Asian aerosols in North America: Frequency and concentration of fine dust

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[1] Using an elemental signature for Asian dust derived from events in April 1998, we probed a long-term set of routine aerosol samples to develop the first empirical assessment of the frequency and intensity of dust transport from Asia to midlatitude North America. Our data reveal a pattern of consistent, frequent transport that contradicts the episodic characterization derived from short-term studies and anecdotal reports. We find that fine (<2.5 μm) Asian dust is a regular component of the troposphere over the eastern Pacific and western North America and is common, at least in spring, across North America. Typical Asian fine dust concentrations (24-hour average) are between 0.2 and 1 $\mu g/m^3$ and only very rarely exceed 5 $\mu g/m^3$. Our data also indicate that Asian dust is concentrated in an altitude zone ranging from about 500 to 3000 m MSL, consistent with isentropic transport processes previously observed in the western Pacific. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* dust, Asia, North America, aerosol, troposphere

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1. Introduction

[2] There is intense international interest in atmospheric transport of natural and anthropogenic contaminants in and across the North Pacific basin because of its potential to impair health and welfare in populated regions of the western Pacific, impact biologic systems throughout the northern Pacific, and impact climate on a basin- to hemispheric scale [*Wilkening et al.*, 2000]. The mineral dust component of the troposphere provides condensation nuclei for cloud formation, fertilizes the oceans, supplies reaction surfaces for atmospheric chemical processes, and interacts with long and short wave radiation to influence the sun-earth radiation balance [*Prospero*, 1996; *Intergovernmental Panel on Climate Change* (*IPCC*), 2001]. Atmospheric mineral dust is one of the most uncertain factors contributing to the effects of aerosols on global climate processes [*IPCC*, 2001]. Determining the sign and intensity of dust-driven radiation forcing requires detailed knowledge of dust characteristics and spatio-temporal distributions [*Tegen and Lacis*, 1996; *Alpert et al.*, 1998].

[3] Mineral dust aerosols are also tracers for the origin of air masses, and by extension, the sources of accompanying anthropogenic pollutants. Such intercontinental pollutants range from combustion products (lead, arsenic, sulfur, elemental carbon, *etc.*) to bio-accumulated persistent organic pollutants such as PCBs and dioxins [*Wilkening et al.*, 2000].

[4] The deserts of western China are known to be very large dust sources, but their contribution to global atmospheric dust loading is not well quantified [*IPCC*, 2001]. Reports of short-term studies or anecdotal observations have created the impression that Asian dust production is primarily a spring phenomenon [*Wang et al.*, 2000; *Merrill*, 1989; *Merrill et al.*, 1989; *Duce et al.*, 1980, *Perry et al.*, 1999; *Jaffe et al.*, 1999]. Analyses of long-term records are necessary to properly characterize the temporal structure of transpacific aerosol transport.

2. The Anecdotal Record

[5] Intense Asian "yellow sand" dust events are observed each spring in China, Korea and Japan and occasionally in Hawaii and North America, contributing to the previously noted impression of transpacific transport as an episodic, springtime phenomenon [*Duce et al.*, 1980; *Perry et al*, 1999; *Jaffe et al.*, 1999]. The scarcity of surface monitoring sites in the midlatitude Pacific Ocean, the inability of present satellite sensors to detect low concentrations of airborne dust, especially over land [*Kaufman et al.*, 1997],

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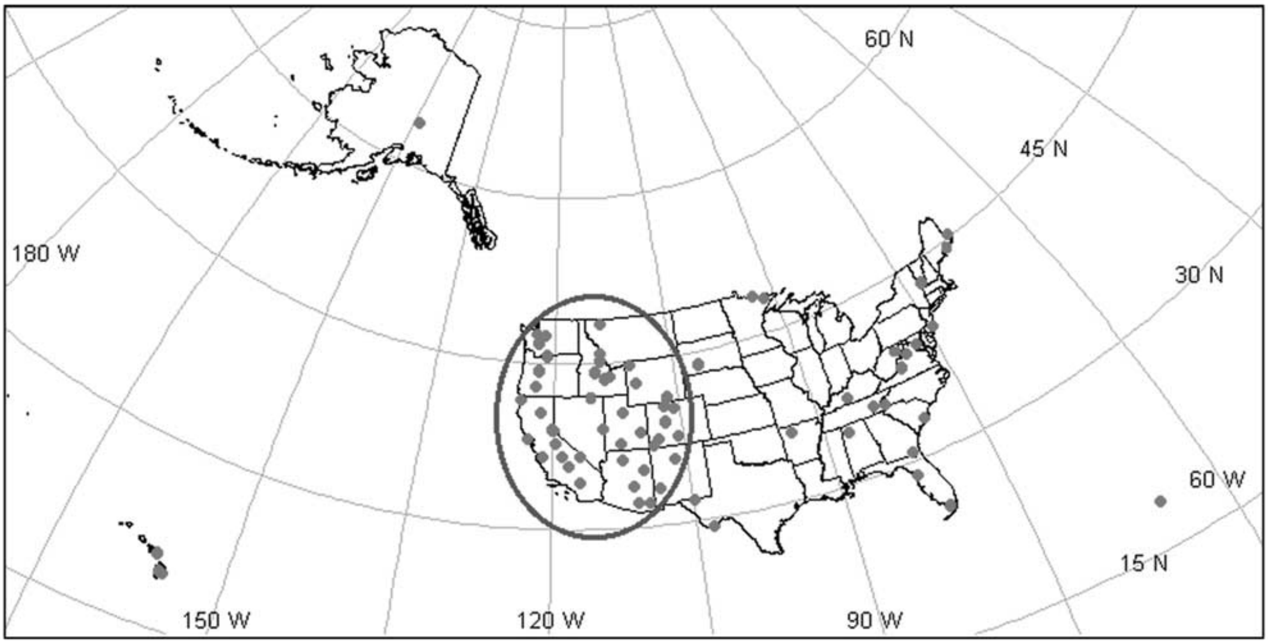

Figure 1. IMPROVE monitoring network during the 1990s. The sites used to characterize the 1998 events are circled.

and the sparse and infrequent data from large-scale surface and airborne aerosol measurement programs in the North Pacific basin combine to created a patchwork record that emphasizes infrequent high-concentration events.

[6] The apparent low frequency of dust transport events is, however, difficult to rationalize with observations of the regularity of dust transport into the mid-Pacific [*Parrington et al.*, 1983; *Gao et al.*, 1992; *Harris and Kahl*, 1990] and the fact that Asian sources have been shown to dominate dust deposition in the North American Arctic both in modern times and at the last glacial maximum (25,000 years ago) [*Rahn et al.*, 1977; *Welch et al.*, 1991; *Biscaye et al.*, 1997; *Bory et al.*, 2002]. The mid-Pacific data suggest that dust-laden air masses are common over the North Pacific, while the arctic data suggest that Asian dust sources operate nearly year-round. It follows that Asian dust should be a persistent component of the troposphere over extra-tropical North America as well. Resolving this apparent paradox requires supplementing the anecdotal record with data on the contemporary long-term frequency and concentration of Asian dust over the eastern North Pacific and North America. We report here an 10+ year record of both high and low concentration events of Asian dust aerosols around the northeastern Pacific Basin and across North America based on data from an extensive network of remote aerosol sampling sites.

3. Methodology

3.1. Analytical Model

[7] Our analysis consists of two steps: first, identify a chemical signature for Asian dust from known events, and second, compare that signature to historical aerosol samples to detect similar events. To accomplish this, we rely on the following assumptions: (1) a strong Asian dust event will impact a large area in North America, resulting in chemically similar contemporaneous samples at multiple sites. (2) During such a large event, the "soil" fractions of samples collected at remote sites far from urban, agricultural, or industrial pollution sources will generally consist almost entirely of Asian dust, with little or no "local" contamination. (3) The chemical characteristics of these extreme events are representative of other, lower-concentration Asian dust events reaching North America. (4) Historical samples whose composition data match those of the extreme event archetypes also represent Asian dust events. (5) The selected chemical signature for Asian dust can be reliably distinguished from those of other potential continental-scale dust sources.

3.2. Database

[8] The Interagency Monitoring for Protected Visual Environments (IMPROVE) sampling network was established in the late 1980s to monitor intrusions of anthropogenic aerosols into National Parks and Wilderness Areas in the United States [*Malm et al.*, 1994; *Sisler et al.*, 1996]. Although not intended for the present application, the IMPROVE network sites are well suited to detecting dilute, continental-scale aerosol clouds because (1) they are generally remote from population centers, minimizing masking by local pollutant emissions; (2) many sites are at elevated locations, reducing the frequency of isolation from the lower free troposphere caused by surface temperature inversions; and (3) the IMPROVE protocol provides a wide range of elemental measurements, permitting extensive multivariate analysis. The IMPROVE sites in operation during the 1990s are plotted in Figure 1 with the western U.S. subset used to develop the Asian dust signature circled.

[9] Our analysis used the elemental data from IMPROVE twice weekly (Wednesday/Saturday) 24-hour integrated samples of particles less than 2.5 μm aerodynamic diameter (PM2.5). Over our analytical period from late 1989 through

early 1999 a continuously operated site presents a record exceeding 1000 samples. IMPROVE analysis includes 24 elements (Al, As, Br, Ca, Cl, Cr, Cu, Fe, K, Mn, Mo, Na, Ni, P, Pb, Rb, S, Se, Si, Sr, Ti, V, Zn, Zr) measured by proton-induced X-ray emission (PIXE) and X-ray fluorescence (XRF), selected ions ($Cl-$, NO_3-, $SO_4=$) by ion chromatography (IC), organic and elemental carbon (OC/EC) by staged thermal desorption and combustion, and total hydrogen by proton elastic scattering (PESA). IMPROVE reports PM2.5 mass down to about 300 ng-m^{-3} with uncertainty of ±200 ng-m^{-3}.

3.3. Defining an Asian Signature

[10] We used cluster analysis to isolate Asian signatures during the known events, develop signatures for potentially interfering non-Asian dust sources (specifically, North American deserts and North Africa), and to test the ability of the signatures to be matched with historical records. Clustering works well with these data because it is tolerant of mixing well-quantified high concentration events with weakly quantified low concentration ones, thus minimizing the number of samples rejected from the analysis. Cluster analysis requires the least data adjustment and fewest assumptions for dealing with missing values or measurements near or below the limits of detection (the latter reported as zero in the IMPROVE data). Clustering works with these data since it only requires that similar samples produce similar analytical results. Elemental ratios for samples near detection limits may have large quantitative errors, but this tends to prevent their clustering with their better-behaved siblings. In the multidimensional clustering space a sample with an element unreported or below detection will score a zero on that axis; multiple nondetects will pull the sample toward the origin, away from the centroid of the target composition. Overall, clustering is more likely to reject erroneous classification in the target type than to promote it. Because our primary goal was to assess the frequency of the presence of Asian dust over North America we chose to trade precision for minimizing the bias that would accompany exclusion of "clean" days, which are known to have a high probability of oceanic or boreal air mass associations.

3.3.1. The April 1998 Dust Events

[11] Two major dust events from April 1998 served as the archetypes for this analysis. The first began with a very large dust storm originating in the Takla Makan desert in Xinjiang, western China on 14 April 1998. This dust cloud spread eastward across northern China, Mongolia, the Korean peninsula, Japan, and the North Pacific, and eventually swept across the United States and Canada. The second originated on 19 April 1998 in the Gobi desert region of north central China and southern Mongolia, and spread along a similar path, eventually reaching Hawaii and Alaska, as well as midlatitude North America. Extensive analyses of these events are reported elsewhere [*Husar et al.*, 2000; *Tratt et al.*, 2000; *Murayama et al.*, 2001; *Uno et al.*, 2001]. The published data and our own analyses of these events support our belief that these events fit our analytic model.

[12] To create a representative set of replicate Asian and non-Asian samples for the April 1998 events we compiled data for samples collected between 15 April and 6 May 1998 for 36 IMPROVE sites in the western United States (236 samples). Figure 2 shows time series fine (PM2.5) soil concentration data from those sites compared with modeled dust arrival for these events from work by *Uno et al.* [2001]. The inset plots the modeled dust concentrations arriving over central California for the time period 15–29 April: the top curve is the sum of modeled transport for dust emitted each day from 14 through 19 April (the lower curves are individual day's emissions). The progression of the two dust clouds across the IMPROVE network is clearly visible in the 3-D view of time series measurement data: the short (left side) axis spans the sampling period 15 April to 6 May; the vertical axis is "soil" concentration (IMPROVE convention: SOIL = Al * 2.2 + Si * 2.49 + Ca * 1.63 + Fe * 2.42 + Ti * 1.94.) in μg/m^3; the long (right) axis arrays the sites in order of decreasing west latitude. (REDW-Redwood, CA; PORE-Point Reyes, CA; CRLA-Crater Lake, OR; THIS-Three Sisters, OR; LAVO-Lassen, CA; SNPA-Snoqualmie Pass, WA; PINN-Pinnacles, CA; CORI-Columbia River Gorge, OR; BLIS-Bliss State Park, CA; YOSE-Yosemite, CA; SOLA-South Lake Tahoe, CA; SEQU-Sequoia, CA; SAGO-San Gorgonio, Ca; DEVA-Death Valley, CA; JARB- Jarbridge, NV; SAWT-Sawtooth, ID; GRBA-Great Basin, NV; SALM-Salmon, ID; CRMO-Craters of the Moon, ID; GLAC-Glacier, MT; SCOV-Scoville, ID; BRCA-Bryce Canyon, UT; GRCA-Grand Canyon, AZ; INGA-Indian Gardens (Grand Canyon), AZ; TONT-Tonto, AZ; YELL-Yellowstone, MT; CANY-Canyonlands, UT; BRID-Bridger, CO; PEFO-Petrified Forest, AZ; MEVE-Mesa Verde, CO; WEMI-Weminuche, CO; WHRI-White River, CO; BRLA-Brooklyn Lake, WY; ROMO-Rocky Mountain, CO; GRSA-Great Sand Dunes, CO; BIBE-Big Bend, TX.) The strong agreement between the measured and modeled data reinforces our interpretation that these mass peaks are Asian dust.

3.3.2. Clustering the Test Data

[13] We selected a suite of 6 elements for initial dust characterization (Si, Fe, Al, Mg, Ca, and Ti) based on degree of data completeness, potential for nondust contamination (industrial pollutants, motor fuels, biomass smoke, etc.), and reported IMPROVE detection limits. To remove the effect of sample mass, the concentration data were transformed to the ratios of Fe, Al, Mg, Ca, and Ti to Si. The relative weighting of the elements in the analysis was balanced by standardizing each elemental ratio to the range of each ratio within the April–May 1998 test data set (i.e., the standardized values all range from zero to one).

[14] Groups of similar samples within the test data set were identified by cluster analysis, with the specificity of the clustering judged by how well it matched those samples for which we had a priori understanding from the well-characterized April 1998 events.

[15] We found the Asian dust events to cluster robustly, with stable numbers in the Asian clusters over a wide range of cluster specifications. Figure 3 shows the progressive clustering among the 236 test samples. The strength of the clustering is obvious: at only 8 clusters, the "Asian" and "non-Asian" groups are largely separated, and the size of this putative "Asian" cluster holds relatively constant while the "non-Asian" group increases from 7 to more than 30 clusters. Since all of the a priori Asian samples are included in this group, we define the core range of compositions

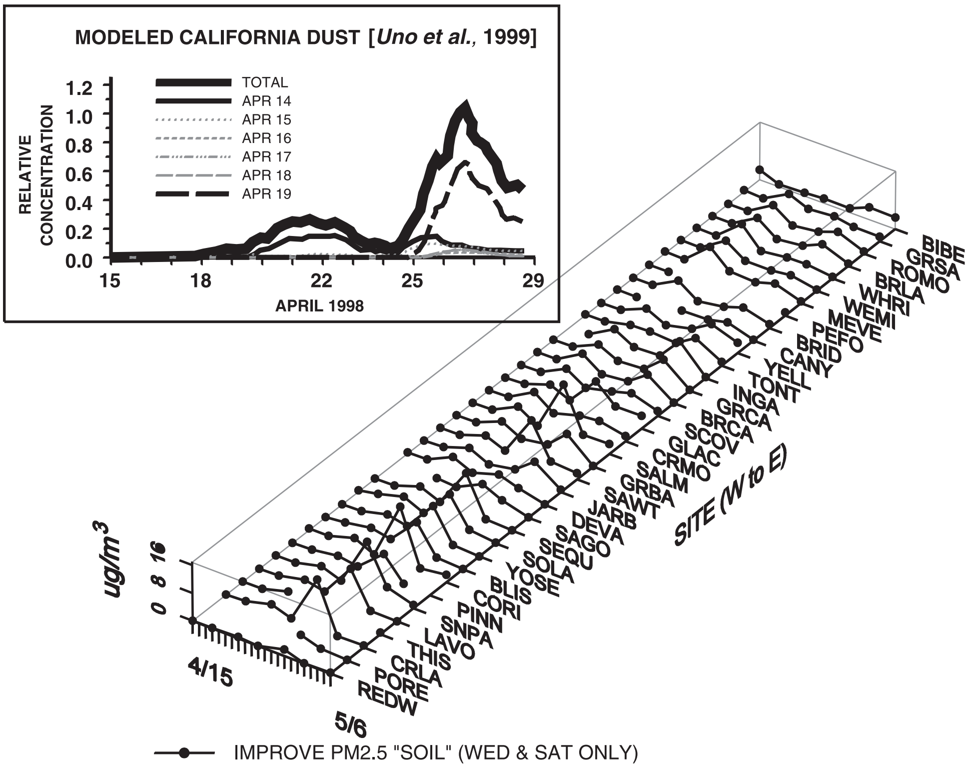

Figure 2. Modeled dust [*Uno et al.*, 2001] and IMPROVE measured fine aerosol soil for 1998 Asian dust events. Uno data: *y* axis is relative dust concentration; *x* axis is arrival date over central California (bold dates are IMPROVE sampling days); numbered curves are for dust entrained in Asia on that date (all dates April 1998).). The 3-D view shows the progression of the two dust clouds across the IMPROVE network with time series data for 36 sites: the short (left side) axis spans the sampling period 15 April to 6 May; the vertical axis is ''soil'' concentration in $\mu g/m^3$; the long (right side) axis lists the sites in order of decreasing west latitude; dots represent 24-hour samples 15, 18, 22, 25, and 29 April and 2 and 6 May; breaks indicate missing data.

within this group (we excluded a few questionable samples) as an operational upper bound for Asian dust recognition. As the clustering becomes increasingly stringent (more than 34 clusters), the ''Asian'' group takes a sharp drop to a lower plateau, which persists until the ''Asian'' group bifurcates at 54 clusters: we interpret the composition range of the lower plateau as a lower operational bound for identifying Asian dust since it contains most, but not all, of our a priori Asian samples.

[16] Starting from the Euclidean distance ranges within the ''Asian'' populations at 32 and 53 clusters, we set two operationally defined levels of specificity for detecting Asian dust: an upper bound based on clustering that included a maximum number of known Asian events with no known false positives, and a lower bound based on the highest specificity consistent with maintaining a single Asian cluster (this level has about 10 percent known false negatives). These two definitions share virtually identical mean compositions, but differ significantly in range of variation. Within the test data the looser (upper bound) definition detects about 20 percent more samples than does the tighter (lower bound) definition.

[17] Since this analysis cannot address how well the April 1998 events represent all Asian events and because we cannot distinguish among the effects of measurement uncertainty, local contamination, and variation in source characteristics for the Asian dust, there is no definitive way to calculate classical confidence bounds on this analysis. However, a qualitative, nonparametric assessment (Figure 4) shows that the separation of populations within the test data is very strong.

[18] The compositional difference across the network between ''Asian'' and ''non-Asian'' events is illustrated in Figure 5, which shows the difference between the compositional heterogeneity when local sources dominate (the 4/15/98 data) and the broad uniformity found for the peak Asian mass event in the IMPROVE record (4/29/98).

[19] Analysis of the test data confirms that the April 1998 events conform to the first two assumptions of our logical model; the compositional data are broadly uniform and not masked by locally generated aerosols across the entire regional footprint of the archetypal events.

3.3.3. Other Continental Scale Dust Sources

[20] The last assumption in our analytical model (that the Asian dust be distinguishable from other dust sources) takes on critical importance in expanding the analysis to all of North America, since it is well known that non-Asian dust sources can have regional scale impacts in North America [*Gillette and Hansen*, 1989; *Cahill et al.*, 1994; *Perry et al.*, 1997; *Prospero*, 1999]. To confirm

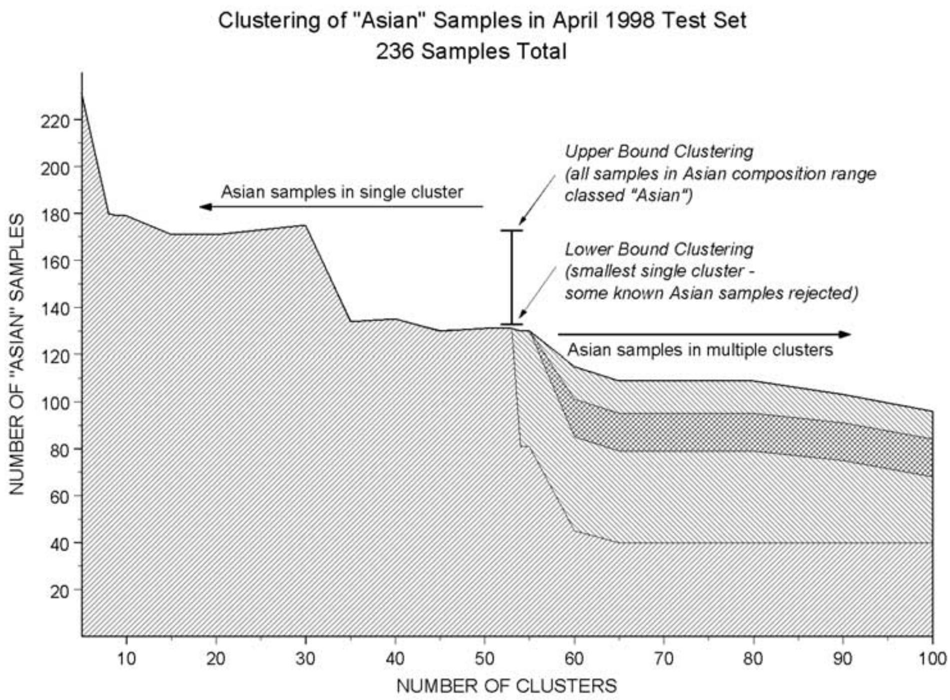

Figure 3. Clustering performance among the April 1998 test samples. At very low cluster numbers (below 8) all known Asian samples fall in a single cluster, but gather with them many that are probably not truly Asian. For cluster numbers from 8 to 30 the identified Asian sample population is stable (approximately 175 samples) as the non-Asian samples fill most of the new clusters. We interpret this first plateau as the upper bound of true Asian identity, although it may contain some ‘‘false positive’’ Asian identifications. At 30 clusters, the specificity drops to plateau again at 131 ‘‘Asian’’ samples for cluster numbers from 30 to 53. We interpret this lower plateau as the lower bound of Asian grouping within this analysis, since the known Asian samples are not confined to a single cluster when higher specificity (more than 53 clusters) is applied.

this critical assumption, we conducted two tests. First, we refined the method by testing it against the IMPROVE data for Virgin Islands National Park in the eastern Caribbean Sea. The Virgin Islands are known to be regularly exposed to dust from North Africa [*Perry et al.* 1997; *Prospero*, 1999], and their great distance from Asia and persistent easterly winds suggest that there should be minimal Asian dust there. Review of data presented by *Perry et al.* [1997] and additional analyses of the chemistry of both our 1998 sample set and IMPROVE data from African dust events at Virgin Islands showed that the Al/Ca and K/Fe ratios efficiently segregated Asian and African dust. *Perry et al.* [1997] report a definitive marker for North African dust is an Al/Ca ratio greater than 3.8; our April–May 1998 Asian test sample Al/Ca ratios are all below 2.6. We observed the K/Fe ratio to be consistently above 0.5 for all the April–May 1998 test samples, while African dust was generally below this level (since K is potentially increased by contamination with biomass smoke, this latter ratio can only be employed to set a minimum K fraction).

[21] Second, we tested the Asian signatures against data from southwestern desert IMPROVE sites. This region exhibits some degree of similarity in erosional and soil-forming processes to parts of the Takla Makan and Gobi deserts and we wanted to eliminate any false positives due to desert soils. Death Valley, California, a very arid site, was selected to detect any interference from desert soils and evaporites off nearby Manly playa or the very large upwind dust source at Owens Lake playa [*Cahill et al.*,

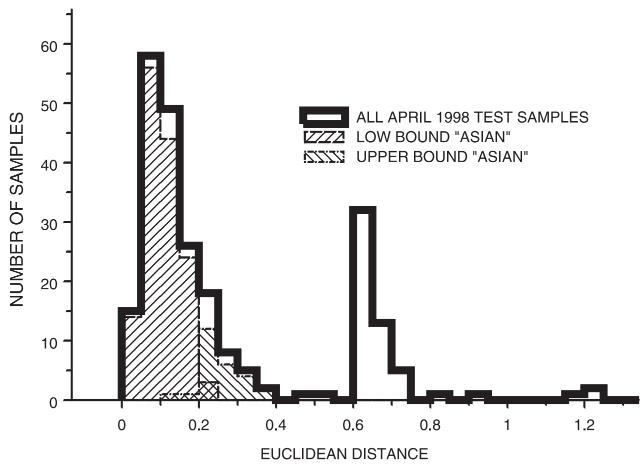

Figure 4. Distribution of Euclidean clustering distances from centroid of lower bound Asian cluster (April 1998 test data). The large difference between the Asian samples and most of the non-Asian samples is evident. Non-Asian samples within the Asian distance range failed elemental ratio tests for overlapping dust types (see section 3.3.3).

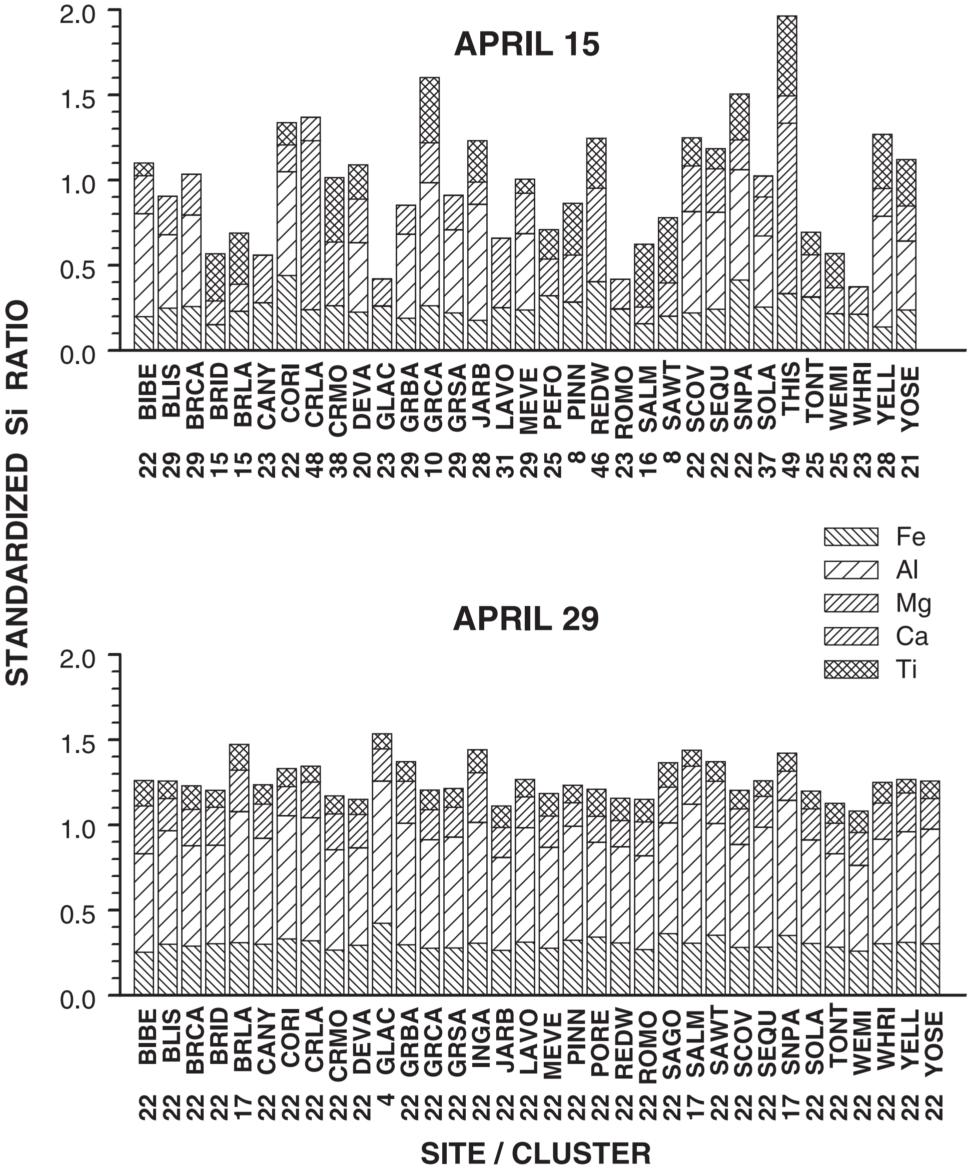

Figure 5. Standardized Silicon elemental ratios for western IMPROVE sites for a regional Asian dust event (4/29) compared to a "non-Asian" day (4/15). *x* axis labels are IMPROVE site codes (see section 3.5) and cluster numbers. Cluster 22 is the more stringent Asian definition (see section 3.3.2).

1994]. Big Bend Texas was selected to test for effects of arid-land grazing and soil disturbance in the surrounding Chihuahuan desert. Comparison of frequency distributions for 14 elemental ratios for putative "Asian" days at these desert sites to the data from the April–May 1998 test showed the expected excess Ca at both sites. Unlike the sharp distinctions in the North African dust comparison, however, there was significant overlap between paired elemental ratio distributions for the desert sites and the test data. Rather than risk misidentification, we decided not to calculate Asian dust statistics for sites in the American desert southwest.

3.4. Searching the Historical Record

[22] We devised algorithms that replicated the standardized ratios and Euclidean distance calculations of the test set and, using Euclidean distance limits and elemental ratio limits derived from the test data, applied them to search the historical aerosol records. Historical observations that fell within the compositional ranges of the test data Asian clusters and passed the exclusionary criteria for North African dust were then classified as "Asian."

3.5. Testing the Historical Record at Mauna Loa

[23] Mauna Loa was selected for testing because it has both an IMPROVE site and published data on its Asian dust exposure [*Harris and Kahl*, 1990; *Holmes and Zoller*, 1996; *Perry et al.*, 1999]. Applying the detection algorithm to the 510 samples in the Mauna Loa IMPROVE record, we detected 119 days that passed the more stringent low limit test, and an additional 53 days that passed the looser high limit test. There are two IMPROVE samplers at Mauna Loa; data reported here are for MALO1, which collected a 72-hour integrated sample beginning at midnight HST each

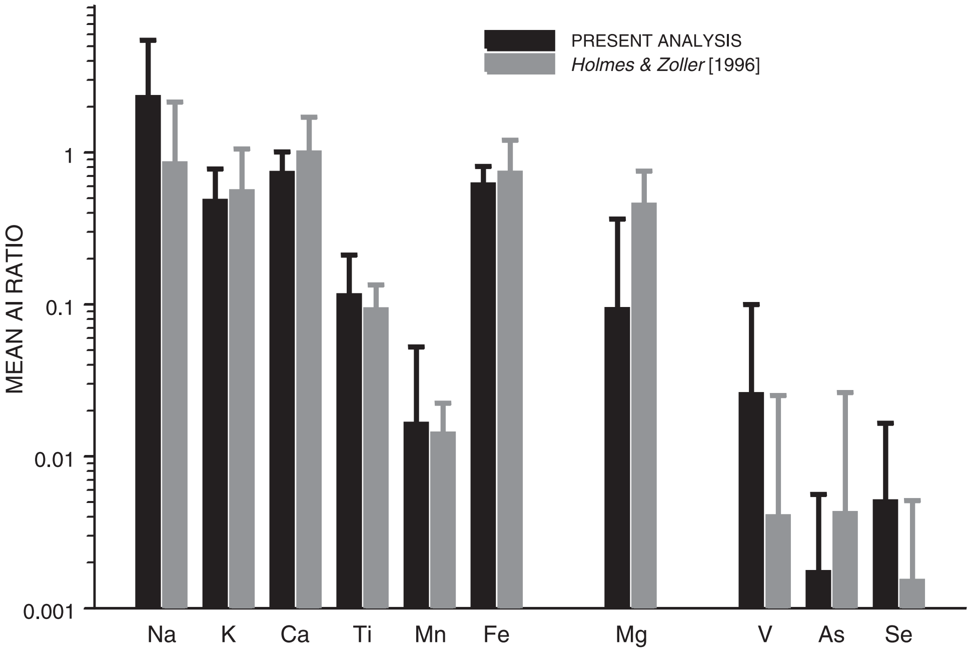

Figure 6. Comparison of mean Aluminum elemental ratios for Asian dust at Mauna Loa as reported by *Holmes and Zoller* [1996] for 250+ samples identified as Asian dust within a 12-year record of weekly dustfall samples (1970 through 1991) with Aluminum ratios from 119 IMPROVE samples collected between 1993 and 1999 and identified by the present method (stringent cut) as containing Asian dust. The plot shows all elements in common to the two data sets. Agreement is strongest among crustal elements (Na, K, Ca, Ti, Mn, Fe) and weaker for combustion-related elements (V, As, Se). Mg comparison is weak since Mg in these IMPROVE samples is frequently below detection limits.

IMPROVE sampling day (Wednesday and Saturday for the period analyzed here).

[24] To verify that our method correctly identified Asian dust independent of some artifact in the IMPROVE record, we compared mean and standard deviation for Aluminum-elemental ratios for our data and the Mauna Loa Asian dust characterization reported by *Holmes and Zoller* [1996]. The previously published ratios are for the Asian subset of a 12-year record of weekly samples taken from 1970 through 1991 (500+ samples, 250+ identified as Asian dust), analyzed by neutron activation. Our data are for 119 IMPROVE samples collected between 1993 and 1999 and identified by the present method (stringent test) as containing Asian dust (analysis by PIXE and XRF). Figure 6 shows all elements in common to the two data sets. Agreement is strongest among crustal elements (Na, K, Ca, Ti, Mn, Fe) and weaker for combustion-related elements (V, As, Se). Mg comparison is this case is inconclusive, since Mg in these IMPROVE samples is frequently below detection limits.

[25] In addition to the chemical similarity, our data replicate the seasonality at Mauna Loa as reported by *Bodhaine* [1983] and as represented in the analyses of *Holmes and Zoller* [1996] (February to June). Our seasonal data are in Figure 7, bottom left.

[26] The Mauna Loa tests demonstrate that our method satisfies the third and fourth assumptions of the analytic model, chemical and temporal consistency in the Asian dust. In addition, the Mauna Loa data also verify the mass-independence of our method: the 1998 events' mainland signatures appear to be chemically "typical" for Mauna Loa samples, despite up to twenty-fold higher concentrations than usually seen at Mauna Loa.

4. Results

[27] We present results for the 1989–1998 IMPROVE data in the form of monthly frequencies for Asian fine dust presence and monthly distributions of fine dust concentrations for selected groups of sites. For the eastern Pacific basin we show the low and high frequency estimates; for clarity, all other sites' frequencies reported here are the low end of the frequency ranges (i.e., the more stringent definition). We offer tentative explanations and interpretations of the patterns in the data for each group. As this is an exploratory look at a very large data set, we emphasize that we are confident of the general patterns of these results but strongly caution against over-interpretation of individual site's results or drawing strong inferences from single data points. While we believe that most Asian dust events are somewhat temporally smoothed in transit across the Pacific, and thus these data are representative of "typical" dust events, the IMPROVE samples actually represent 24-hour integration (72-hour at Mauna Loa), so that dust concentration variation within events is unknown, as are instantaneous peak dust concentrations.

4.1. Northeastern Pacific Basin

[28] Figures 7 and 8 show, respectively, our calculated frequency of Asian dust and the distribution of dust mass by month from the decade of IMPROVE data analyzed.

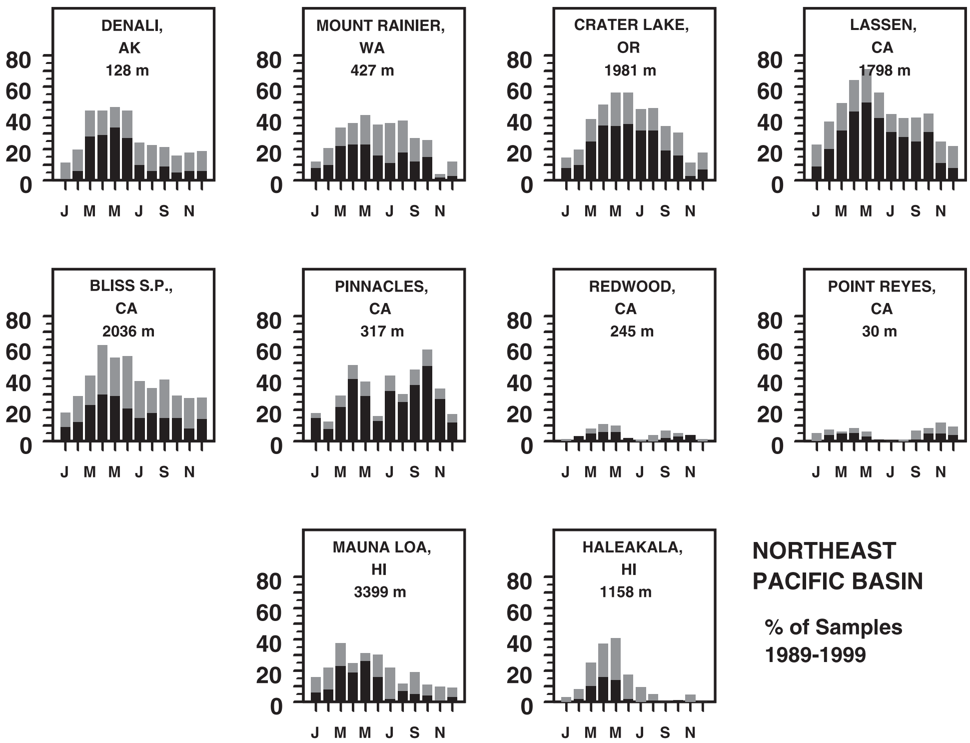

Figure 7. Monthly percent frequency of Asian fine dust (1989–1999) for selected IMPROVE sites around the northeast Pacific basin. Black and gray are low and high estimates as described in section 3.3.2. Mauna Loa data are from MALO1 3-day continuous samples.

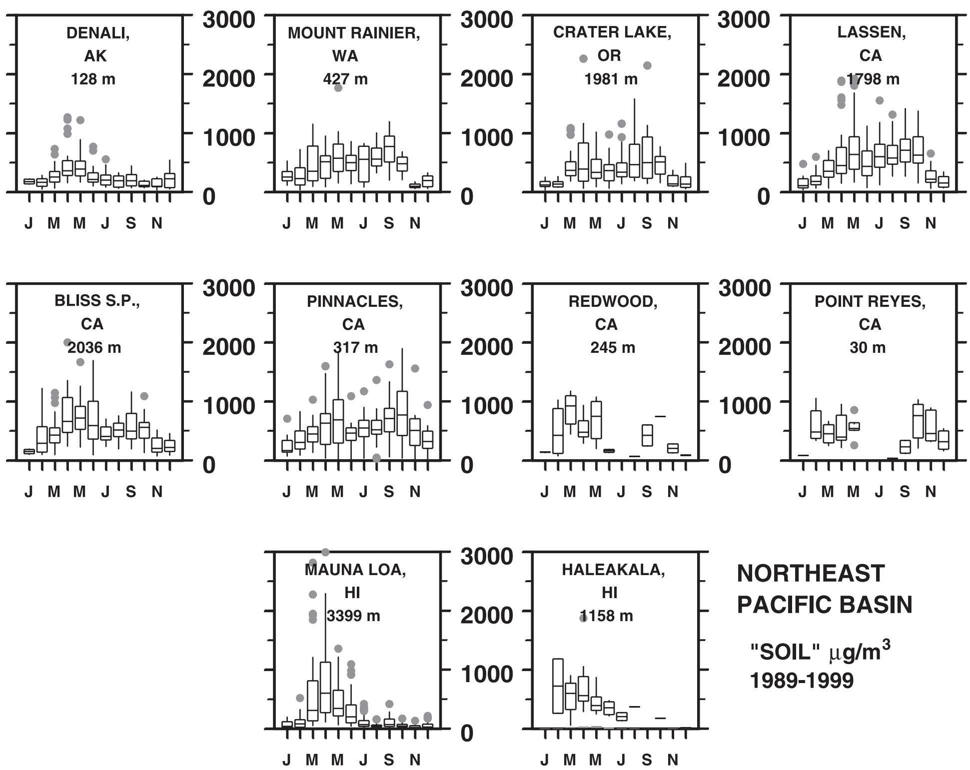

Figure 8. Monthly concentration of Asian fine dust for selected IMPROVE sites around the northeast Pacific basin. Mauna Loa data are from MALO1 3-day continuous samples.

4.2. Hawaii

[29] Mauna Loa, at 3499-m elevation shows the "conventional" springtime pattern as previously reported [*Harris and Kahl*, 1990; *Holmes and Zoller*, 1996; *Perry et al.*, 1999] and discussed above. Haleakala, situated at a modest 1158 m elevation shows the same temporal pattern, but at much lower frequencies across all months. The general seasonal pattern is consistent with processes described by *Merrill et al.* [1997]. We believe that the difference between these Hawaiian sites reflects a different frequency of Asian air mass exposure due to the fact that Haleakala is generally below, and Mauna Loa generally above, the persistent Trade Wind inversion. Situated near the Tropic of Cancer, Hawaii is generally isolated from midlatitude Asian sources by being within the zone of tropical easterly circulation, but from December to May the tropical wind bands are displaced to the south of Hawaii and strong westerly circulation can reach the islands to deliver Asian aerosols, especially at elevated Mauna Loa. The seasonality of tropical Pacific exposure to Asian aerosols is not unique to Hawaii; rather, the Hawaii data provide confirmation and context for other anecdotal observations such as reported for Eniwetok Atoll by *Duce et al.* [1980].

4.3. Alaska

[30] Denali, located at low altitude (128 m) on the northern slope of the Alaska range near Mt. McKinley, exhibits a temporal pattern much like that at Mauna Loa. Lacking additional subarctic sites for comparison, we cannot distinguish between horizontal and vertical gradients as factors in the seasonality of the Denali data, however the pattern is consistent with the reported south-north increase in seasonality of aerosol loading in Alaska [*Polissar et al.*, 1998].

4.4. California, Oregon and Washington

[31] In California, Oregon and Washington the larger number of sites available for study helps fill out the picture. The midlatitude Pacific coast sites exhibit very strong altitude sensitivity.

[32] For example, Point Reyes, California, is on the coastline just north of San Francisco, near sea level but protected from the local aerosols generated in the surf/coastal dune zone by a low ridge. While such a site might seem ideal for detecting transoceanic transport, in fact, our results suggest that it is effectively isolated from the free tropospheric transport zone except for sporadic contact during spring and fall. We believe this is due to Point Reyes being generally immersed in the stable marine layer that regularly overlies the cold California Current (see section 4.6).

[33] The picture of persistent Asian effect is very obvious at the higher Western Cordillera sites (Crater Lake in southern Oregon, Lassen in northern California, and Bliss in central California near Lake Tahoe, a span of nearly 500 km).

[34] There appears to be commonality of source for all these sites despite the frequency differences. Not only do the dust events share a common chemistry (set by the selection criteria), they also have a common distribution of concentrations, suggesting that location is relatively unimportant in dilution of the material. We interpret this structure in the data to suggest an "air mass" source and to effectively preclude a single North American source as an explanation for these data.

4.5. Continental Transect

[35] Figures 9 and 10 show frequency and mass concentration distributions for a suite of sites starting at the Pacific coast (Redwood National Park in California) and running along the northern tier of states to the Atlantic coast (Acadia National Park in eastern Maine). The western portion of this transect shows the Pacific marine layer effect and increasing Asian exposure with altitude, and a strong maximum at sites in the northern Rocky Mountains (see section 4.6). The spatial pattern suggests an increasing Asian exposure northward into Canada, but it may be an artifact of the suppression of local dust by the extensive forest surrounding the Rocky Mountain sites, making Asian samples less subject to dilution by local dust, and thus easier to detect.

[36] In the lee of the continental divide, Asian dust measured at surface sites is significantly reduced, with appreciable exposure limited to the deep mixing conditions of spring. Unfortunately, mountains (and thus IMPROVE sites) in the eastern United States are generally not high enough to reach the levels at which strong exposure is observed in the west. Based on data from a few sites near 1000-m elevation, both exposure and concentration appear to diminish weakly west to east. We also note that in New England, where the transport flow is offshore, there is no apparent marine effect on observed Asian aerosol exposure.

4.6. Atlantic Region

[37] Figures 11 and 12 show fine aerosol dust source climatologies for a string of sites ranging from sea level New England through the Appalachian highlands and southwards across the southeastern coastal plain, Florida, and into the Caribbean Sea. As discussed above, this region is exposed to long-range transport of fine dust from North Africa, with frequencies ranging from persistent in the south to rare, midsummer events in the north [*Perry et al.*, 1997; *Prospero*, 1999]. The plots show both the Asian and North African values extracted from the IMPROVE data. The contrast between the Asian and North African sources is dramatic. When it is present, North African dust appears at much higher concentrations than Asian dust throughout the region, but the Asian signature appears as a common, albeit infrequent, "background" even in the southernmost sites. While we were originally skeptical of these results, recent events have demonstrated that Asian aerosols can easily reach the western Atlantic Ocean and the northern Caribbean Sea. An enormous Asian dust storm originating in the deserts of China on 6–7 April 2001 appeared over the United States a few days later, with much notice in the press. When the cold front pushing the dust clouds passed out over the Atlantic on 19 April, it was imaged by the SEAWifs satellite. The U.S. Naval Research Laboratory, Monterey processed the imagery and meteorological data and produced a map separating the aerosols into sulfate, soil, and smoke. Imagery and maps are available at http://www.nrlmry.navy.mil/aerosol/Case_studies/20010413_epac/.

4.7. Vertical Structure of the Asian Plume

[38] The range of geographic settings of the IMPROVE sites permit inference of the spatial structure of the Asian

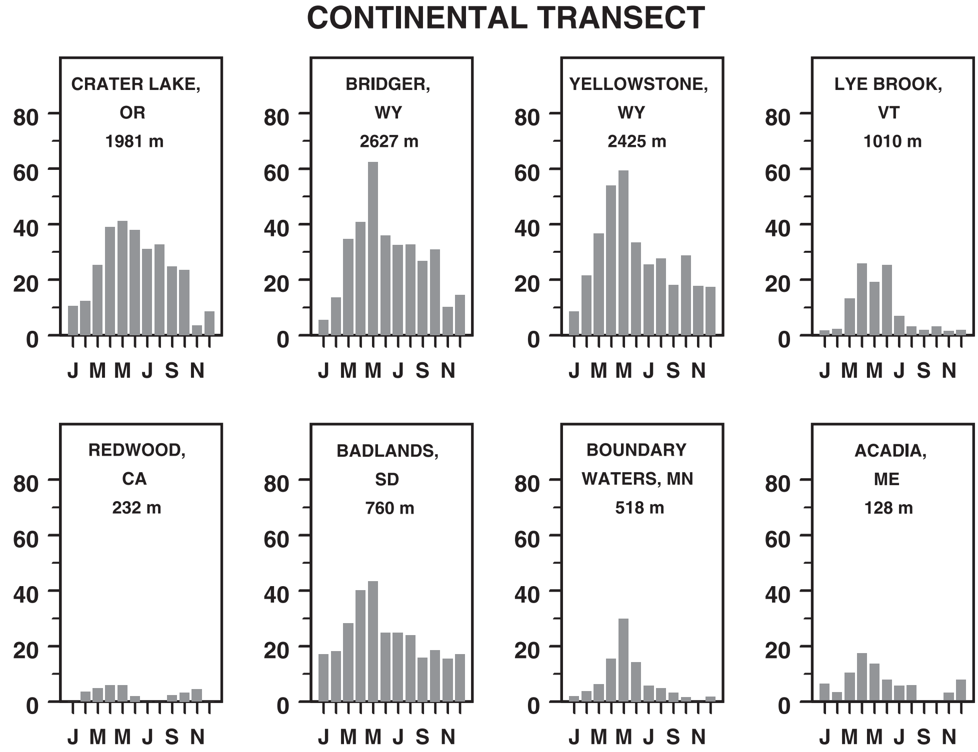

Figure 9. Monthly fine Asian dust frequency (percent of samples, stringent test) along a transect from the Pacific coast to New England. Montane sites on top, lowlands sites below.

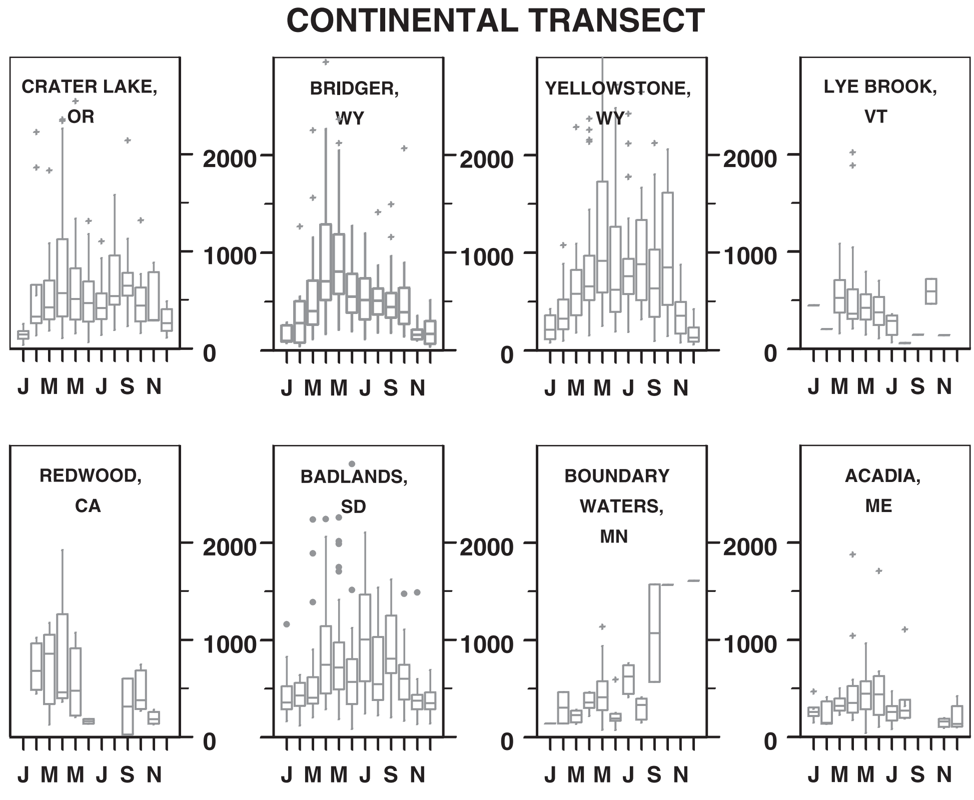

Figure 10. Monthly fine Asian dust concentration (ng/m^3) along a transect from the Pacific coast to New England. Low altitude sites east of the Rocky Mountains exhibit depressed frequency except in spring. Means are more meaningful than box dimensions for site-months with low numbers of events (e.g., September at boundary waters represents only two events in nine years).

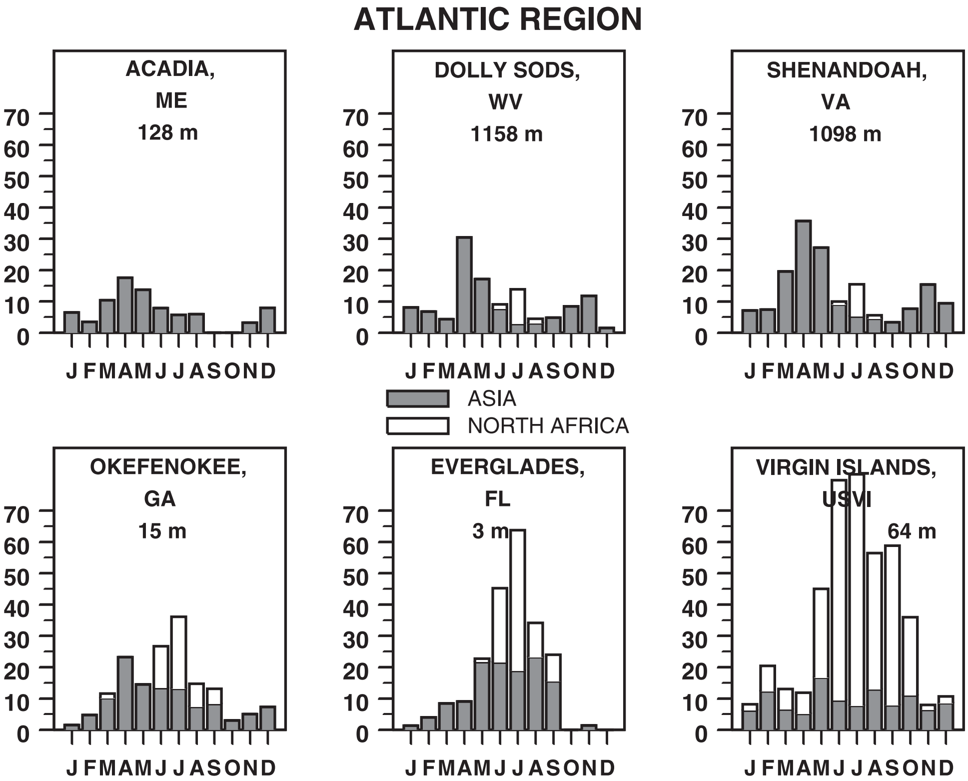

Figure 11. Monthly fine Asian and North African dust frequency (percent of samples) along a transect from New England to the Caribbean Sea. Gray represents Asian events; white represents North African events. Asian frequency drops southward, North African frequency drops northward. Also note different seasonality: Asian transport is broadly distributed with a spring peak, North African influence is summer-only except in the Caribbean Sea.

continental plume in three dimensions. Discussing the western coast sites, we posed an hypothesis of marine layer effect to explain the low frequency at coastal sites. This is supported by comparing data over a range of elevations. Figure 13 presents Asian dust frequency data and Asian and non-Asian Na concentration frequency distributions from Point Reyes (30 m elevation), Redwood National Park (245 m elevation, on the coastal slope in far Northern California), Pinnacles National Monument (317 m elevation, on the crest of the coast ranges in central California), Mt. Rainier, Washington (427 m elevation, on the flank of the mountain above the Puget Sound lowland), and Crater Lake (1963 m elevation, atop the Cascade crest in southern Oregon). Point Reyes and Redwood remain within the marine layer most days. The intermediate elevation sites (Pinnacles and Rainier) are alternately above and below the marine inversion. At Crater Lake marine air is infrequent and dilute. The Asian frequency and Na mass data show the expected pattern: nearly constant mineral aerosol Na mass on "Asian" days, decreasing sea-salt Na mass with altitude on non-Asian days, and increased Asian dust frequency with altitude.

[39] The data from high elevation sites in the northern Rocky Mountains suggests that concentrated Asian dust transport also has an upper limit, although much less firm than the lower bound created by the Pacific marine layer. The data presented in Figure 14 show no appreciable decrease in Asian dust frequency with altitude, but suggest a weakening of concentration, especially a loss of high concentration events and general lessening of mass outside the peak season. We believe this reflects the layered structure of Asian dust events in the western Pacific translated across the ocean basin (see discussion).

4.8. Summary of Findings

[40] The data presented here lead us to state the following findings. (1) The Asian dust that occasionally appears in high concentration over the northeastern Pacific has a unique chemical signature by which it can be traced in time and space. (2) Fine Asian dust events have a common source profile that makes multiple events recognizable by a common chemical signature over a wide range of mass concentrations. (3) Asian dust is transported across the North Pacific nearly continuously, except in winter. Our data suggest a winter hiatus in dust generation or transport, but do not preclude a plausible alternative interpretation that dust transported in winter is deposited by precipitation. Additionally, the apparent winter hiatus may be accentuated by operational limitations in the IMPROVE network (some missing data) due to the logistical difficulties of retrieving samples from elevated sites in winter. (4) In the eastern Pacific, Asian dust transport occurs predominantly above the marine boundary layer, but somewhat confined to the lower troposphere (a zone of 500–2500 m altitude). (5) Fine Asian dust (PM2.5) concentrations in the North Amer-

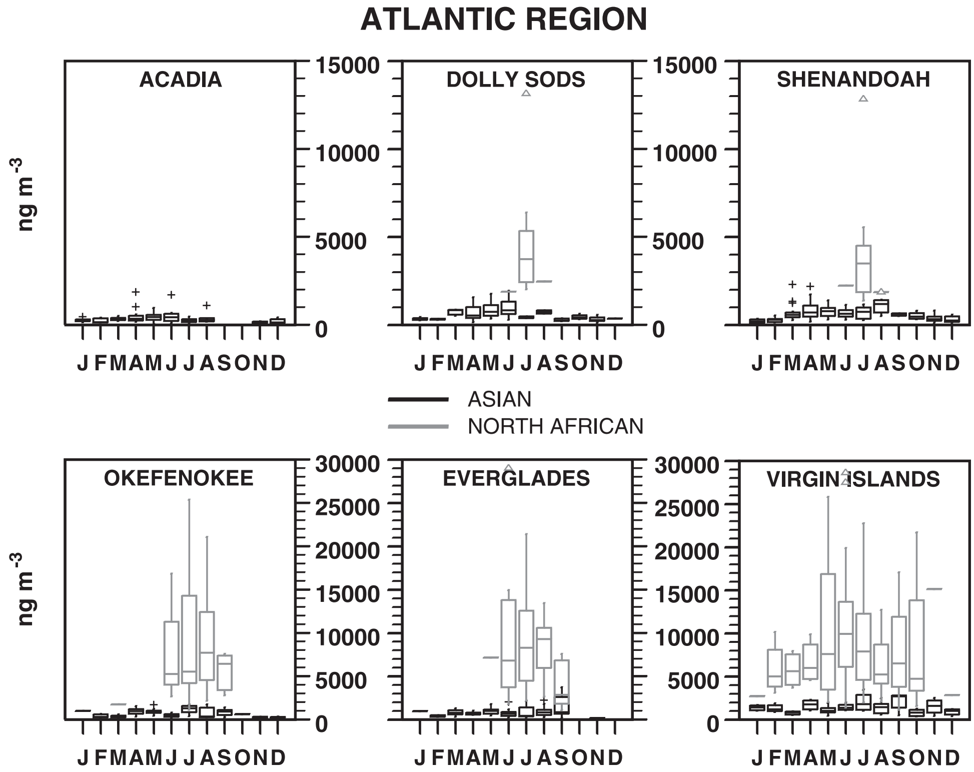

Figure 12. Monthly fine Asian and North African dust concentrations (ng/m^3) along a transect from New England to the Caribbean Sea. Asian and North African events are treated as mutually exclusive. Statistics for North African events are weak at the northern sites (e.g., June, July, and August North African data at Shenandoah represent one, seven, and one events, respectively, in eleven years).

ican troposphere generally range from 0.2 to 1 μg/m^3, infrequently exceeding 5 μg/m^3.

5. Discussion

[41] The subjective impression generated by infrequent sampling and widely reported extreme events is that Asian dust transport to North America is an episodic springtime phenomenon. Our examination of a decade-long record of fine aerosol data from the IMPROVE network contradicts that view, showing that Asian dust, and by implication accompanying air pollutants, are transported to North America throughout spring, summer, and fall. In addition, we find that Asian dust appears throughout North America, from the Caribbean to Alaska, not just on the Pacific coast. Such a departure from "conventional wisdom" begs the question of whether our concept of persistent and pervasive Asian influence can be supported by other evidence. While full treatment of these issues is far beyond the scope of this paper, we offer a sampling of supporting evidence from other sources. We proceed by addressing each of our findings.

1. Major Asian dust clouds can be recognized and tracked by their chemical composition. We believe that this is not controversial. Researchers in Asia have long noted that "yellow sand" events have a characteristic color and composition [*Chong and Yoon*, 1996; *Okada et al.*, 1990], and *Husar et al.* [2000] reported a distinctive chemical pattern for the April 1998 events that matched the spatial footprint of the dust as observed by satellite. Moreover, Asian dust should differ significantly from local dust within the fine fraction of the IMPROVE samples as an expected consequence of its transport. *Schutz and Rahn* [1982] showed that the mineral (and elemental) composition of crustal material varies with particle size, allowing the winnowing process (differential settling) of long-range transport to change the chemical composition of dust by eliminating coarse particles and increasing the relative fraction of platy and lower density crystals (micas, clays, etc.). *Braaten and Cahill* [1986] established that transported Asian soils in spring events at Mauna Loa contained significant fractions less than 0.1 μm diameter, which contrasts strongly with mass median diameter of 0.7 ± 0.1 for local Hawaiian dust reported by *Perry et al.* [1999]. Transported Asian dust is also much smaller than typical North American dust, the latter being almost entirely in the size range between 2.5 and 1.5 μm [*Raabe et al.*, 1988; *Cahill and Wakabayashi*, 1993].

2. Asian dust events have a common source. The repeated detection of the same chemical signature in a time series of aerosol samples can arise either from multiple events from a single source (our hypothesis) or independent impacts from multiple identical sources. Multiple lines of evidence suggest that the dust in "yellow sand" events comes from a limited geographic area [*Chong and Yoon* 1996; *Okada et al.*, 1990; *Sun et al.*, 2001].

3. Asian dust transport is nearly continuous. There are two related issues regarding the frequency of Asian transport across the Pacific. First, whether our interpreta-

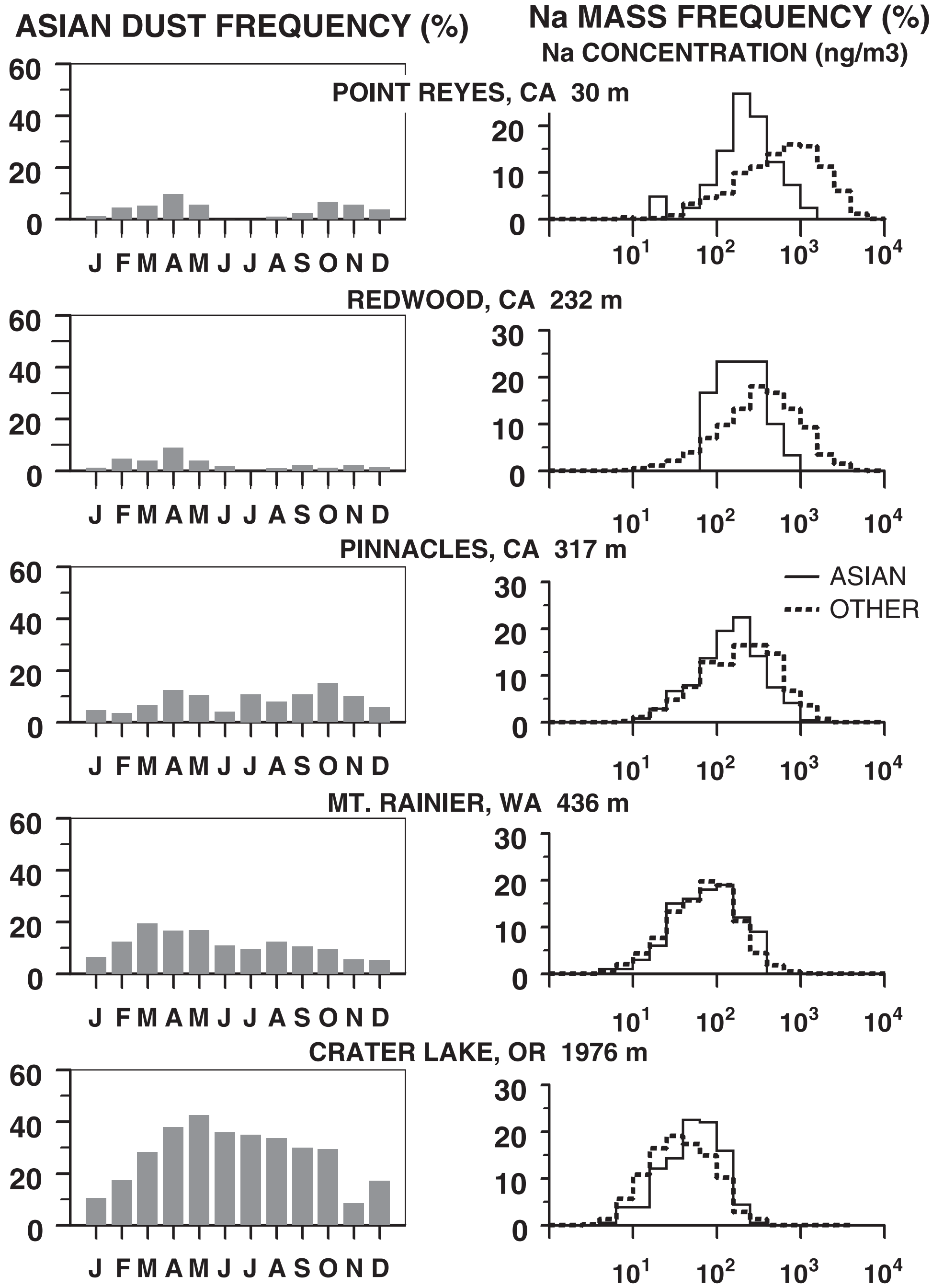

Figure 13. Frequency of Asian dust and sodium concentration distributions for "Asian" and "Non-Asian" days at eastern Pacific near-coastal sites. Frequency range for Mt. Rainier and Crater Lake is twice that of other plots. The Asian days show only a weak progression of decreasing Na concentration with altitude, probably due to meteorological selection (only strong Asian high pressure events penetrate to low altitude sites), while sea-salt aerosol ("non-Asian" curve) decreases rapidly with altitude. This is the pattern expected if our marine layer hypothesis is true that there is mutual exclusion between exposure to Asian air masses and the marine boundary layer.

tion of the aerosol data is consistent with Pacific basin meteorology, and second, whether the data from the decade of the 1990s are representative of long-term conditions. In addition to references cited above, we find particular support for the meteorological component in the work of *Yienger et al.* [2000], who used a general circulation model to track transpacific transport of gases for a year. That study found that transport at the surface (940-mb) was episodic, but that transport aloft was a regular synoptic feature of airflow over the North Pacific. This is consistent with the sustained high frequency and altitude-dependency found in the aerosol data reported here, as well as the published meteorological analyses [*Merrill et al.*, 1989, 1997]. Regarding possible changes in

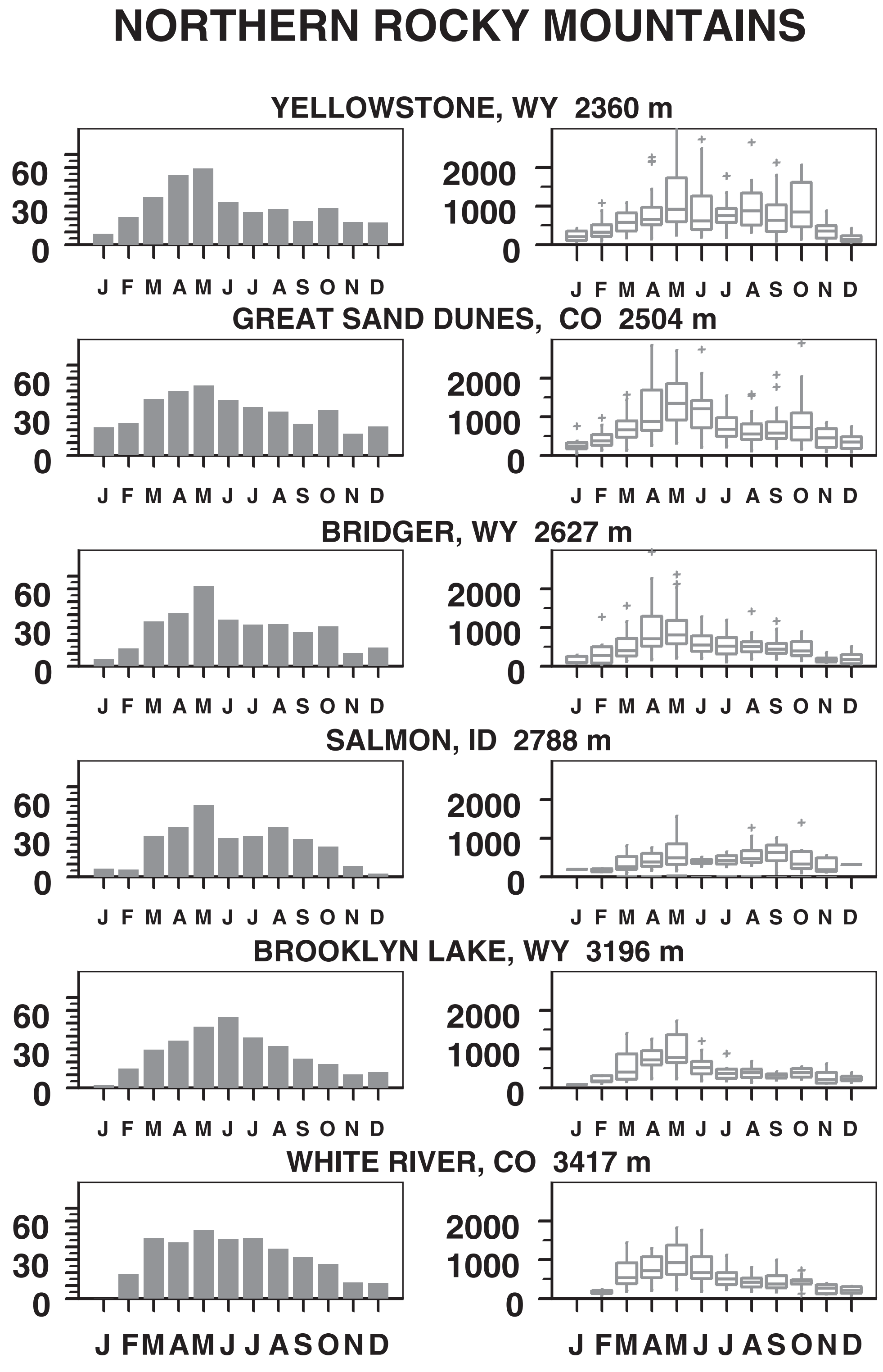

Figure 14. Asian frequency and dust mass concentration distributions for high altitude IMPROVE sites in the northern Rocky Mountains. Asian dust frequency is roughly comparable across all sites, while high concentration events appear to diminish with altitude. These data suggest an upper limit to concentrated transport somewhere between 2500 and 3000m MSL. While not definitive, these data comport with observations in the western Pacific (see section 4.6).

dust generation over a period of years, the picture is less clear. Changes in the relative mix of "natural" and anthropogenic dust sources, surface characteristics, soil moisture, and other factors can all modulate dust production. Some Chinese authorities claim that dust generation has been recently accelerated by a combination of drought and anthropogenic soil disturbance [*U.S. Embassy to the Peoples Republic of China*, 2001];

however, the modeling of *Wang et al.* [2000] suggests that local surface relative humidity, soil grain size, and wind speed are the controlling factors, in which case decadal or longer variation may be driven by a combination of anthropogenic, meteorological, and climatic processes. We are unable to resolve this latter question with the data at hand.

4. Regardless of the causes of interannual modulation of dust generation, Asian dust appears to have dominated fine dust in the middle to high latitudes of North America for millennia. *Biscaye et al.* [1997] reported that isotopic tracers and clay mineralogy for dust samples retrieved from the GISP2 ice core (Summit, Greenland, 2400 m elevation) and dated 23 ka to 26 ka (last glacial maximum) best fit with source material from the Gobi desert. While equally detailed mineralogical analysis of the later portions of the core have not been reported, physical analysis of dust from the nearby GRIP core [*Steffensen*, 1997] shows no obvious change in dust size distributions or other characteristics between glacial and Holocene times, and *Bory et al.* [2002] have recently reported that the modern dust at GRIP is of Asian origin. In addition, dust aerosols collected in modern firn from Baffin Island [*Zdanowicz et al.*, 2000] have also been attributed to Asian sources.

5. Asian dust is rare within the marine boundary layer. The vertical stratification of Asian fine dust found in the IMPROVE data probably represents a combination of processes. *Wang et al.* [2000] characterize dust generation in China as driven by high winds associated with cold fronts and report that dust transport follows an isentropic surface extending from the high elevation source areas eastward over the Pacific. This layered structure has also been observed by lidar in Japan [*Uematsu et al.*, 1983; *Sakai et al.*, 2000] and reconstructed from Korean Air Force pilot reports over Korea [*Chong and Yoon*, 1996]. These reports are fully consistent with the vertical pattern we see in the IMPROVE data around the eastern Pacific basin and into the Rocky Mountains.

6. Conclusion

[42] Our analyses of the IMPROVE data and collateral information from other studies establishes fine Asian dust as a persistent and pervasive component of the troposphere over the North Pacific and North America. These data will be better understood in the light of findings from short-term intensive studies such as ACE Asia (http://saga.pmel.noaa.gov/aceasia/AAIntro.html) and the Intercontinental Transport and Chemical Transformation 2002 program (ITCT 2K2; http://www.etl.noaa.gov/programs/2002/itct/), while this long-term record will help put the intensive data into proper perspective. Together, they will provide a guide to evaluating transport of anthropogenic pollutants into and across the Pacific. Fully understanding the geochemical, biological, and climatic implications of these findings will require additional research to separate the effects of meteorology, natural erosion, and human activity. Once these processes have been resolved, it will be possible to use data on contemporary Asian dust dynamics to enhance our understanding of the broader problem of the role of eolian processes in modern and quaternary dynamics of loess deposition, ocean fertilization, and radiative climate forcing.

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