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2	Long-term dust climatology in the western United States reconstructed from routine
3	aerosol ground monitoring
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#### 32 Abstract:

This study introduces an observation-based dust identification approach and applies this 33 method to reconstruct long-term dust climatology in the western United States.Long-term dust 34 climatology is important for quantifying the effects of atmospheric aerosols on regional and 35 36 global climate. Although there exist many routine aerosol monitoring networks, it is often 37 difficult to obtain dust records from these networks, because these monitors are either deployed 38 far away from dust active regions (most likely collocated with dense population) or contaminated 39 by anthropogenic sources and other natural sources, such as wildfires and vegetation detritus. 40 Here we propose an approach to identify local dust events relying solely on aerosol mass and composition from general-purpose aerosol measurements. Through analyzing the chemical and 41 42 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 43 44  $PM_{2.5}/PM_{10}$  ratio; 3) higher concentrations and percentage of crustal elements; 4) lower percentage of anthropogenic pollutants; and 5) low enrichment factors of anthropogenic elements. 45 After establishing these identification criteria, we conduct hierarchical cluster analysis for all 46 validated aerosol measurement data over 68 IMPROVE sites in the western United States. A 47 48 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 U.S. Deserts, namely the Great Basin Desert, the 49 50 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 51 52 an interesting four-year activity cycle (one in 2000-2003 and the other in 2004-2007). The years 53 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 54 single digit dust records. Among these deserts, the Chihuahuan Desert (59 cases) and the 55 Sonoran Desert (62 cases) are by far the most active source regions. In general, the Chihuahuan 56 57 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 58 second peak in autumn from September to November. The large quantity of dust events 59 60 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. 61

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63 *Keywords:* Dust, aerosols, climatology, air quality, IMPROVE, drought

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## 65 Introduction

Due to its various effects on air quality and climate (Intergovernment Panel on Climate 66 Change, IPCC, 2007), dust aerosol lifted from disturbed soil has been extensively studied 67 through ground observation, remote sensing and model simulations (Gillette and Passi, 1988; 68 Gong et al., 2003; Reid et al., 2003; Zhang et al., 2003). For both remote sensing and modeling 69 studies, ground measurements are critically important for verifying derived results. Specific 70 ground-based monitoring networks have been established to facilitate dust detection (Zhang et 71 72 al., 2003) and to assist in calibrating and improving aerosol models (Gong et al., 2003). In most 73 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). 74 75 Therefore, it is difficult to utilize these monitors to identify dust events because the monitoring sites are either deployed far away from dust active regions (most likely collocated with dense 76 population) or contaminated by anthropogenic sources. Even at rural or background sites, other 77 78 natural sources, such as wildfires and vegetation detritus, and long-range transported dust can 79 contribute to monitor readings (e.g., Edgerton et al., 2009; Jaffe et al., 2004). Consequently, it is difficult to directly utilize the measurement data from such monitoring networks to detect dust 80 81 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 82 83 utilizing such a large set of data results is a missed opportunity to gain insight into dust activities from the perspective of "ground truth". 84

85 A myriad of observation-based methods have been proposed to identify dust events using satellite observation, computer models and ground and laboratory measurements. These methods 86 87 vary in complexity and applicability, but in general fall into three categories: laboratory-based 88 approach, and remote sensing-based approach and ground monitor-based approach. In the early years, radioative elements, such as Radon-222, have been used as a tracer of dust transport from 89 Africa (Prospero, 1970). In later studies, the mineral dust component in sampled aerosols was 90 91 determined by the weight of ash residue from the high-temperature burning of sampling filter 92 after being extracted with deionized water (Prospero, 1999). Another laboratory study differentiated dust particles from other types of transportable particles collected on board the 93 94 NOAA Research Vessel Ronald H. Brown through individual-particle analysis using an

automated scanning electron microscope (SEM) and a field emission scanning electron
microscope (FESEM) (Gao et al., 2007).

97 With the rapid expansion of remote sensing data, several studies have attempted to detect dust outbreaks using satellite images and other derived products (Kauffman et al., 2000; 98 Prospero et al, 2002; Rivera-Rivera et al., 2010; Lee et al, 2009). The pioneer works by Prospero 99 and colleagues have associated dust sources with barren areas with "depressed" elevations 100 101 relative to their surroundings (Ginoux et al., 2001) based on satellite-based global observations from the NIMBUS 7 Total Ozone Mapping Spectrometer (TOMS) (Prospero et al., 2002). They 102 found that the major dust sources are invariably associated with topographical lows in arid or 103 104 semiarid regions with rainfall below 250 mm (Prospero et al., 2002). A recent work by Ginoux et al (2010) combines land use data with the Moderate Resolution Imaging Spectroradiometer 105 (MODIS) Deep Blue algorithm to identify natural and anthropogenic dust sources over the 106 western Africa. This approach is further developed to pin-point active dust sources in the North 107 America by selecting grid cells based on the frequency of high aerosol optical depth (AOD) 108 events (AOD = 0.75) (Draxler et al., 2010). In an effort to quantify the relative impacts of 109 110 Saharan and local dust in Elche in Southeastern Spain, Nicolas et al. (2008) combined satellite images from the NASA SeaWiFS, two dust prediction models (NAAPS and DREAM), a back-111 112 trajectory model (HYSPLIT) and NCEP meteorological reanalysis data to detect the outbreaks of African dust events. Using Positive Matrix Factorization (PMF), they identified six PM<sub>10</sub> 113 114 sources, including local soil and African dust, which are distinguished by the correlation of the source intensity with Ti. In Asia, an operational dust retrieval algorithm has been developed 115 116 based on the FY-2C/SVISSR through combining visible and water vapor bands observations of the geostationary imager to distinguish dust plumes from surface objects and clouds (Hu et al., 117 118 2008). In the United States, data from both polar-orbiting and geostationary satellites have been used to characterize source areas of large dust outbreaks (Lee et al., 2009; Rivera-Rivera et al., 119 120 2010). It should be mentioned that all of these dust source identification methods are based on satellite remote sensing that needs to be independently verified using ground observations. For 121 122 instance, Schepanski et al. (2007, 2012) combined a back-tracking method with high temporal 123 satellite aerosol data (15-min Aerosol Index (AI) from the Ozone Monitoring Instrument (OMI)) to identify dust sources over the Saharan region. They found that the spatial distribution of dust 124

source areas inferred from OMI 15-min AI is distinctly different from that by using the dailyMODIS Deep Blue aerosol data (Schepanski et al., 2012).

127 Beside these laboratory and remote sensing studies, dust identification methods have also been developed based exclusively on aerosol mass concentration and its correlation with 128 meteorological conditions. Kavouras and co-workers (2007) developed a semi-quantitative 129 method to assess local dust contribution in the western United States utilizing multivariate linear 130 131 regression of dust concentrations against categorized wind parameters. In their study, dust concentrations are assumed equal to the sum of fine soil and coarse particles using an operational 132 definition adopted from Malm et al. (1994, 2000a, 2000b). Escudero et al. (2007) proposed a 133 method to quantify the daily African dust load by subtracting the daily regional background level 134 from the PM<sub>10</sub> concentration value. Ganor et al (2009) developed and tested an automated dust 135 identification algorithm for monitoring location in Israel. Their algorithm determined a dust 136 event by three conditions: half-hour  $PM_{10}$  average level exceeds 100  $\mu g/m^3$ , this high level 137 maintained for at least three hours, and the peak  $PM_{10}$  ever reaches 180 µg/m<sup>3</sup>. In most aerosol 138 observations, however, the dust emission conditions or visual identification information are not 139 140 available. Consequently, it is challenging to identify local windblown dust events based on 141 particle concentration or chemical species because of the variability in meteorological 142 conditions, dust strength and the distance from source areas (e.g. Luo et al. 2003).

We propose here a comprehensive dust identification approach and apply this method to 143 reconstruct long-term dust climatology over the western United States. During local dust storms, 144 air samples demonstrate distinct physical and chemical characteristics, "fingerprints" that can be 145 146 used to pinpoint these events based on element abundance and size distribution. This ground-147 monitoring based method identifies individual local dust events using five dust indicators, including mass concentrations, chemical composition and size distribution. These indicators are 148 149 chosen from case studies of the aerosol data collected during three large dust storms identified 150 independently by satellite remote sensing. Some of these indicators are being used in previous dust identification works. We demonstrate here that the concurrent application of all five criteria 151 lends greater confidence to the reconstructed dust dataset in the absence of other complementary 152 measures. Hierarchical cluster analysis is subsequently conducted to apply these indicators to 153 daily aerosol data, so that a group of local dust aerosol samples that best matches these 154 155 identification criteria can be separated from other aerosols of other origins. The use of cluster

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analysis not only allows us to process large dataset, but also provides identifying threshold 156 values through clustering all aerosol data based on their statistical similarity in physical and 157 chemical characteristics. In addition, we apply this approach to scan the IMPROVE data from 158 2000 to 2007, and identify 182 local dust samples over 30 locations in the western United States. 159 A dataset of identified local dust events provide useful information for regulators to pinpoint 160 natural dust events and for researchers to verify remote sensing products and atmospheric 161 162 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 163 representation of chemically speciated dust aerosols allows atmospheric modeler to directly 164 compare model predicted crustal and trace elements with field measurements. Atmospheric 165 modelers, when equipped with such information, will be able to explicitly simulate the 166 concentrations and deposition of critical nutrients (e.g., Fe) and toxic elements to study the 167 climate, health and biogeochemical effects of dust aerosols. 168

## 169 **2. Methodology**

## 170 *2.1 Approach to identify local dust records*

171 This approach consists of several consecutive steps. First, we review the satellite data from the MODIS sensors to identify well-recorded large dust events that originate within the United 172 173 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 174 175 (IMPROVE) network in the western United States. If there are valid IMPROVE measurements, these cases will serve as the dust "samples" to explore potential rules for identifying local dust 176 177 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 178 parameters:  $PM_{10}$  and  $PM_{2.5}$  (particles smaller than 10 and 2.5  $\mu$ m in diameter, respectively) 179 mass concentrations, ratio of PM<sub>2.5</sub> to PM<sub>10</sub>, percentage of crustal elements in PM<sub>2.5</sub>, percentage 180 of industrial, residential or biomass burning elements in PM2.5, and enrichment factors of several 181 182 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 187 local dust event. For instance, long-range transported Asian and African dust has been previously reported to cause air quality degradation in both the western and the eastern United States 188 189 (Prospero, 1999; Jaffe et al., 2004; Fairlie, et al., 2007). To ensure the source of dust aerosols is 190 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<sub>2.5</sub> to PM<sub>10</sub> ratio, 191 which increases as dust plumes age. The US EPA uses a value of 0.15 - 0.26 for PM<sub>2.5</sub> to PM<sub>10</sub> 192 193 ratio for soil dust emissions from human activities (MRI, 2005). 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 194 contaminated with non-local dust sources. This ratio is chosen based on the emission splitting 195 196 factors used fugitive dust particles by the US EPA (MRI, 2005), and previous field measurements of the PM<sub>2.5</sub> to PM<sub>10</sub> ratio during dust events (e.g., 0.45 in Cheng, et al., 2005). 197 Considering that most IMPROVE are not in the immediate proximity of dust source areas, we 198 allow the cutoff ratio to be slightly larger (0.39) in the data processing. It should be noted that 199 we consider all dust emissions from North America, including these from the Chihuahuan Desert 200 in Mexico as local dust because the southern Chihuahuan Desert is a frequent dust source for 201 aerosols in the southwestern US, especially Texas and New Mexico. The low PM<sub>2.5</sub>/PM<sub>10</sub> ratio is 202 also expected to exclude high PM concentration contributed by biomass burning, which is 203 204 dominated by fine particles, resulting in a high PM<sub>2.5</sub>/PM<sub>10</sub> ratio (Reid et al., 2005). A simple sensitivity test was conducted in the Discussion section to examine how sensitive the results are 205 206 to the choice of the cutoff value.

Because dust particles can be mobilized by both wind erosion and human activities, we 207 208 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 209 210 aerosols are associated with abundant crustal elements, which differentiate them from aerosols 211 from biomass burning 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 212 the largest sector of primary PM emissions. The five major fugitive dust sources in the United 213 214 States are vehicle emissions from unpaved road (47%), paved road (7%), agricultural operation 215 (29%), and construction (11%), and mining/quarrying (7%). Each of these sources involves either fossil fuel combustion or other human activities in the immediate vicinity of dust sources. 216 217 Therefore, compared to natural dust, anthropogenic dust aerosols contain higher anthropogenic218 originated elements, such as elemental carbon (from fossil fuel or biomass combustion) or heavy 219 metals (such as Zn, Pd and Cu) from industrial operations (Chow et al., 1993; 2003; Reff et al., 220 2009). For instance, high levels of black carbon, Pb, Zn are found in paved road particles while high levels of nitrate (NO<sub>3</sub>), Cr and Ni are found in unpaved road dust (Chow et al., 1993). 221 222 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 223 224 enrichment factors (EFs) of anthropogenic pollutants as the indicators to distinguish natural dust from anthropogenic dust. In this study, the enrichment factors (EFs) are calculated for a series of 225 elements using Si as the reference element and the abundance of crustal elements at the Earth's 226 227 surface as given by Taylor and McLennan (1985),

$$EF_{X} = \frac{(X / Si)_{aerosol}}{(X / Si)_{crustal}}$$
(1)

where (X/Si)<sub>aerosol</sub> and (X/Si)<sub>crustal</sub> represent the ratio of a certain species (X) to Si in sampled dust 229 230 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 231 232 anthropogenic origin. By examining the variation of the above parameters, we can establish useful criteria for the subsequent statistical analysis to identify other local dust events that are not 233 234 revealed by satellite data. Considering all the relevant parameters discussed above, five criteria will be the focus of subsequent statistical analysis: 1) PM<sub>10</sub> and PM<sub>2.5</sub> concentrations; 2) Ratio of 235 236 PM<sub>2.5</sub> to PM<sub>10</sub>; 3) Concentrations of crustal elements Si, Ca, K, Fe, Ti; 4) Concentrations of 237 anthropogenic pollutants, As, Zn, Cu, Pb, sulfate ,nitrate, Organic carbon (OC), and EC; and 5) 238 enrichment factors (EF) of anthropogenic pollution elements Cu, Zn, Pb and K.

The third step of the procedure is to cluster all daily aerosol data according to these 239 240 indicators, and to pinpoint a local dust group. This is achieved by applying hierarchical cluster 241 analysis to IMPROVE daily data site by site for all selected sites. Cluster analysis is a statistical method that creates clusters of items or objects that have similarity within the same cluster but 242 with differences between clusters. This technique has been previously applied to air quality 243 studies to investigate source origins of air pollutants (e.g. Slanina et al., 1983; Dorling et al., 244 1992; Tong et al., 2005; Van Curen, et al., 2002). As discussed earlier, dust episodes are usually 245 extraordinary events with large perturbations in both aerosol concentrations and chemical 246 composition compared to that during non-dusty periods. Assuming spatial homogeneity in dust 247

chemical composition within a dust source region, we used the hierarchical cluster analysis to 248 group all IMPROVE aerosol measurements based on the similarities in chemical and physical 249 250 characteristics. The spatial homogeneity in dust composition, as observed in many desert regions, is likely due to the fact that the aerosol over the desert itself is well mixed as a result 251 from long-time continuous deposition and uptake of materials from the ground (Schutz and 252 Sebert, 1987). The cluster analysis is conducted using the statistical software SPSS Statistics 253 254 17.0 (SPSS Inc.). As hierarchical cluster analysis is applied to each site, more than 600 daily data covering 2000-2007 period are involved for ~90% of 68 sites except for a few sites with missing 255 data. In the cluster analysis, the concentrations of Si, Ca, K, Fe, Ti, As, Cu, Pb, S, Zn and V, 256 PM<sub>2.5</sub>/PM<sub>10</sub> ratio, and the enrichment factors of Ca, K, Fe, As, Cu, Pb and Zn, are used to 257 construct six clusters. The concentrations of five aerosol components, Al, sulfate, nitrate, OC and 258 259 EC are excluded from this analysis to avoid the unbalanced sampling issue, because a large portion of these data are either missing or invalid. Between-Group Linkage clustering method 260 261 and Pearson Correlation to measure inter-cluster intervals are configured to assemble the most similar cases into a same group. This method measures the correlations of x and y variables of 262 263 case i according to the following formula (SPSS Statistics 17.0 Algorithms, SPSS Inc.):

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$$C_{xy} = \frac{\sum_{i} (Z_{xi}Z_{yi})}{N}$$
(2)

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

Where  $C_{xy}$  the correlation between variable x and variable y,  $Z_{xi}$  and  $Z_{yi}$  are the standardized Zscore 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<sub>i</sub>) for the N cases. The cases with higher correlation, which means higher similarity, are put into one cluster (or group).

In this study, the cluster analysis processes daily aerosol data (i.e., 24-hour every third day according to the IMPROVE sampling protocol) at each site to identify a dust group. During the study period, there are more than 600 daily data records from 90% of studied 68 sites except for a few sites with less than 300 data records. After the cluster analysis, the identified dust records at each site are further grouped according to the geographic locations and temporal ranges for subsequent analysis.

#### 276 2.2 Observational data

277 The aerosol observation data from the IMPROVE network were chosen for two reasons. 278 The IMPROVE monitoring sites, with a few exceptions, are deployed in the national parks and wilderness areas in the United State (Pitchford and Malm, 1994), including many sites in close 279 proximity or downwind to major dust source regions. Second, the IMPROVE network is also one 280 of the two national air quality monitoring networks that measure both mass concentrations and 281 282 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 283 national coverage, but there is no aerosol composition data available from this network. Another 284 national aerosol monitoring network, the Chemical Speciation Network (CSN), is deployed 285 mostly in urban areas, making it unsuitable for dust monitoring due to anthropogenic 286 contamination. There are also some regional networks, such as the Southeastern Aerosol 287 Research and Characterization Study (SEARCH), which measures aerosol mass and composition 288 at both urban and rural sites (Edgerton et al., 2009). The currently operating eight SEARCH sites, 289 290 however, are located in the southeastern US and are too far away from major dust sources.

291 A subset of 68 sites from the IMPROVE network are used in this study. This subset of IMPROVE sites, deployed in eight western states (Fig. 1), is chosen based on the findings in 292 293 previous studies that have identified the geographical distribution of active dust sources in the North America (Gillette and Passi, 1988; Malm et al., 2004; Van Curen, et al., 2002; Wells et al., 294 295 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  $PM_{10}$ 296 297 and PM<sub>2.5</sub> mass concentrations, and PM<sub>2.5</sub> 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, 298 299 Pb, Rb, S, Se, Si, Sr, Ti, V, Zn, Zr) measured by proton-induced X-ray emission (PIXE) and Xray fluorescence (XRF), selected ions (Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>-</sup>) by ion chromatography (IC), organic to 300 301 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 302 303 calculated from the mass concentrations of five major soil-derived elements (Al, Si, Ca, Fe, K, 304 and Ti) in their assumed oxides (Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, CaO, K<sub>2</sub>O, FeO, Fe<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, respectively) (Malm, 2000a, 2000b): 305

$$Soil_f = 2.2[Al] + 2.49 [Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti]$$
 (3)

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 2000-2007 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 east of Kansas are excluded from this analysis, since there are no major active dust sources in this region.

Besides the IMPROVE data, satellite remote sensing of dust aerosols is used to independently 313 identify local dust events. The MODIS sensors aboard both Terra and Aqua have been making 314 global daily observations of atmospheric aerosols since 2002 (Terra started in 2000). A total of 315 316 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. 317 Over the ocean, MODIS relies on the aerosol spectral signature from 0.55 to 2.13 µm to separate 318 319 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 320 visible wavelength to observe surface reflectance (Kaufman et al., 1997). 321

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### 323 **3. Identifying windblown dust events**

### 324 *3.1 Analysis of satellite detected dust events*

During the study period, there were thirteen large dust storms occurring in the southwestern 325 United States that have been identified from NASA Earth Observatory's Natural Hazards dust 326 products (http://earthobservatory.nasa.gov/NaturalHazards). The purpose of analyzing these 327 328 known dust events is to learn from these data of the distinct physical and chemical properties of 329 local dust samples. The IMPROVE sampling protocol is to collect a 24h duration sample every three days. Therefore, it is difficult for the ground monitors to capture all dust events identified 330 331 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 332 all dust events. Therefore, it is not easy to pinpoint a dust case that is simultaneously recorded by 333 both satellite sensors and ground monitors. Here we focus on three such rare dust cases that were 334 recorded by both ground and satellite observations on April 15, 2003, November 27, 2005 and 335 April 12, 2007. The MODIS imageries show that the three storms originated from different 336 337 source regions. The former two storms were conceived from the Chihuahuan 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 April 15, 2003 storm, the Big Bend National Park, TX (BIBE1) site and the GUMO1 site for the November 27, 2005 storm, and the Agua Tibia, CA (AGTI1) site and the San Gorgonio Wilderness, CA (SAGO1) site for the April 12, 2007 storm (Fig. 2).

The aerosol mass and chemical composition measurements at these monitors are then used to 345 extract the commonality of typical local dust samples. In each case, we compare the observed 346 concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, crustal (Si, Ca, Fe, K) and anthropogenic elements (Cu, Zn, Pb), 347 sulfate, nitrate, OC and EC in  $PM_{2.5}$ , the  $PM_{2.5}/PM_{10}$  ratio, the percentage of the above species 348 349 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 350 dusty periods: 1) compared to that on non-dusty days, the  $PM_{10}$  concentration during a dusty day 351 was elevated by 2-10 times from the pre-storm and post-storm levels; 2) although the 352 353 concentration of PM<sub>2.5</sub> also increased during a dust storm, the PM<sub>2.5</sub>/PM<sub>10</sub> ratio dropped significantly to approximately 0.2, a value typically representing freshly emitted soil particles; 3) 354 355 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<sub>2.5</sub>, 356 including Cu, Zn, Pb, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and OC, all decreased from their corresponding pre-storm and 357 post-storm levels, although the absolute concentrations may have increased or decreased 358 359 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 varied at different sites, 360 361 with an increase at the GUMO1 site and the BIBE1 site during the November 2005 dust storms and a decrease at the AGTI1site and the SAGO1 site during the April 2007 dust storm. The 362 concentrations of particulate sulfate and nitrate during dust events are controlled by two 363 processes: the dilution by dusty but otherwise clean air of background pollution (Guo et al., 2004) 364 365 and the supply of sulfate and nitrate from soil particles and the uptake and/or formation of nitrate 366 and sulfate on dust particles. Similar phenomena have been reported in previous studies (Arimoto et al., 2006; Wang et al., 2005, Sun et al., 2004) in which the concentrations of nitrate 367 368 and sulfate increased during dust days, since mineral dust particles provide alkalic surface and

catalytic for the scavenging and heterogeneous conversion of  $SO_2$  and  $NO_x$  into sulfate and nitrate. For the selected cases, the absolute concentrations of sulfate and nitrate increase, but the relative abundance of these components decreases (Fig 2); 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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#### 382 *3.2 Cluster analysis*

After establishing these identifying indicators, we use cluster analysis to test the hypothesis 383 384 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 385 data over the 68 study sites, using the concentrations of PM<sub>10</sub>, PM<sub>2.5</sub>, elements (Si, Ca, K, Fe, Ti, 386 As, Cu, Zn, Pb, V), the PM<sub>2.5</sub> to PM<sub>10</sub> ratio, the enrichment factors of K, Ca, Cu, Zn and Pb as 387 388 the clustering criteria as discussed earlier. We found that over 30 of the 68 sites, there is one aerosol data group that demonstrates similar physical and chemical characteristics as observed in 389 390 the previously satellite identified dust events. At the remaining sites, none of the IMPROVE data 391 show any consistent pattern of dust events.

392 Figures 3 and 4 show the results of cluster analysis of all IMPROVE observation data from 2000 to 2007 at the GUMO1 site, which experienced the large number of dust storms during the 393 study period. The cluster analysis divides all data into six groups, and the first group has the 394 highest  $PM_{10}$  concentrations (Fig. 3a). The mean  $PM_{10}$  concentration in this group is 395 approximately 60  $\mu$ g/m<sup>3</sup>, 3–10 fold of the typical background levels in the western United States 396 (Malm et al., 1994). The PM<sub>2.5</sub> concentrations in this group are also higher than in other groups, 397 although the differences among groups are relatively smaller than that for the PM<sub>10</sub> 398 399 concentrations (Fig. 3b). The concentrations of the four crustal elements are significantly higher

400 in group 1 than in other groups (Fig. 3c). During non-dusty days, the concentrations of these crustal elements in PM<sub>2.5</sub> are low (less than 0.1  $\mu$ g/m<sup>3</sup>) in most cases, except for Si the 401 concentration of which reaches 1.0  $\mu$ g/m<sup>3</sup> occasionally. During dust storms, the Si concentration 402 varies from 1.0  $\mu$ g/m<sup>3</sup> to 6.0  $\mu$ g/m<sup>3</sup>. For the three trace metals that are mainly attributed to 403 anthropogenic sources, their concentrations in group one is among the lowest, but the difference 404 405 is not as distinguishable as that of  $PM_{10}$  or crustal elements (Fig. 3e), likely resulting from varying meteorological conditions and uneven distribution of emission sources. For the four 406 major aerosol components (sulfate, nitrate, OC and EC), the distinction among these groups is 407 408 further blurred. This 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  $PM_{2.5}/PM_{10}$ 409 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 410 in other groups have a wide range, from 0.1 to over 0.9 (Fig. 3f). The higher ratio reflects either 411 higher contribution of anthropogenic sources and biomass burning, or the aging of aerosol 412 plumes. 413

414 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, 415 their relative abundance (in percentage) is also higher than in other groups (Fig 4a). The opposite 416 417 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, 418 419 indicating their soil origin. In comparison, the silicon referenced enrichment factors are much higher in all other groups, except group 4, which shows consistent low enrichment factors and 420 421 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 422 soil dust (such as from unpaved road or mining operation). Based on the consistent and distinct 423 chemical and physical patterns that simultaneously match the five stipulated criteria, we hence 424 425 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 section 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
- 432 burning) or with different chemical composition (i.e., aerosols originated from other sources).
  - 433

### 434 **4. Summary of identified dust records**

## 435 *4.1 Summary of identified dust records*

Cluster analysis of all aerosol data identifies a total of 182 dust records from 30 of the 68 436 sites (Table 1). These sites with dust records are also marked in red in Figure 1. These 30 sites 437 are located in the states of Texas, New Mexico, Arizona, South California, Nevada and Colorado. 438 Such spatial distribution is consistent with the distribution of the four U.S. Deserts, namely the 439 440 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 441 442 wind power in spring lifts surface soil grains (Gillette and Hansen, 1989). Overall, the spatial distribution is similar to the dust source map reported in previous studies (Malm et al., 2004; 443 444 Kavouras et al, 2007).

Among the 30 dust sites, there are three sites, including Phoenix (PHOE1) and Douglas 445 446 (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 447 448 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 449 450 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 crustal and 451 452 anthropogenic elements result from strong mixing of wind-generated emissions and urban 453 plumes, a unique setting for studying the interactions between dust and urban pollutants.

454

## 455 *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). The "well-known" seasonal trend of local windblown dust in US, however, has not been independently evaluated against robust measurement data except for Kavouras et al. 461 (2007), who used wind and visibility data to identify local windblown dust and investigated the462 seasonal trend.

463 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 464 emissions. Figure 6 shows the interannual variations of local dust records in the five dust regions 465 from 2000 to 2007. Although the IMPROVE monitors are not expected to capture all dust events, 466 467 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 468 away from each other. Therefore, the observed dust events are unlikely to overlap those detected 469 470 at other locations. Only during extraordinarily large dust events, such as the April 15, 2003 storm, the dust plume can be detected by multiple IMPROVE monitors (four in this case) and the data 471 472 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 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 Chihuahuan Desert (59 cases) and the Sonoran Desert (62 cases) are by far the most active source regions. In general, the Chihuahuan 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.

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 Chihuahuan 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 492 activities in the Sonoran Desert can largely offset the diminished activities in the Chihuahuan 493 Desert. Actually, May 2003 is the month with the largest number of local dust records during the 494 eight-year period, with 16 dust records obtained by 10 IMPROVE monitors there. The 495 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 496 two months earlier than in Sonoran Desert. The lowest number of dust events is found in January 497 and August, during both periods dust activities were found only in the Mojave Desert. During the 498 study period, there are eight sites that have observed more than eight local windblown dust 499 events, with the GUMO1 site in Texas having the largest number (27) of dust records. In 500 501 addition, the Queen Valley site in Arizona (QUVA1), the Big Bend site in Texas (BIBE1), the Salt Creek site in New Mexico (SACR1), the Chiricahua site (CHIR1), the Saguaro West site 502 (SAWE1), and the Ike's Backbone site (IKBA1), all in Arizona, have captured 19, 16, 12, 9, 9, 503 and 9 dust events, respectively. These monitors are either located in or downwind to the 504 previously identified dust source regions associated with the geological characteristics of high 505 soil erodibility (Kavouras et al., 2007). 506

507

### 508 **5. Discussion**

### 509 *5.1 Limitations with the dust identification approach*

The major limitation of our approach is that the dust indictor parameters may not be universally available from routine aerosol monitoring networks. 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. The lack of these aerosol parameters limits the applicability of our approach to dust studies for those regions.

Alternatively, we consider here three simplified methods that use only basic aerosol mass concentrations, and compare their capability to pinpoint dust events to that of the full method using all five indicators. The first simplified approach uses two dust indicators, the  $PM_{10}$  mass concentration (> 40 µg/m<sup>3</sup>) and the  $PM_{2.5}/PM_{10}$  ratio (< 0.35) as the filtering criteria. The  $PM_{10}$ cutoff and the  $PM_{2.5}/PM_{10}$  ratio cutoff are taken from the lower and upper 95% values of the corresponding parameters in the local dust group identified by the full method as proposed in this

study, respectively. The second method is similar to the first one, except using a PM<sub>2.5</sub>/PM<sub>10</sub> 523 cutoff of 0.20, the median value of the dust group. This ratio is also used by the US EPA to split 524 fugitive dust PM<sub>10</sub> into PM<sub>2.5</sub> (MRI, 2005). Compared to the first one, the second method is 525 considerably more exclusive. The third method simply uses  $PM_{10} > 100 \mu g/m^3$  as the identifying 526 indictor, following Ganor et al. (2009). Due to the IMPROVE sampling protocols, 24-hour mean 527 528  $PM_{10}$  concentrations are used here, instead of hourly  $PM_{10}$  data as in Ganor et al. (2009). Table 2 compares the performance of the three simplified methods to identify dust events to that of the 529 530 full method. Here we define the performance using two categorical evaluation metrics as introduced by Kang et al. (2009): Hit Rate and False Alarm Ratio. Hit Rate is the percentage of 531 "true" dust events identified by the simple method to all events by the full method, while the 532 False Alarm Ratio is the percentage of "false" events (i.e., not considered local dust events by 533 534 the full method) to all events selected by the simple methods. The first simplified method has the 535 highest hit rates, catching 27% of the dust events identified by the full method. Meanwhile, it is also associated with the highest false alarm ratio, with 68% of the dust events it selected deemed 536 false by the full method. When the  $PM_{2.5}/PM_{10}$  ratio is further constrained to 0.20, the false 537 alarm ratio has been reduced significantly (to 16%), but at the cost of hit rate, which shows that 538 539 the second method can catch only 13% of the all dust events. The revised Ganor method demonstrates dust identifying capability between the two simplified methods. Although these 540 simplified methods show varying effectiveness to identify local dust events, it should be pointed 541 out that chemical fingerprint is still needed to assure the origin of measured aerosols. For 542 example, the measurement data over the three urban sites can satisfy all selection criteria for 543 local dust events, except the high levels of anthropogenic components. Such information reveals 544 545 either human contamination of the dust aerosols, or human motivated dust sources (such as road dust from unpaved road). Regardless of its complexity, our proposed approach is likely to work 546 most efficiently when all five identification criteria are concurrently applied. 547

548 5.2 Dust activities in the western United States

549 Our study reveals that dust events in the United States occur in almost all seasons, suggesting 550 the prevailing impact of windblown dust across the year. This seasonal variation is consistent 551 with previous model simulation over the United States (Gillette and Hansen, 1989; Park et al., 552 2010). The windblown dust emissions peak in the spring, due to high wind speed, low soil 553 moisture, and a lack of vegetation cover over erodible land surface. The springtime maximum 554 over North America was also reported in a long-term general circulation model study by Tegen 555 and Miller (1998). Some previous study (e.g. Gatz and Prospero, 1996) has used the infrequency 556 of summertime dust plumes to exclude the possibility of the impact of dust storms on regional air 557 quality. This work and several previous modeling studies suggest that it is possible to see 558 summertime impact of dust aerosols originated from the western United States.

559 Although the IMPROVE sites are not expected to capture all dust events on a local scale, in 560 particular for the Great Basin Desert and southern Chihuahuan Desert where monitors are sparse, a dataset of observed dust events developed from using our approach can help evaluate the 561 completeness or efficiency of satellite-based approaches. Meanwhile, a variety of computer 562 models have been developed to study the life cycle of dust aerosols and their effects on the 563 regional and global climate systems. Although dust sources over North America contributes to 564 only 3% of global dust budget (Ginoux et al., 2001), previous model simulations have 565 highlighted the importance of dust aerosols in regional air quality and climate modeling over the 566 567 western United States (Draxler et al., 2010). The same observed dust dataset can provide detailed comparisons between model and observations on an event level. Furthermore, the rich pool of 568 569 aerosol chemical composition data associated with these identified dust records are useful to compile chemical profiles for splitting dust aerosols. Recent advances of aerosol modeling (such 570 571 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 572 573 emitted dust particles. A companying paper (Dan et al., Chemical composition of natural dust particles in the United States, in preparation) utilizes the data of the identified dust events and 574 575 chemical composition to derive chemical profiles that could be used in future dust aerosol 576 emission and modeling works.

577 Dust activities display a four-year cycle during the eight-year study period. While more data are needed to verify this four-year cycle observed in this study, we discuss briefly here the 578 579 possible driving forces behind this interannual variability. Climate models have predicted that a transition to a more arid climate is under way in the southwestern United States, where multiyear 580 581 drought and the 1930s Dust Bowl will become the new climatology within a time frame of years 582 to decades (Seager et al., 2007). Windblown dust emissions are controlled by a number of important parameters, such as wind speed, soil moisture, surface roughness and erodible dust 583 584 supply (Marticorena et al., 1995; Gillette et al., 1988; 2004). Among these controlling factors,

585 surface wetness can be used as an indicator to drought condition, which is often associated with dust activities in arid environment. Here we examine the monthly surface wetness over the five 586 587 dust regions using the Modern Era Retrospective-analysis for Research and Application (MERRA) dataset from the NASA's Goddard Space Flight 588 Center (http://gmao.gsfc.nasa.gov/research/merra/). Figure 8 shows that the lowest surface wetness is 589 found in different months among these regions. The Chihuahuan Desert generally sees an early 590 591 dry season, while the Sonoran Desert and the Mojave Desert are often associated with a prolonged and drier summer. Although soil moisture controls several factors that influence dust 592 emissions, the monthly surface wetness data here are not in good correlation with the observed 593 594 dust pattern. The monthly regional mean of surface wetness may not represent the local condition under which dust emissions are initiated. As discussed by many field and model 595 596 studies, windblown dust emission is a complicated process that has not been fully understood. In addition, these processes are increasingly complicated by human disturbance of the land surface, 597 such as the rapid urbanization in southern Arizona (Sorooshian et al., 2011). Future analysis of 598 599 the meteorological parameters and surface conditions over these regions is need to further 600 investigate the underlying mechanisms causing the interannual variations. Given the climate model prediction of a drier climate in the Southwest, it is interesting to continue observing how 601 602 the dust activities will respond to the changes in regional and global climate systems.

603

#### 604 **6.** 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 aerosols, we demonstrate how to use an observation-based approach to pinpoint 182 local dust records over 30 locations in the western United States over a eight-year study period.

The results presented in this study are subject to several limitations. The IMPROVE monitors are unevenly deployed over different dust source regions. Therefore, the observationbased dust data may not represent the overall emissions from each region. For instance, there are only five monitors in the Great Basin Desert, 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 Chihuahuan Desert. The high number of dust records over these regions reflects both dense detection and active dust emissions.

Although our method specifically targets local dust samples, it can be easily extended to 620 621 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 622 623 elements. Compared to local dust events, the increase in mass concentration from the non-dusty level may be smaller, and the  $PM_{2.5}/PM_{10}$  ratio is much higher during a long-range transport 624 event (Cheng et al., 2005). The volcanic ash or dry fog formed from emitted sulfate dioxide is 625 associated with high levels of sulfate content in the aerosols (Bao et al., 2010). The high sulfate 626 627 and low anthropogenic elements can be used to distinguish these data from that featuring coal-628 burning aerosol. Similarly, biomass burning originated aerosols contain high levels of potassium, organic carbon and black carbon, and the aerosols are predominantly in the fine mode (Reid et 629 al., 2005). Through a reasonable procedure of remote sensing-assisted data training, our method 630 631 can be applied to identify a number of distinct aerosol sources in research and regulatory applications. Our approach emphasizes using ground monitoring data for dust identification. In 632 633 this approach, the use of satellite data is limited to selecting independently identified dust events 634 for the purpose of methodological training. There are other approaches that rely predominantly 635 on remote sensing data to pin-point dust events (Prospero et al, 2002; Lee et al, 2009; Rivera-Rivera et al., 2010). Our study proposes an alternative way to pin-point dust events that can work 636 complimentarily with these satellite-based methods to identify dust events. 637

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Site	SiteID	Longitude	Latitude	<b>PM10</b> (mg/m3)	<b>PM2.5</b> (mg/m3)	PM2.5/PM10 Ratio	Local Dust Events (YYMMDD)
1	BOAP1	-106.85	33.87	42.44	6.99	0.16	11016
2	GICL1	-108.24	33.22	35.59	7.71	0.22	70328
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
б	GUMO1	-104.81	31.83	73.2	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	61.2	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	59.6	17.22	0.3	011016, 030515, 030530, 070412, 060716
13	TONT1	-111.11	33.65	61.6	13.89	0.23	060716, 070412, 070708, 070720, 071006
14	PEFO1	-109.769	35.07	55.4	13	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	0.19	020508, 020511, 020520, 040903, 061228
17	DOME1	-118.14	35.73	65.6	6.86	0.1	31030
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	21002
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	20228
23	GRBA1	-114.22	39.01	104.62	18.85	0.18	20228
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	70720
26	GRSA1	-105.52	37.72	51.28	11.1	0.23	000517, 020511, 030503, 050603
27	MEVE1	-108.49	37.2	65.2	13.68	0.22	030202, 030415, 050419
28	DOUG1	-109.54	31.35	81.27	21.2	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

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.

Table 2. Comparisons of three simplified dust identification methods to the full method proposed
in this study. Here the dust records identified by the full method are used as reference data to
calculate hits and false alarms.

Method	Hits <sup>d</sup>	False Alarms <sup>e</sup>	Hit Rate $^{\rm f}$	False Alarm Ratio <sup>g</sup>
Simplified Method I <sup>a</sup>	49	381	27%	68%
Simplified Approach II <sup>b</sup>	24	86	13%	16%
Ganor Approach <sup>c</sup>	38	42	21%	29%

a. Method I uses two threshold values ( $PM_{10} > 40 \ \mu g/m^3$ , and  $PM_{2.5}/PM_{10}$  ratio < 0.35) to identify dust events;

b. Method II is similar to that of Method I, except that the  $PM_{2.5}/PM_{10}$  threshold value is set to be 0.20;

c. The Ganor method (revised from Ganor et al., 2009) uses 24-hour  $PM_{10}$  concentration > 100  $\mu$ g/m<sup>3</sup> as the sole criteria.

d. Hits are the number of dust records identified by both the simple method and the full method;

e. False alarms are the dust records selected by the simple method, but not by the full method.

f. Hit Rate is the percentage of "true" dust events identified by the simple method to all events by the full method,

g. False Alarm Ratio is the percentage of "false" events (i.e., not considered local dust events by the full method) to all events selected by the simple methods.



Figure 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. The background is the area classified as arid or semi-arid land.



Figure 2. Variations of  $PM_{10}$ ,  $PM_{2.5}$  and chemical components of  $PM_{2.5}$  at the BIBE1, GUMO1, AGTI1 and SAGO1 sites during, before and after three dust storms. These dust events have been pinpointed by MODIS satellite data. "Fraction in PM2.5" indicates the fraction of the concerned aerosol component to total  $PM_{2.5}$  mass, and  $EF_{Si}$  indicates the enrichment factors of concerned species using Si as the referent element.



Figure 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)  $PM_{10}$  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)  $PM_{2.5}/PM_{10}$  ratio. The bottom and top edges of the box indicates the 25th and 75th percentile and the line in the box indicates the 50%. Group 1 was identified as the local dust group.



Figure 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 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. The bottom and top edges of the box indicates the 25th and 75th percentile and the line in the box indicates the 50%. Group 1 was identified as the local dust group.



Figure 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 April 15, 2003 storm and the November 27, 2005 storm.



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



Figure 7. The monthly frequency of local dust cases from 2000 to 2007 in the five dust source regions, namely, the Chihuahuan Desert (CHD), the Sonoran Desert (SOD), the Mojave Desert (MOD), the Great Basin Desert (GBD) and the Colorado Plateau (COP).



Figure 8. Monthly variations of surface wetness over five dust source regions during the study period. The surface wetness is derived from the NASA Modern Era Retrospective-analysis for Research and Application (MERRA) dataset. Five dust regions include the Chihuahuan Desert (CHD), the Sonoran Desert (SOD), the Mojave Desert (MOD), the Great Basin Desert (GBD) and the Colorado Plateau (COP).