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An
**observation-based
approach for local
dust identification**

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An observation-based approach to identify local natural dust events from routine aerosol ground monitoring

D. Q. Tong^{1,2}, M. Dan^{1,3}, T. Wang³, and P. Lee¹

¹US National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory, Silver Spring, MD 20910, USA

²Northeastern Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun, China

³Beijing Municipal Institute of Labor Protection, Beijing, China

Received: 8 December 2011 – Accepted: 26 January 2012 – Published: 7 February 2012

Correspondence to: D. Q. Tong (daniel.tong@noaa.gov), M. Dan (danmo2001@hotmail.com)

Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Dust is a major component of atmospheric aerosols in many parts of the world. Although there exist many routine aerosol monitoring networks, it is often difficult to obtain dust records from these networks, because these monitors are either deployed far away from dust active regions (most likely collocated with dense population) or contaminated by anthropogenic sources and other natural sources, such as wildfires and vegetation detritus. Here we propose a new approach to identify local dust events relying solely on aerosol mass and composition from general-purpose aerosol measurements. Through analyzing the chemical and physical characteristics of aerosol observations during satellite-detected dust episodes, we select five indicators to be used to identify local dust records: (1) high PM_{10} concentrations; (2) low $PM_{2.5}/PM_{10}$ ratio; (3) higher concentrations and percentage of crustal elements; (4) lower percentage of anthropogenic pollutants; and (5) low enrichment factors of anthropogenic elements. After establishing these identification criteria, we conduct hierarchical cluster analysis for all validated aerosol measurement data over 68 IMPROVE sites in the Western United States. A total of 182 local dust events were identified over 30 of the 68 locations from 2000 to 2007. These locations are either close to the four US Deserts, namely the Great Basin Desert, the Mojave Desert, the Sonoran Desert, and the Chihuahuan Desert, or in the high wind power region (Colorado). During the eight-year study period, the total number of dust events displays an interesting four-year activity cycle (one in 2000–2003 and the other in 2004–2007). The years of 2003, 2002 and 2007 are the three most active dust periods, with 46, 31 and 24 recorded dust events, respectively, while the years of 2000, 2004 and 2005 are the calmest periods, all with single digit dust records. Among these deserts, the Chihuahua Desert (59 cases) and the Sonoran Desert (62 cases) are by far the most active source regions. In general, the Chihuahua Desert dominates dust activities in the first half of the eight-year period while the Sonoran Desert in the second half. The monthly frequency of dust events shows a peak from March to July and a second peak in autumn from September to November. The large

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quantity of dust events occurring in summertime also suggests the prevailing impact of windblown dust across the year. This seasonal variation is consistent with previous model simulations over the United States.

1 Introduction

5 Due to its various effects on air quality and climate (Intergovernment Panel on Climate Changes, IPCC, 2007), dust aerosol lifted from disturbed soil have been extensively studied through ground observation, remote sensing and model simulations (Gillette and Passi, 1988; Gong et al., 2003; Reid et al., 2003; Zhang et al., 2003). For both remote sensing and modeling studies, ground measurements are always needed to verify
10 derived results. Specific ground-based monitoring networks have been established to facilitate dust detection (Zhang et al., 2003) and to assist in calibrating and improving aerosol models (Gong et al., 2003). In most cases, however, ground aerosol monitoring networks are deployed for other purposes, such as monitoring visibility (Pitchford and Malm, 1994) and protecting human health (Bell et al., 2007). Therefore, it is difficult to utilize these monitors to identify dust events because the monitoring sites are
15 either deployed far away from dust active regions (most likely collocated with dense population) or contaminated by anthropogenic sources. Even at rural or background sites, other natural sources, such as wildfires and vegetation detritus, and long-range transported dust can also contribute to monitor readings (e.g., Edgerton et al., 2009; Jaffe et al., 2004). Consequently, it is difficult to directly utilize the measurement data
20 from such monitoring networks to detect dust from local sources or to assess dust model performance. The regulatory monitoring networks, however, represent the majority of air quality monitoring around the world. The incapability of utilizing such a large set of data results in a missed opportunity to gain insight into dust activities from the perspective of “ground truth”.
25

Previous efforts have been made to identify dust contribution to ground samples. In the early years, radioactive elements, such as Radon-222, have been used as a tracer

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of dust transport from Africa (Prospero, 1970). In later studies, the mineral dust component in sampled aerosols was determined by the weight of ash residue from the high-temperature burning of sampling filter after being extracted with deionized water (Prospero, 1999). Wind and visibility data were also used to identify local windblown dust, but not chemical composition (Kavouras et al., 2007). In other dust composition studies, the definition of dust samples is derived from visual identification of dust events or other complementary measures (e.g., Kim et al., 2003). In most aerosol observations, however, the dust emission conditions or visual identification information are not available. Consequently, it is challenging to identify local windblown dust events based on particle concentration or chemical species because of the variability in meteorology conditions, dust strength and the distance from source areas.

We propose here a comprehensive analysis approach to identify local dust events relying on aerosol mass and composition from general-purpose aerosol measurement. During local dust storms, air samples demonstrate distinct physical and chemical characteristics, “fingerprints” that can be used to pinpoint these events based on element abundance and size distribution. The US Environmental Protection Agency (EPA) has established the Exceptional Event Rule (EER), which has been effective since March 23, 2007 (Federal Register, 2007). The EER has listed wind-blown dust emissions (the “high wind” event) as an exceptional event that could be excluded from being counted as a standard-violating exceedance. This approach we propose here is potentially useful to assist state and federal regulators in spotting dust occurrences in air quality management based solely on ground aerosol composition measurements. We apply this approach to scan the IMPROVE data from 2000 to 2007, and identify 182 local dust samples over 30 locations. A dataset of identified local dust events provide useful information for regulators to pinpoint natural dust events and researchers to verify remote sensing and atmospheric modeling results. In addition, the detailed chemical data collected during these identified dust events make it possible to determine the chemical composition of dust aerosols. The representation of chemically speciated dust aerosols allows atmospheric modeler to directly compare dust crustal and trace

elements with field measurements of individual aerosol elements. Atmospheric modelers, when equipped with such information, will be able to explicitly simulate the concentrations and deposition of critical nutrients (e.g., Iron) and toxic elements to study the climate, health and biogeochemical effects of dust aerosols.

2 Methodology

2.1 Approach to identify local dust records

This approach consists of several consecutive steps. First, we review the satellite data from the MODIS (Moderate Resolution Imaging Spectroradiometer) sensors to identify well-recorded large dust events that originate within the United States. Based on the time and location of these satellite detected storms, we obtain the ground measurement data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network in the Western United State. If there are valid IMPROVE measurements, these cases will serve as the dust “samples” to explore potential rules for identifying local dust aerosols. The second step of this approach is to examine the physical and chemical characteristics of the “known dust” samples. We are particularly interested in the following parameters: PM_{10} and $PM_{2.5}$ (particles smaller than 10 and 2.5 μm in diameter, respectively) mass concentrations, ratio of $PM_{2.5}$ to PM_{10} , percentage of crustal elements in $PM_{2.5}$, percentage of industrial, residential or biomass burning elements in $PM_{2.5}$, and enrichment factors of several crustal and anthropogenic elements.

The rationale behind choosing these parameters varies. In general, a dust event is associated with reduced visibility, resulting from increased levels of fine and coarse particles in the air (Malm et al., 1994). Therefore, $PM_{2.5}$ and PM_{10} concentrations during dust events are considerably higher than the typical levels. High PM concentrations, however, do not warrant a local dust event. For instance, long-range transported Asian and African dust has been previously reported to cause air quality degradation

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in both the Western and the Eastern United States (Prospero, 1999; Jaffe et al., 2004; Fairlie, et al., 2007). To ensure the source of dust aerosols is local, we exclude the high PM data that is also associated with high $PM_{2.5}/PM_{10}$ ratio. Field and laboratory measurements of freshly emitted soil dust aerosols reveal a low $PM_{2.5}$ to PM_{10} ratio, which increases as dust plumes age. The US EPA uses a value of 0.2 for $PM_{2.5}$ to PM_{10} ratio for soil dust emissions from human activities (Tong et al., 2012a). In this work, we remove the high PM data with the $PM_{2.5}/PM_{10}$ ratio higher than 0.35, considering these samples being contaminated with non-local dust sources. It should be noted that we consider all dust emissions from North American, including these from the Chihuahuan Desert in Mexico as local dust because the Southern Chihuahuan Desert is a frequent dust source for aerosols in the Southwestern US, especially Texas and New Mexico. The low $PM_{2.5}/PM_{10}$ ratio is also expected to exclude high PM concentration contributed by biomass burning, which is dominated by fine particles, resulting in a high $PM_{2.5}/PM_{10}$ ratio (Reid et al., 2005).

Because dust particles can be mobilized by both wind erosion and human activities, we apply three additional criteria to distinguish windblown dust from anthropogenic fugitive dust or other intensive aerosol types (such as volcanic ash, wildfire, or vegetative detritus). Soil dust aerosols are associated with abundant crustal elements, which differentiate them from aerosols from biomass burning, volcanic ash, or fossil fuel combustion. This feature alone, however, can not distinguish natural dust from anthropogenic fugitive dust. In the United States, anthropogenic fugitive dust is the largest sector of primary PM emissions. The five major fugitive dust sources in the United States are vehicle emissions from unpaved road (47%), paved road (7%), agricultural operation (29%), and construction (11%), and mining/quarrying (7%) (Tong et al., 2012a). Each of these sources involves either fossil fuel combustion or other human activities in the immediate vicinity of dust sources. Therefore, compared to natural dust, anthropogenic dust aerosols contain higher levels of anthropogenically originated elements, such as elemental carbon (from fossil fuel or biomass combustion) or heavy metals (such as Zn, Pd and Cu) from industrial operations (Chow et al., 2003; Reff

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et al., 2009). For instance, high levels of black carbon, Pb, Zn are found in paved road particles while high levels of nitrate (NO₃), Cr and Ni are found in unpaved road dust (Chow et al., 2003). Similarly, OC, K and Ca concentrations are high in animal husbandry dust and Ti, V, Mn concentrations in construction dust (Chow et al., 2003).

Therefore, we use the concentrations and enrichment factors (EFs) of anthropogenic pollutants as the indicators to distinguish natural dust from anthropogenic dust. In this study, the enrichment factors are calculated for a series of elements using Si as the reference element and the abundance of crustal elements at the Earth's surface as given by Taylor (1985),

$$EF_X = \frac{(X/Si)_{\text{aerosol}}}{(X/Si)_{\text{crustal}}} \quad (1)$$

where $(X/Si)_{\text{aerosol}}$ and $(X/Si)_{\text{crustal}}$ represent the ratio of a certain species (X) to Si in sampled dust aerosols and in the Earth's surface soil, respectively. Species with EFs close to unity are considered to have a strong natural origin, while species with higher EFs have mainly an anthropogenic origin. By examining the variation of the above parameters, we can establish useful criteria to be used in the subsequent statistical analysis to identify other local dust events that are not revealed by satellite data. Considering all the relevant parameters discussed above, five criteria will be the focus of subsequent statistical analysis: (1) PM₁₀ and PM_{2.5} concentrations; (2) Ratio of PM_{2.5} to PM₁₀; (3) Concentrations of crustal elements Al, Si, Ca, K, Mg, Fe; (4) Concentrations of anthropogenic pollutants, As, Zn, Cu, Pb, sulfate, nitrate, Organic carbon (OC), and EC; and (5) enrichment factors of anthropogenic pollution elements Cu, Zn, Pb and K.

Assuming reasonable homogeneity in dust chemical composition in a small region, we used the hierarchical cluster analysis to group all IMPROVE aerosol measurements based on the similarities in chemical and physical characteristics. Cluster analysis is a statistical method that creates clusters of items or objects that have similarity with the others in the same cluster but with differences between clusters. This technique has

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been previously applied to air quality studies to investigate source origins of air pollutants (e.g., Slanina et al., 1983; Dorling et al., 1992; Tong et al., 2005). As discussed earlier, dust episodes are usually extraordinary events with large perturbations in both aerosol concentrations and chemical composition compared to that during non-dusty periods. The cluster analysis is conducted using the statistical software SPSS Statistics 17.0 (SPSS Inc.). In the cluster analysis, the concentrations of Si, Ca, K, Fe, Ti, As, Cu, Pb, S, Zn and V, $PM_{2.5}/PM_{10}$ ratio, and the enrichment factors of Ca, K, Fe, As, Cu, Pb and Zn, are used to construct 6 clusters. The concentrations of four aerosol components, sulfate, nitrate, OC and EC are excluded from this analysis to avoid the unbalanced sampling issue, because a large portion of these data are either missing or invalid. In this study the hierarchical cluster analysis is configured with Between-Group Linkage clustering method, using Pearson Correlation to measure inter-cluster intervals. This method measures the correlations of x and y variables of case i according to the following formula (SPSS Statistics 17.0 Algorithms, SPSS Inc.):

$$C_{xy} = \frac{\sum_i (Z_{xi} Z_{yi})}{N} \quad (2)$$

$$Z_{xi} = \frac{Xi - \overline{X_N}}{\sqrt{\sum (Xj^2 - \overline{X_N}^2) / (N - 1)}} \quad (3)$$

Where C_{xy} the correlation between variable x and variable y , Z_{xi} and Z_{yi} are the standardized Z -score value of x and y for the case i , respectively, N is the number of cases, and $\overline{X_N}$ is the average value of x of the case i (x_i) for the N cases. The cases with higher correlation, which means higher similarity, are put into one cluster (or group).

2.2 Observational data

The aerosol observation data from the IMPROVE network were chosen for two reasons. The IMPROVE monitoring sites are predominantly deployed in the national parks

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and wilderness areas in the United State (Pitchford and Malm, 1994), including many sites in close proximity or downwind to major dust source regions. Secondly, the IMPROVE network is also one of the two national air quality monitoring networks that measure both mass concentrations and chemical composition of atmospheric aerosols.

There are other national or regional monitoring networks existing in the United States. The EPA Air Quality System (AQS) network has a national coverage, but there is no aerosol composition data available from this network. Another national aerosol monitoring network, the Chemical Speciation Network (CSN), is deployed mostly in urban areas, making it unsuitable for dust monitoring due to anthropogenic contamination. There are also some regional networks, such as the Southeastern Aerosol Research and Characterization Study (SEARCH), which measures aerosol mass and composition at both urban and rural sites (Edgerton et al., 2009). The currently operating eight SEARCH sites, however, are located in the Southeastern US and are too far away from major dust sources.

A subset of 68 sites from the IMPROVE network are used in this study. This subset of IMPROVE sites, deployed over in eight western states (Fig. 1), is chosen based on the findings in previous studies that have identified the geographical distribution of active dust sources in North America (Gillette and Passi, 1988; Malm et al., 2004; Van Curen, et al., 2002; Wells et al., 2007; Draxler et al., 2010). These regions are generally associated high wind power over barren land. The IMPROVE samplers have four modules designed to collect samples to measure PM_{10} and $PM_{2.5}$ mass concentrations, and $PM_{2.5}$ chemical components (Malm et al., 1994). These aerosol components include 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^{2-}) by ion chromatography (IC), organic to elemental carbon ratio (OC/EC) by staged thermal desorption and combustion, and total hydrogen by proton elastic scattering (PESA) (Malm, 2000). Fine soil in the IMPROVE data is calculated from the mass concentrations of five major soil-derived elements (Al, Si, Ca, Fe, K, and Ti) in their assumed oxides (Al_2O_3 , SiO_2 , CaO , K_2O , FeO , Fe_2O_3 ,

TiO₂, respectively) (Malm, 2000):

$$\text{Soil}_i = 2.2[\text{Al}] + 2.49[\text{Si}] + 1.63[\text{Ca}] + 2.42[\text{Fe}] + 1.94[\text{Ti}] \quad (4)$$

where [Al], [Si], [Ca], [Fe] and [Ti] are the measured concentrations of particulate aluminum, silicon, calcium, iron and titanium, respectively. All observational data for the period 2000–20007 are used in the subsequent analyses. All data flagged in the dataset for not attaining quality control standards were removed, with the exception of those flagged for moderate changes in flow rate. Data from the IMPROVE sites in the Central US are excluded from this analysis, since there are no major active dust sources in this region. According to the PM_{2.5} emission inventories, anthropogenic dust emissions also typically dominate the crustal aerosol composition in this region (Tong et al., 2012a).

Besides the IMPROVE data, satellite remote sensing of dust aerosols is used to independently identify local dust events. The Moderate resolution Imaging Spectroradiometer (MODIS) sensors aboard both Terra and Aqua have been making global daily observations of atmospheric aerosols since 2002 (Terra started in 2000). A total of seven wavelength channels (ranging from 0.47 to 2.13 μm) are used by MODIS to retrieve aerosol properties. Separate algorithms are developed for aerosol retrieval over land and ocean. Over the ocean, MODIS relies on the aerosol spectral signature from 0.55 to 2.13 μm to separate pollution particles (smaller in size) from coarse sea-salt and dust particles (Tanré et al., 1997). Over the land, MODIS uses the 2.1 mm channel to monitor surface-cover properties, and the visible wavelength to observe surface reflectance (Kaufman et al., 1997).

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3 Identifying windblown dust events

3.1 Analysis of satellite detected dust events

During the study period, satellite remote sensing has identified several large dust storms occurring in the United States. The purpose of analyzing these known dust events is to learn from these data of the distinct physical and chemical properties of local dust samples. The IMPROVE sampling protocol is to collect a 24 h duration sample every three days. Therefore, it is difficult for the ground monitors to capture all dust events identified by the satellite sensors. Meanwhile, because of limited temporal and spatial coverage, cloud contamination and high surface reflectivity over deserts, satellite remote sensing can not detect all dust events. Therefore, it is not easy to pinpoint a dust case that is simultaneously recorded by both satellite sensors and ground monitors. Here we focus on three such rare dust cases that were recorded by both ground and satellite observations on 15 April 2003, 27 November 2005 and 12 April 2007. The aforementioned 2003 dust storm was also reported by local media because of motorcyclist casualties caused by extremely low visibility over highways during the storm (Tong et al., 2012b). The MODIS imageries show that the three storms originated from different source regions. The former two storms were conceived from the Chihuahua Desert in Northern Mexico, while the latter one from the Mojave Desert in Southern California. By examining the MODIS imageries and the PM_{10} concentrations at ground monitors, we choose one or two IMPROVE sites that have captured a significant amount of dust aerosols at their samplers. These sites include the Guadalupe Mountains National Park, TX (GUMO1) site for the 15 April 2003 storm, the Big Bend National Park, TX (BIBE1) and GUMO1 sites for the 27 November 2005 storm, and the Death Valley National Park, CA (DEVA1) site and the San Geronio Wilderness, CA (SAGO1) site for the 12 April 2007 storm (Fig. 2).

The aerosol mass and chemical composition measurements at these monitors are then used to extract the commonality of typical local dust samples. In each case, we compare the observed concentrations of PM_{10} , $PM_{2.5}$, crustal (Si, Ca, Fe, K)

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and anthropogenic elements (Cu, Zn, Pb), sulfate, nitrate, OC and EC in $PM_{2.5}$, the $PM_{2.5}/PM_{10}$ ratio, the percentage of the above species and the enrichment factors of anthropogenic elements before, during, and after these dust episodes (Fig. 2). A few interesting patterns are shown in the aerosol samples collected during dusty periods:

(1) compared to that on non-dusty days, the PM_{10} concentration during a dusty day was elevated by 2–10 times from the pre-storm and post-storm levels; (2) although the concentration of $PM_{2.5}$ also increased during a dust storm, the $PM_{2.5}/PM_{10}$ ratio dropped significantly to approximately 0.2, a value typically representing freshly emitted soil particles (Tong et al., 2012a); (3) both the concentrations and percentage of crustal elements, including Si, Ca, Fe, and K, increased during dusty days; (4) The percentages of anthropogenic components in $PM_{2.5}$, including Cu, Zn, Pb, SO_4^{2-} , NO_3^- , and OC, all decreased from their corresponding pre-storm and post-storm levels, although the absolute concentrations may have increased or decreased depending on the site and the species. The concentration of EC during the dusty days was reduced to almost zero at all sites, but sulfate and nitrate concentrations reached their maximum values at the GUMO sites during the April 2003 and November 2005 dust storms; and (5) the silicon enrichment factors of Cu, Zn and Pb, which indicates anthropogenic contamination, decreased dramatically on dusty days.

Although the number of dust storms analyzed here is limited, the consistence of these patterns at all sites suggests that it may be feasible to identify local dust events through the use of routinely monitored aerosol parameters. Based on the observations above, we propose the following five indicators to be used to identify local dust records in the subsequent hierarchical cluster analysis: (1) high PM_{10} concentrations; (2) low ratio of $PM_{2.5}$ to PM_{10} ; (3) higher concentrations and percentage of crustal elements; (4) lower percentage of anthropogenic pollutants; and (5) low enrichment factors of anthropogenic elements.

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3.2 Cluster analysis

After establishing these identifying indicators, we use cluster analysis to test the hypothesis that there is one aerosol group, the local dust group, simultaneously matching all the above selection criteria. We perform hierarchical cluster analysis for all validated aerosol measurement data over the 68 study sites, using the concentrations of PM₁₀, PM_{2.5}, elements (Al, Si, Ca, Mg, K, Fe, Ti, As, Cu, Zn, Pb, V), sulfate, nitrate, OC, and EC, the PM_{2.5} to PM₁₀ ratio, the enrichment factors of K, Ca, Cu, Zn and Pb as the clustering criteria as discussed earlier. We found that at 30 of the 68 sites, there is one aerosol data group that demonstrates similar physical and chemical characteristics as observed in the previously satellite identified dust events. At the remaining sites, none of the IMPROVE data show any consistent pattern of dust events.

Figures 3 and 4 show the results of cluster analysis of all IMPROVE observation data from 2000 to 2007 at the GUMO1 site. The cluster analysis divides all data into six groups, and the first group has the highest PM₁₀ concentrations (Fig. 3a). The mean PM₁₀ concentration in this group is approximately 60 μg m⁻³, 3–10 fold of the typical background levels in the Western United States (Malm et al., 1994). The PM_{2.5} concentrations in this group are also higher than in other groups, although the differences among groups are relatively smaller than that for the PM₁₀ concentrations (Fig. 3b). The concentrations of the four crustal elements are significantly higher in group 1 than in other groups (Fig. 3c). During non-dusty days, the concentrations of these crustal elements in PM_{2.5} are low (less than 0.1 μg m⁻³) in most cases, except for Si the concentration of which reaches 1.0 μg m⁻³ occasionally. During dust storms, the Si concentration varies from 1.0 to 6.0 μg m⁻³. For the three trace metals that are mainly attributed to anthropogenic sources, their concentrations in group 1 is the lowest among all groups, but the difference is not as distinct as that of PM₁₀ or crustal elements (Fig. 3e), likely resulting from varying meteorological conditions and uneven distribution of emission sources. For the four major aerosol components (sulfate, nitrate, OC and EC), the distinction among these groups is further blurred. This

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is because these aerosol components can be contributed by both natural soil dust and non-dust sources (Fig. 3e). Finally, the data in group one have the lowest $PM_{2.5}/PM_{10}$ ratios. The $PM_{2.5}/PM_{10}$ ratio ranges from 0.1 to slightly above 0.3, with a mean of 0.2. The ratios in other groups have a wide range, from 0.1 to over 0.9 (Fig. 3f). The higher ratio reflects either higher contribution of anthropogenic sources and biomass burning, or the aging of an aerosol plume.

Figure 4 shows additional distinct physical and chemical characteristics of group 1 from other data groups. Not only the actual mass concentrations of crustal elements are higher in group 1, their relative abundance (in percentage) is also higher than in other groups (Fig. 4a). The opposite is true for the three anthropogenic trace metals, the percentage of which is the lowest among all groups. The enrichment factors for these metal elements are extremely close to unity in group 1, indicating their soil origin. In comparison, the silicon referenced enrichment ratios are much higher in all other groups, except group 4, which shows consistent low enrichment factors and higher crustal elements. This group, although not as clearly characterized as group 1, may represent similar soil dominated aerosol samples, such as smaller dust events or anthropogenic soil dust (such as from unpaved road or mining operation). Based on the consistent and distinct chemical and physical patterns that simultaneously meet the five stipulated criteria, we hence identify group 1 from the cluster analysis as the local dust aerosol group.

Figure 5 shows all identified dust events along the time series of $PM_{2.5}$ and PM_{10} concentrations at the GUMO1 site during the entire study period. Most of the high PM_{10} cases are identified by the cluster analysis as local dust events (highlighted with dashed circles), including the two large dust storms discussed in Sect. 3.1. However, there are other cases of high PM_{10} concentrations being excluded by the cluster analysis as local dust samples. These data are either associated with high $PM_{2.5}$ to PM_{10} ratios (long-distance dust transport or biomass burning) or with different chemical composition (i.e., aerosols originated from other sources).

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4 Summary of identified dust records

4.1 Summary of identified dust records

Cluster analysis of all aerosol data identifies a total of 182 dust records from 30 of the 68 sites (Table 1). These sites with dust records are also marked in red in Fig. 1. These 30 sites are located in the states of Texas, New Mexico, Arizona, South California, Nevada and Colorado. Such spatial distribution is consistent with the distribution of the four US Deserts, namely the Great Basin Desert, the Mojave Desert, the Sonoran Desert, and the Chihuahuan Desert. Outside the deserts, there are two sites in Colorado where previous model studies have found that high wind power in spring lifts surface soil grains (Gillette and Hansen, 1989; Tong et al., 2012b). Overall, the spatial distribution is similar to the dust source map reported in previous studies (Malm et al., 2004; Kavouras et al., 2007; Tong et al., 2012b).

Among the 30 dust sites, there are three sites, including Phoenix (PHOE1) and Douglas (DOUG1) in Arizona, and Fresno (FRES1) in California, that demonstrate distinct patterns in chemical composition. Although all located in arid or semiarid regions, these sites are also noticeably influenced by anthropogenic sources from nearby urban areas. In fact, a separate cluster has been identified for these sites, where the concentrations and percentage of primary anthropogenic pollutants such as Cu, Zn, Pb and EC, as well as their enrichment factors are much higher than at rural or remote dust source areas. The concurrent high levels of crustal and anthropogenic elements result from strong mixing of wind emissions and urban plumes, a unique setting for studying the interactions between dust and urban pollutants.

4.2 Temporal and spatial variability in dust events

The temporal variability of dust aerosols is also an interesting feature to air quality and climate modeling. Previous windblown dust studies, mostly relying on model simulations, predicted a springtime maximum over the North America (Gillette and Hansen, 1989; Tegen and Miller, 1998; Tong et al., 2012b). The “well-known” seasonal trend of

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local windblown dust in US, however, has not been independently evaluated against robust measurement data except for Kavouras et al. (2007), who used wind and visibility data to identify local windblown dust and investigated the seasonal trend.

The dust events identified in this work display large temporal and spatial variability. All dust cases from the three urban sites are excluded here because of their proximity to urban emissions. Figure 6 shows the interannual variations of local dust records in the five dust regions from 2000 to 2007. Although the IMPROVE monitors are not expected to capture all dust events, the dust records over these static sites nevertheless reflect the year-to-year change in dust activities over these areas. Unlike urban monitors, the IMPROVE monitors are distributed far away from each other. Therefore, the observed dust events are unlikely to overlap those detected at other locations. Only during extraordinarily large dust events, such as the 15 April 2003 storm, a dust plume can be detected by multiple IMPROVE monitors (four in this case) (Tong et al., 2012b) and the data at these sites are considered valid since all five filtering criteria are met.

During the eight-year study period, the total number of dust events displays an interesting four-year activity cycle. In the first cycle, the number of dust events increase from 8 per year in 2000 to 45 per year in 2003. In the second cycle, dust activities dropped to below 10 per year in 2004, and then persistently increase to 20 per year in 2007. It is not clear if such an interannual pattern exists in other years. The years of 2003, 2002 and 2007 are the three most active dust periods, with 46, 31 and 24 recorded dust events, respectively. The years of 2000, 2004 and 2005 are the calmest dust periods, all with single digit dust records.

Figure 6 also reveals the different activity patterns in different dust regions. The Chihuahuah Desert (59 cases) and the Sonoran Desert (62 cases) are by far the most active source regions. In general, the Chihuahuah Desert dominates dust activities in the first half while the Sonoran Desert in the second one (Fig. 6). The interannual trend is primarily driven by the dust activities from these regions. The Mojave Desert contributes 23 dust events during this period, while the Great Basin Desert and the Colorado Plateau contribute only seven and eight dust events, respectively.

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The dust records suggest clear seasonal variability in dust activities. The monthly frequency of dust events (Fig. 7) shows a peak from March to July and a second peak in autumn from September to November. Among all months, the highest number of dust records is in April, when the dust emissions in both the Chihuahua and Sonoran Deserts are most active. The month of May sees almost the same number of dust events as April, because the increase of dust activities in the Sonoran Desert can largely offset the diminished activities in the Chihuahua Desert. Actually, May 2003 is the month with the largest number of local dust records during the eight-year period, with 16 dust records obtained by 10 IMPROVE monitors there. The abundance of ground measurements during this period makes it an ideal case for a future dust modeling study over the United States. The peak dust season in the Chihuahuan Desert is about two months earlier than in the Sonoran Desert. The lowest number of dust events is found in January and August, during both periods dust activities were found only in the Mojave Desert. During the study period, there are eight sites that have observed more than eight local windblown dust events, with the GUMO1 site in Texas having the largest number (27) of dust records. In addition, the Queen Valley site in Arizona (QUVA1), the Big Bend site in Texas (BIBE1), the Salty Creek site in New Mexico (SACR1), the Chiricahua site (CHIR1), the Saguaro West site (SAWE1), and the Ike's Backbone site (IKBA1), all in Arizona, have captured 19, 16, 12, 9, 9, and 9 dust events, respectively. These monitors are either located in or downwind to the previously identified dust source regions associated with the geological characteristics of high soil erodibility (Kavouras et al., 2007).

4.3 Discussion

Several previous studies have attempted to identify potential dust source regions in the United States, which has been reviewed by Tong et al. (2012b). In general, wind erosion can occur over many land types, including deserts, dry lake beds, and exposed agricultural lands where fine and loose soil grains are available for wind lifting. The pioneer works by Prospero and colleagues have associated dust sources with barren

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Furthermore, the rich pool of aerosol chemical composition data associated with these identified dust records are useful to compile chemical profiles for emission splitting of dust aerosols. Recent advances of aerosol modeling (such as the latest version of the Community Multiscale Air Quality (CMAQ) model) require emission information of not only the mass flux and size distribution, but also the chemical composition of emitted dust particles. A companion paper (Dan et al., Chemical composition of natural dust particles in the United States, 2012) utilizes the data of the identified dust events and chemical composition to derive chemical profiles that could be used in future dust aerosol modeling works.

Our study reveals that dust events in the United States occur in almost all seasons, suggesting the prevailing impact of windblown dust across the year. This seasonal variation is consistent with previous model simulation over the United States (Gillette and Hansen, 1989; Park et al., 2010; Tong et al., 2012b). The windblown dust emissions peak in the spring, due to high wind speeds and a lack of vegetation cover over erodible land surface. The springtime maximum over North America was also reported in a long-term general circulation model study by Tegen and Miller (1998). Some previous study (e.g., Gatz and Prospero, 1996) has used the infrequency of summertime dust plumes to exclude the possibility of the impact of local dust on regional air quality. This work and several previous modeling studies suggest that it is possible to see summertime impact of dust aerosols originated from the Western United States.

5 Conclusions

Dust is a major component of atmospheric aerosols in many parts of the world. There are, however, very few monitoring networks that are exclusively designed and deployed to observe sand and dust particles. General-purpose aerosols monitoring networks, however, exist in a large number. The approach we propose here can utilize the general aerosol observations to identify local dust events. Using the publicly available IMPROVE data, we demonstrate how to use this approach to pinpoint 182 local dust records over 30 locations in the Western United States over a eight-year study period.

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The results presented in this study are subject to several limitations. The IMPROVE monitors are unevenly deployed over different dust source regions. Therefore, the observation-based dust data may not represent the overall emissions from each region. For instance, there are only five monitors in the Great Basin Dessert, and none is sitting in the heart of the barren land. The low number of recorded dust events (about one in each year) may be related to the sparse monitors in this region. Meanwhile, the IMPROVE monitors are more densely deployed over the Sonoran Desert and north Chihuahua Desert. The high number of dust records over these regions reflects both site density and active dust emissions.

Our approach involves both physical and chemical data of aerosol measurements, therefore requiring a comprehensive monitoring and analysis networks such as the IMPROVE program used in this study. In many cases, especially over major dust active regions over Africa and Asia, routine measurements of aerosol size distribution and chemical composition are not available. This limits the applicability of our approach to dust studies for these regions. A simplified approach, which uses only PM_{10} mass concentrations ($> 40 \mu g m^{-3}$) and the $PM_{2.5}/PM_{10}$ ratio (< 0.2) as the filtering criteria, is found reasonably effective to identify local dust events. It should be pointed out that, however, chemical fingerprint is still needed to assure the origin of measured aerosols. For example, the measurement data over the three urban sites can satisfy all selection criteria for local dust events, except the high levels of anthropogenic components. Such information reveals either human contamination of the dust aerosols, or human motivated dust sources (such as road dust from unpaved road, see Tong et al., 2012a; for more information). Regardless of its complexity, our proposed approach is likely to work most efficiently when all five identification criteria are concurrently applied.

Although our method specifically targets local dust samples, it can be easily extended to pinpoint other intermittent emission sources, such as long-range transported dust, volcanic ash, and biomass burning. Long-range transported dust is also associated with an increase in crustal elements. Compared to local dust events, the increase in mass concentration from the non-dusty level may be smaller, and the $PM_{2.5}/PM_{10}$

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Table 1. Identified local dust events from the IMPROVE monitoring network from 2000 to 2007. The concentrations and ratios listed in the table represent the mean values if there is more than one identified dust episode.

Site	SiteID	Longitude	Latitude	PM ₁₀ ($\mu\text{g m}^{-3}$)	PM _{2.5} ($\mu\text{g m}^{-3}$)	PM _{2.5} /PM ₁₀ ratio	Local dust events (YYMMDD)
1	BOAP1	-106.85	33.87	42.44	6.99	0.16	011016
2	GICL1	-108.24	33.22	35.59	7.71	0.22	070328
3	SACR1	-104.4	33.46	72.15	15.95	0.22	010410, 010925, 030415, 030602, 030605, 031226, 050311, 051203, 060312, 060619, 060622, 071114
4	WHIT1	-105.54	33.47	89.55	20.42	0.23	020426, 060216
5	BIBE1	-103.18	29.3	53.25	12.36	0.24	000322, 000422, 000921, 010209, 011124, 020210, 020309, 020312, 020330, 020402, 020616, 021110, 030328, 030406, 030415, 051127
6	GUMO1	-104.81	31.83	73.20	15.63	0.22	000422, 000517, 010422, 010603, 010624, 010715, 011016, 020309, 020420, 020502, 020511, 020526, 020610, 020613, 020619, 030202, 030304, 030415, 030418, 030515, 030723, 031208, 031226, 040608, 051127, 060318, 070223
7	CHIR1	-109.39	32.01	73.34	17.05	0.24	000408, 011109, 030521, 030717, 051127, 060601, 060716, 061222, 070328
8	IKBA1	-111.68	34.34	62.53	18.76	0.29	010621, 020514, 030515, 030521, 030530, 030726, 040903, 070412, 070720
9	QUVA1	-111.29	33.29	61.20	13.64	0.22	011016, 020426, 020514, 030202, 030515, 030521, 030617, 030620, 030714, 030717, 030909, 041021, 060216, 060414, 060716, 060725, 070418, 070708, 071018
10	SAGU1	-110.74	32.17	57.79	18.13	0.31	011109, 030521, 030717, 060625, 070328, 070412
11	SAWE1	-111.22	32.25	75.59	20.23	0.25	011109, 030521, 030711, 030717, 030909, 070328, 070412, 070415, 070521
12	SIAN1	-110.94	34.09	59.60	17.22	0.3	011016, 030515, 030530, 070412, 060716
13	TONT1	-111.11	33.65	61.60	13.89	0.23	060716, 070412, 070708, 070720, 071006
14	PEFO1	-109.769	35.07	55.40	13.00	0.24	030509, 050404, 050419
15	AGTI1	-116.97	33.46	72.46	13.69	0.19	010817, 021125, 030106, 070412
16	DEVA1	-116.85	36.51	63.95	12.00	0.19	020508, 020511, 020520, 040903, 061228
17	DOME1	-118.14	35.73	65.60	6.86	0.1	031030
18	JOSH1	-116.39	34.07	69.56	15.97	0.27	000812, 011001, 020731, 030819, 050802, 060625
19	SAGA1	-118.03	34.3	45.12	6.43	0.14	021002
20	SAGO1	-116.91	34.19	71.09	10.97	0.16	021125, 070412, 070521
21	SEQU1	-118.83	36.49	78.61	10.06	0.16	020710, 031030
22	HOOV1	-119.18	38.09	149.29	45.76	0.31	020228
23	GRBA1	-114.22	39.01	104.62	18.85	0.18	020228
24	WARI1	-118.82	38.95	70.39	12.85	0.19	030921, 040310, 040903, 050916, 050922
25	INGA1	-112.13	36.08	107.08	32.39	0.3	070720
26	GRSA1	-105.52	37.72	51.28	11.10	0.23	000517, 020511, 030503, 050603
27	MEVE1	-108.49	37.2	65.20	13.68	0.22	030202, 030415, 050419
28	DOUG1	-109.54	31.35	81.27	21.20	0.26	070328, 071108
29	PHOE1	-112.1	33.5	76.82	15.93	0.21	011016, 020511, 020722, 020917, 030202, 030515, 030530, 030714, 030717, 030909, 060405, 060414, 060625, 070412, 070720
30	FRES1	-119.77	36.78	88.88	16.65	0.19	040915, 060914, 060929, 061026, 070912

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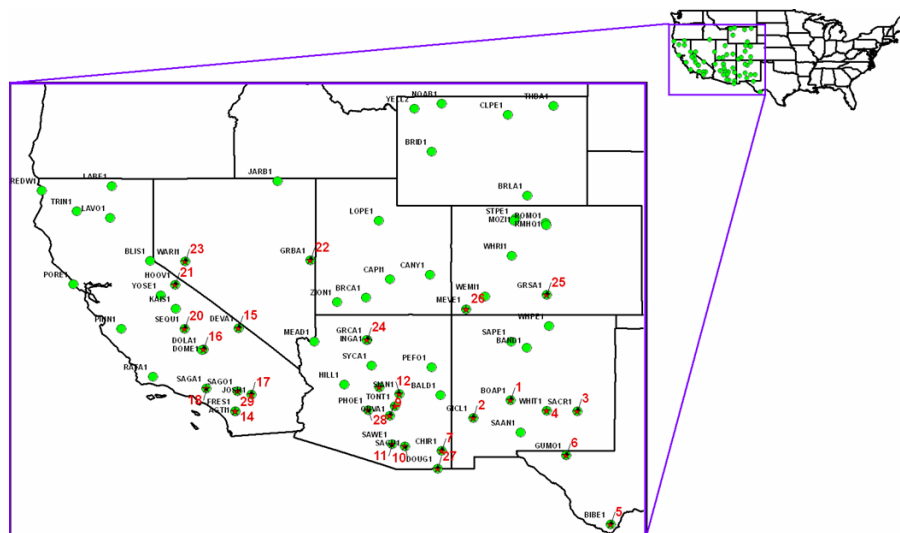


Fig. 1. Locations of the 68 selected IMPROVE monitors from which the aerosol observation data are used in this study. The 30 sites (marked in red) indicate the locations where at least one local dust storm has been identified between 2000 and 2007 using the approach proposed in this work.

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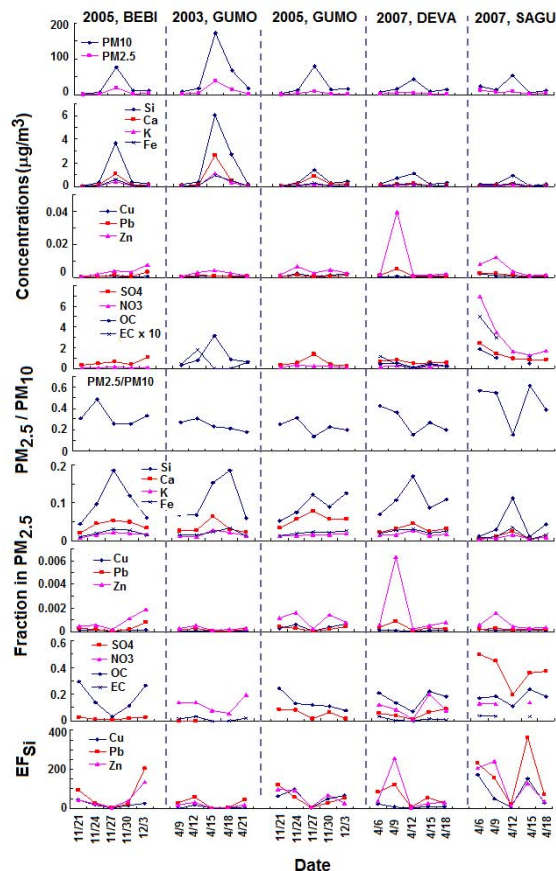


Fig. 2. Variations of PM_{10} , $PM_{2.5}$ and chemical components of $PM_{2.5}$ at the BEBI1, GUMO1, DEVA1 and SAGU1 sites during, before and after three dust storms. These dust events have been pinpointed by MODIS satellite data.

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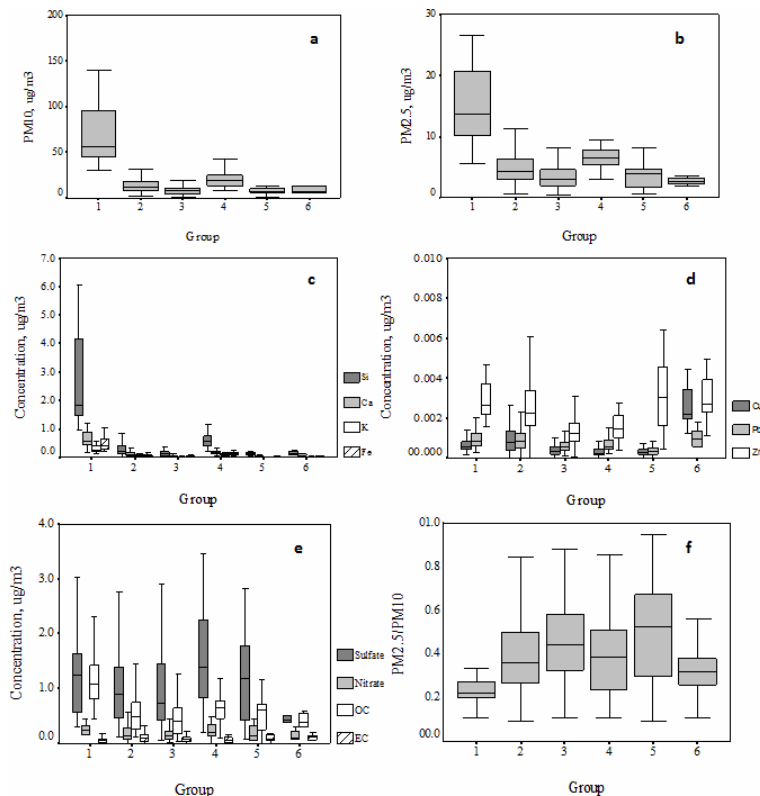


Fig. 3. Physical and chemical characteristics of aerosol samples in different clusters as generated by the hierarchical cluster analysis of all IMPROVE observation data from 2000 to 2007 at the GUMO1 site: **(a)** PM₁₀ mass; **(b)** PM_{2.5} mass; **(c)** crustal elements, Si, Ca, K, Fe; **(d)** anthropogenic trace elements, Cu, Zn, Pb; **(e)** Sulfate, Nitrate, OC and EC; and **(f)** PM_{2.5}/PM₁₀ ratio. Group 1 was identified as the local dust group.

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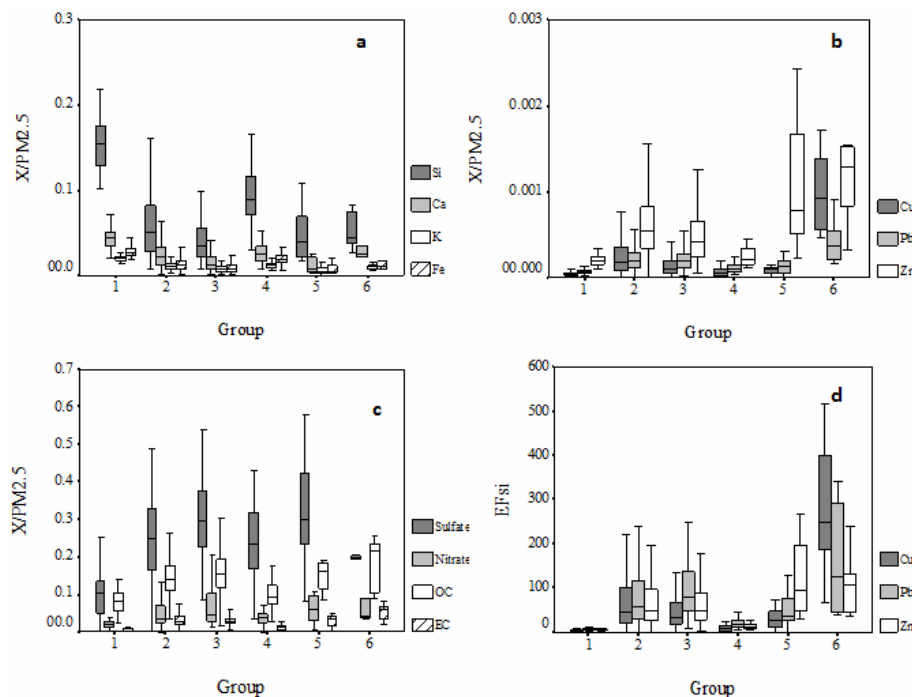


Fig. 4. Physical and chemical characteristics of aerosol samples in different clusters as generated by the hierarchical cluster analysis of all IMPROVE observation data from 2000 to 2007 at the GUMO1 site (continued): **(a)** mass fractions of Si, Ca, K, and Fe in $PM_{2.5}$; **(b)** mass fractions of Cu, Zn, and Pb in $PM_{2.5}$; **(c)** mass fractions of Sulfate, Nitrate, OC and EC in $PM_{2.5}$; **(d)** enrichment factors of Cu, Zn and Pb using Si as the reference element between different groups classified by cluster analysis. Group 1 was identified as the local dust group.

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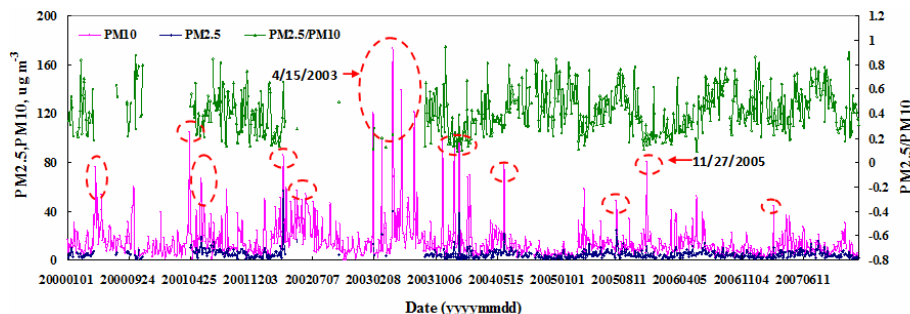


Fig. 5. Time series of PM_{10} and $PM_{2.5}$ mass concentrations and their ratio at the Guadalupe Mountains National Park, TX (GUMO1) site between 2000 and 2007. Red circles indicate local dust events identified using the dust identification approach. The approach has effectively captured all satellite pinpointed dust events, including the 15 April 2003 storm and the 27 November 2005 storm.

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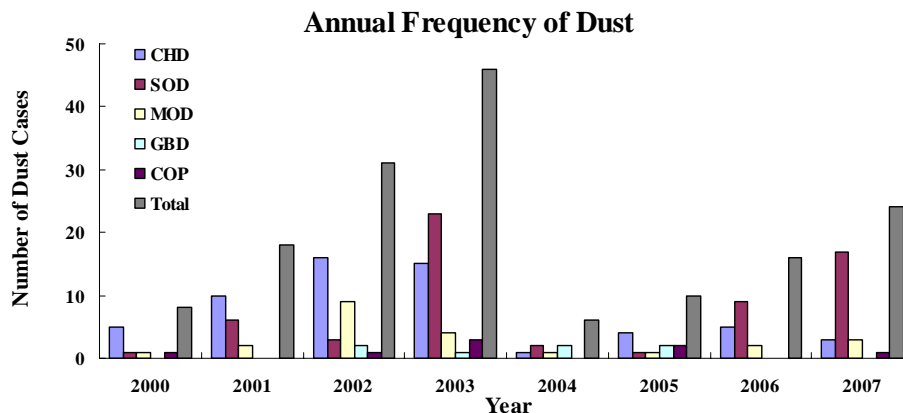


Fig. 6. The annual frequency of local dust cases in the five deserts regions from 2000 to 2007. The dust regions are Chihuahuan Desert (CHD), Sonoran Desert (SOD), Mojave Desert (MOD), Great Basin Desert (GBD), and Colorado Plateau (COP).

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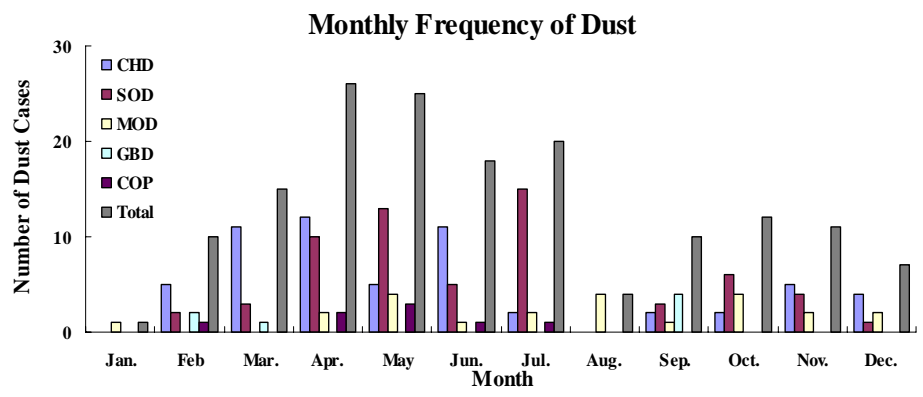


Fig. 7. The monthly frequency of local dust cases in the five deserts regions from 2000 to 2007.

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