental aerosol. Trinity showed only a weak marine effect, while there was a general lack of Cl or reacted salt at Lassen; together they show that unmodified marine air is rare at montane sites except near the coast. This highlights the separation between the lower free troposphere in which the Asian aerosol travels and the stable marine layer that is in contact with the ocean.

[54] A significant finding of this analysis is the importance of nighttime fumigation as a mechanism for delivering free troposphere air to surface monitoring sites in the mountains. The general experience of local nighttime inversion formation at lowland and valley monitoring sites is not an appropriate conceptual model for understanding the effects of intercontinental transport to elevated sites. The persistent effects of diurnal cycling of pollutant regimes is clearly seen in Figure 16. During the period of strong synoptic circulation (prior to about 5/7) the diurnal composition patterns at both Trinity and Lassen are controlled by regional free tropospheric flows. After 5/7, the weather becomes less active and local diurnal circulation becomes stronger; this results in much stronger transport of lowland

air to Trinity, which responds with strong diurnal variation in aerosol source. The effect at Lassen, another km in altitude, is much less pronounced.

[55] Despite the observation of increasing local effects at the mountain sites late in the study period, however, regional processes still have a strong influence and appear to synchronize aerosol dynamics at both sites. From the beginning of sampling up to early May, the temporal curves at the mountain sites show strong temporal correlation (Figure 15, bottom). Even when circulation is more locally controlled, as evidenced by the spikes and diurnal cycling associated with local emissions in the latter third of the sampling period, the mountain sites still are strongly influenced by Asian aerosol, especially at night, and multiday variation of aerosol loads is broadly coordinated. Despite significant distance between the mountain sites and differing geographic settings, the Asian signature’s time trend (PC1 at each site) is highly correlated over the entire 6 weeks of the ITCT 2K2 experiment (Figure 15, bottom).

[56] Overall, the ITCT 2K2 surface aerosol sampling observed two distinct regimes: a marine-dominated aerosol at Trinidad Head, and an Asian-dominated aerosol at Trinity and Lassen. Locally produced aerosols were observed, but regionally generated North American aerosols were generally weak at all three sites during this experiment.

8. ITCT 2K2 and the Long-Term Record at Mount Lassen

[57] In order to evaluate how well the ITCT 2K2 period represents “typical” conditions in the region, we compared the ITCT 2K2 samples from Lassen with the overall Asian aerosol impact at Mount Lassen for the period 1988–2000 reported by VanCuren [2003].

[58] Qualitatively, the published analysis and the present data appear similar: The decadal analysis found that about half of the coarse aerosol (2.5–10 μm diameter) and about three-quarters of the fine (<2.5 μm) was associated with the Asian plume; the soil analysis reported here shows about 15% of the coarse soil and about 65% of fines at Lassen were of Asian origin.

[59] While we do not have total mass from the DRUM samples, we can compare soil and sulfur: Statistics from the two data sets are shown in Table 3. The present data fall well within the range of values reported in the long-term data.

[60] Isolating the months of April and May, we compared the elemental composition by applying PCA to the two data sets as though they were both filter samples for particles < 2.5 μm diameter. These results are presented in Figure 17. The limited number of samples and large uncertainties for low concentrations conspire to limit the comparison to the first two components. Within the scope of this analysis PCs 1 and 2 appear to be equivalent.

[61] On the basis of these comparisons, the ITCT 2K2 data are representative of “typical” springtime conditions in the region.

9. Summary of Findings

[62] The ITCT 2K2 DRUM aerosol data show the following.

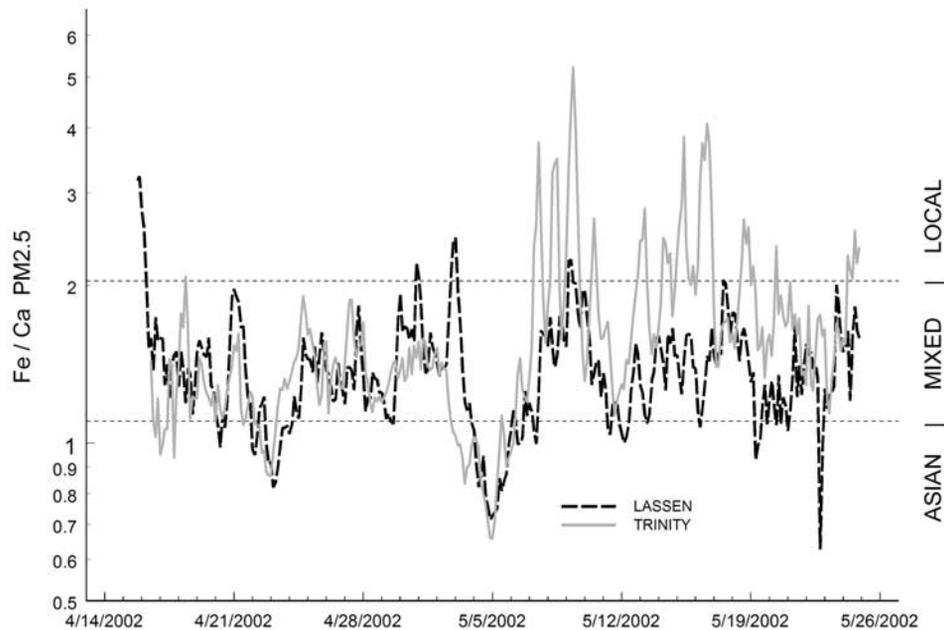


Figure 16. Time series of Fe/Ca ratios for PM_{2.5} (sum of stages 3–8) for Lassen and Trinity. Date marks are local midnight. Dashed lines are limits of mixing model. General trend is coordinated at the two sites. Asian contribution to fine soil is persistent at both sites through 5/7. Local daytime upslope winds alternating with nocturnal fumigation with Asian aerosols from free troposphere occurs at both sites. The diurnal effect is more pronounced at Trinity, consistent with its lower elevation. The strong diurnal period at Trinity coincides with the dominance of local diurnal effects seen in the Trinidad Head data. See color version of this figure in the HTML.

[63] 1. Marine aerosols were dominant at the sea level site throughout the sampling period, despite more or less continuous Asian aerosol transport above the marine boundary layer. The marine air generally contained a recognizable combustion component, whether arriving from the ocean or landward direction. A distinctive Asian aerosol mass only reached sea level during a frontal passage between 4/22 and 4/25, when marine aerosols at Trinidad Head were replaced by transported Asian continental aerosols.

[64] 2. The dominant aerosols at the elevated sites were of Asian continental origin throughout the sampling period, accounting for most of the aerosol mass and dominating the chemical composition.

[65] 3. Asian aerosol was present continuously during periods of strong synoptic circulation; during weaker flow regimes it peaked at night due to subsidence fumigation from the free troposphere.

[66] 4. The aerosol mass observed at the 1 km elevated site (Trinity) was consistently dominated by Asian continental aerosols, but also contained a recognizable fraction of marine aerosol, as well as some local soil dust.

[67] 5. The aerosol mass observed at the 2 km elevated site (Lassen) was consistently dominated by Asian continental aerosols with some local dust; marine aerosols were insignificant contributors to aerosol mass.

10. Conclusions

[68] This multisite experiment was designed to assess the composition, temporal patterns, and vertical stratification of aerosols over the northeastern Pacific Ocean. The findings

support the inferences drawn from statistics from intermittent samples [VanCuren and Cahill, 2002; VanCuren, 2003]. The continuous data not only verify the earlier results, but clarify the processes controlling tropospheric aerosol impacts at surface monitoring sites. It is clear from the present data that the apparent weakness of oceanic influence and strong Asian influence at sites in the western cordillera of North America is not solely a result of decreasing flux of marine aerosols moving inland, but is truly a product of separation of trans-Pacific transport from the ocean surface by frequent and persistent meteorological stratification that precludes contact between the lower free troposphere and the ocean surface.

[69] At interior locations, local aerosol impacts at montane sites are governed by diurnal limits on vertical dispersion. Upslope flow is limited to periods of solar heating, thus transport of lowland air pollutants to montane sites is limited to a few hours of the day. During night and morning hours, montane sites experience thermally driven subsidence, which permits downward fumigation of free tropospheric air containing Asian pollutants.

[70] Viewed in isolation, the results of the ITCT 2K2 aerosol sampling show that, in spring, the lower free troposphere over the northeastern Pacific Ocean and near-coastal western North America are dominated by continental outflow from Asia, with little or no marine influence, and suggest that the previously reported episodic nature of trans-Pacific transport to this region is more a phenomenon of varying concentration than one of discontinuous flow.

[71] When viewed in the light of the prolonged “transport season” evident in the long-term aerosol history for Mount

Table 3. Comparison of ITCT-2K2 Data With Published Data for 392 Days With Asian Dust (1988–2000) at Mount Lassen IMPROVE site [VanCuren, 2003]^a

	IMPROVE 1988–2000		ITCT-2K2	
	Mean	Std. Dev.	Mean	Std. Dev.
PM2.5-10	7.9	5.3		
SOIL	3.9	3.8	3.1	3.2
PM2.5	3.9	2.4		
Soil	0.7	1.0	1.3	1.3
Sulfur	0.2	0.3	0.2	0.1

^aConcentrations are in $\mu\text{g}/\text{m}^3$.

Lassen and Crater Lake [VanCuren, 2003], it is likely that the pattern of nearly continuous transport observed in this study is similar to the structure of trans-Pacific transport through summer and fall as well. This is consistent with observations made over the western Pacific [Moore *et al.*, 2003] and published model calculations [Xiao *et al.*, 1997; Song and Carmichael, 2001]. In short, both long-term and intensive ground level observations indicate that the background tropospheric aerosol over western North America is generated in Asia.

[72] These findings also bear on the proper appreciation of Asian and marine aerosols' respective climate impacts. The

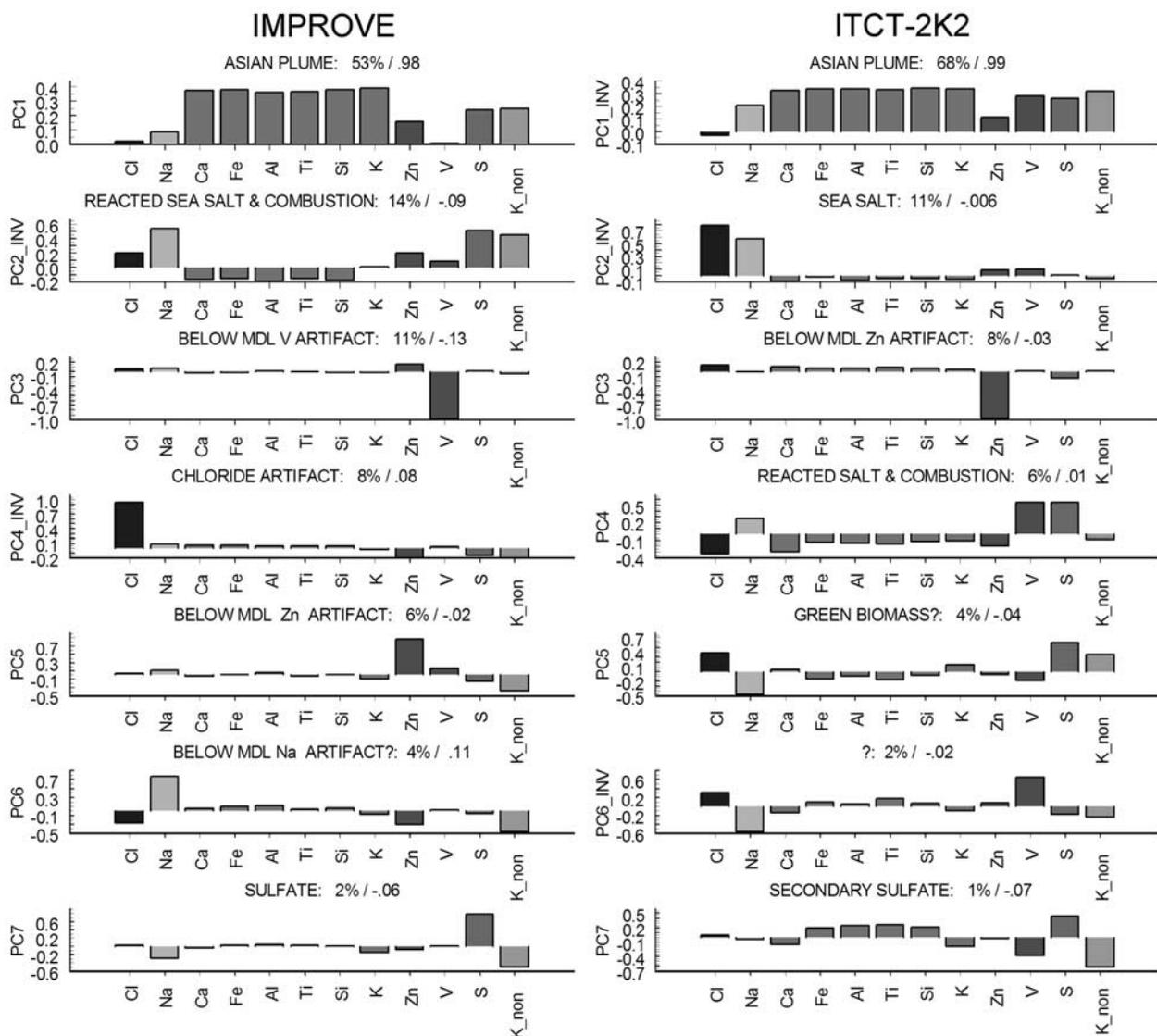


Figure 17. Left column shows principal component elemental associations for all sample days April through October for years 1988–2000 in Mount Lassen IMPROVE PM2.5 samples (components scored for percent of total variance, not mass). Right column shows principal component elemental associations for pseudo-PM2.5 composite from ITCT 2K2 samples at Mount Lassen. The first components are equivalent; ITCT 2K2 data resolve sea salt alone as well as the marine combustion component. Remaining components are similar in both data sets, but interpretations are somewhat uncertain owing to low sample count, effects of frequent values below minimum detection (MDL), large uncertainties for low concentrations of V, Na, and Zn in both data sets, and Cl ion uncertainties in the IMPROVE data. See color version of this figure in the HTML.

observed extreme stratification suppresses deposition of Asian continental aerosols to the surface of the eastern North Pacific Ocean, thus greatly extending their atmospheric lifetimes. Such a persistent flux of Asian aerosols suggests that Asian emissions (both natural and anthropogenic) play a significant role in driving both direct and indirect aerosol effects on free tropospheric radiation balance over the much of the north Pacific basin (above the marine boundary layer), and that the role of marine aerosols in such processes is largely confined to the boundary layer. This implies that terrestrial emissions in Asia may significantly modulate free troposphere aerosol radiation budgets over the North Pacific. If so, further investigation is needed to assure that these processes are faithfully reproduced in general circulation models and their impacts on past and future climate are properly accounted.

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