

The role of dust storms in total atmospheric particle concentrations at two sites in the western U.S.

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[1] Mineral aerosols are produced during the erosion of soils by wind and are a common source of particles (dust) in arid and semiarid regions. The size of these particles varies widely from less than 2 μm to larger particles that can exceed 50 μm in diameter. In this study, we present two continuous records of total suspended particle (TSP) concentrations at sites in Mesa Verde and Canyonlands National Parks in Colorado and Utah, USA, respectively, and compare those values to measurements of fine and coarse particle concentrations made from nearby samplers. Average annual concentrations of TSP at Mesa Verde were 90 $\mu\text{g m}^{-3}$ in 2011 and at Canyonlands were 171 $\mu\text{g m}^{-3}$ in 2009, 113 $\mu\text{g m}^{-3}$ in 2010, and 134 $\mu\text{g m}^{-3}$ in 2011. In comparison, annual concentrations of fine (diameter of 2.5 μm and below) and coarse (2.5–10 μm diameter) particles at these sites were below 10 $\mu\text{g m}^{-3}$ in all years. The high concentrations of TSP appear to be the result of regional dust storms with elevated concentrations of particles greater than 10 μm in diameter. These conditions regularly occur from spring through fall with 2 week mean TSP periodically in excess of 200 $\mu\text{g m}^{-3}$. Measurement of particles on filters indicates that the median particle size varies between approximately 10 μm in winter and 40 μm during the spring. These persistently elevated concentrations of large particles indicate that regional dust emission as dust storms and events are important determinants of air quality in this region.

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1. Introduction

[2] Atmospheric aerosols have a wide range of effects on the global environment including influences on climate, visibility, biogeochemistry, and human safety and health [Aurup, 2010; Mahowald *et al.*, 2010]. There are a variety of sources of aerosols including industrial emissions, transportation emissions, and the wind erosion of soils. Most aerosol research and measuring efforts focus on coarse particle matter (PM) with aerodynamic diameters between 2.5 and 10 μm (PM_{2.5–10}) and the fine particle-size classes less than 2.5 μm (PM_{2.5}). These relatively small particles receive much attention because they can travel for thousands of kilometers through the atmosphere, and they play a major role in radiative forcing of the atmosphere, thereby affecting climate [Andreae and Crutzen, 1997]. Very small particles,

often associated with industrial activity, can also penetrate deeply into the lungs leading to a variety of impacts on pulmonary function and other aspects of human health [Aurup, 2010]. Despite these many compelling reasons to focus research efforts on small aerosols, particles larger than 10 μm are often also present in the atmosphere, particularly near dryland areas that are sources of mineral aerosols [Lawrence *et al.*, 2010].

[3] The potential influence of particles larger than 10 μm in diameter on overall atmosphere dust loads near dryland source regions is large but highly variable. Although measurements of total suspended particles (TSP) and of particles larger than 10 μm in diameter were common in the US prior to the 1980s [Lundgren *et al.*, 1984; Noll *et al.*, 1985; Stout and Lee, 2003], such measurements are now infrequent in the U.S. because the U.S. Clean Air Act regulations focus solely on the PM_{2.5} and PM_{2.5–10} fractions of atmospheric particles. As a result, there is infrequent monitoring of larger particles. Similar air-quality regulation is in place in many industrialized countries around the world based on the assumptions that particles larger than 10 μm —referred to here as PM_{>10} particles—do not travel long distances through the atmosphere and have little effect on human health, at least relative to the smaller particles [c.f. Aurup, 2010]. As discussed below, both of these assumptions may not be as straightforward as they seem, thereby indicating the need to better understand the relative importance of these larger particles in arid regions of the United States.

Additional supporting information may be found in the online version of this article.

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Figure 1. Map of sampling locations and related study areas.

[4] Very coarse particles ($PM_{>10}$) are readily emitted from desert soil surfaces [Bacon *et al.*, 2011; Li *et al.*, 2009] but have high depositional velocities and so typically are assumed to travel short distances through the atmosphere [Sehmel, 1980]. However, information from a range of studies within and adjacent to drylands suggests that $PM_{>10}$ can travel tens to thousands of kilometers during particularly windy conditions [reviewed in Lawrence and Neff, 2009]. In a study of dust deposited during large dust storms to snow cover in the San Juan Mountains of Colorado, Lawrence *et al.* [2010] observed dust particles ranging in size from less than 1 to 100 μm with the bulk of particles in the 20–60 μm size range after a minimum transport distance in excess of 200 km. A number of other studies including in areas downwind of large deserts indicate that particles with diameters in excess of 10 μm can travel for hundreds to thousands of kilometers prior to deposition [Alastuey *et al.*, 2005; Betzer *et al.*, 1988; Chan *et al.*, 2005; McTainsh *et al.*, 1997; Middleton *et al.*, 2001; Rutherford *et al.*, 1999]. If these particles are being transported over moderately long distances (several hundred km or more) in the atmosphere, then these large mineral aerosols from remote sources could reach large population centers around the world’s dust-producing regions.

[5] Among the numerous effects of dust, the health impact of exposure to high concentrations of very coarse particles is poorly understood. Epidemiological studies, however, indicate that dust storms in Asia lead to elevated mortality and cardiovascular disease [Kwon *et al.*, 2002]. Such effects might not be attributable solely to the smallest particle sizes carried by these storms in light of statistical evidence that respiratory morbidity of children in China is related to the TSP concentration (distinct from that of the

fine and coarse fractions) with respect to respiratory function, including bronchitis, persistent cough, and persistent phlegm [Zhang *et al.*, 2002]. The TSP fraction can be responsible for a large fraction of the total atmospheric metal load in urban areas [Schleicher *et al.*, 2011]. In Australia, the incursion of dust storms into urban areas has been followed by an increase in asthma symptoms and asthma-associated hospitalization [Rutherford *et al.*, 1999]. These studies combined with evidence for moderate distance transport of large particles clearly indicates the need to comprehend better the influence of desert dust on atmospheric particle concentrations including the temporal and spatial dynamics of particles larger than the current regulatory $PM_{2.5}$ and PM_{10} standards.

[6] Dust storms are a frequent occurrence in many arid regions and recent work using calcium in rainfall as a proxy for dust deposition suggests large increases in dust deposition across large areas of the western US [Brahney *et al.*, 2013]. Despite indications of changing dust deposition in the western US, there is limited information on the seasonality, spatial distribution, and overall concentration of particles during dust storms in the arid western US [but see Kavouras *et al.*, 2007]. Concentrations of $PM_{2.5-10}$ and $PM_{<2.5}$ in the arid western US are highly variable [Li *et al.*, 2013] but are generally very low on the Colorado Plateau despite frequent dust storms in this region [e.g., Hahnenberger and Nicoll, 2012]. In this study, we present the results of continuously biweekly monitoring of TSP concentrations for one year at Mesa Verde National Park (MVNP) and three years at Canyonlands National Park (CNP). The purpose of these measurements was to characterize seasonal variation of total particle concentrations at these sites and to compare these observations to data for known dust events and from samples of $PM_{2.5}$ and $PM_{2.5-10}$ concentration.

2. Methods

2.1. Sites and Samplers

[7] Two TSP samplers were installed on the Colorado Plateau in close proximity to Interagency Monitoring of Protected Visual Environments (IMPROVE) network samplers (Figure 1). The IMPROVE network was established in 1985 to provide long-term monitoring of visibility in protected air sheds (particularly in National Parks and Wilderness areas) including routine measurement of $PM_{2.5}$ and $PM_{2.5-10}$ particle concentrations. The IMPROVE network does not routinely measure TSP concentrations. One TSP sampler was installed in January 2011 at approximately 1.5 m above ground level on a rocky mesa top approximately 4.5 m away from an IMPROVE sampler in Mesa Verde National Park (Lat 37.1984, Lon -108.4907 , 2170 m elevation). The other sampler was installed 3 m above ground level on a mesa top and approximately 10 m away from an IMPROVE sampler in the Sky region of the Canyonlands National Park (Lat 38.4589, Lon -109.821 , 1798 m elevation). The samplers were installed to take advantage of local facilities and although the Mesa Verde sampler is at a lower height than the Canyonlands sampler, it is located on bare rock about 50 m from the edge of the mesa. The Canyonlands sampler is similarly located within 50 m of the edge of a large rocky mesa top. Both samplers have

very few local (<100 m) sources of dust and are primarily exposed to airflow that originates from the direction of nearby cliffs. For these reasons, we expect very limited contributions from sources of dust in the immediate vicinity of the samplers.

[8] The Canyonlands sampler, installed in June 2008, was a Thermo GS2310 High Volume Air Sampler outfitted with dual-stage motor and a Thermo G313 Mass-Flow Controller and mechanical timer (Thermo Electron Corporation, Franklin, MA, USA). The Mesa Verde sampler, installed in late 2010, was a Staplex TSP-CF device outfitted with a dual-stage motor and digital mass-flow controller and airflow recorder and timer (Staplex, Brooklyn, NY, USA). Both samplers were calibrated to continuously draw 1132 L min^{-1} of air through Whatman EPM 2000 $8 \times 10 \text{ cm}$ filters (Whatman, Clifton, NJ, USA).

[9] The TSP samplers used in this study meet the US EPA standards for TSP sampling of particles less than $100 \mu\text{m}$ in diameter, and our protocols follow the US EPA recommendations for TSP collection and analysis [US EPA, 1999]. The sampler designs used in this study have been evaluated for the efficiency of particle capture with varying particle diameters and external wind speeds. There is evidence that the sampling efficiency of very large particles ($> 30 \mu\text{m}$) declines with increasing wind speed. For this reason, we would expect that our estimates of total particle load during dust events may under-sample concentrations in the large range of particle diameters, although the exact degree of under-sampling depends on a variety of factors and is therefore challenging to quantify under field conditions [Kenny *et al.*, 2005]. In this regard, the results presented here are a conservative estimate of TSP concentrations because of the potential to underestimate the presence of very large particles during high wind events.

2.2. Data Collection and Analysis

[10] Prior to sampling, the filters were weighed to the nearest microgram on a Mettler Toledo AX26 microbalance (Mettler Toledo, Columbus, OH, United States) under controlled temperature and humidity conditions. After weighing, filters were pretreated for three days in a Millipore deionized water (DI) leach. During the leaching process, DI water was changed daily. On the third day of leaching, filters were individually rinsed before being hung to dry. Once dry, filters were stacked in numerical order wrapped in foil and placed in a 500°C muffle furnace for 6 h. There was no measurable change in mass in filters before and after the leaching and combustion treatments. After combustion, filters were put in plastic bags presoaked in deionized water and individually triple rinsed. Each plastic bag was labeled with the filter ID number and the filter mass. Filters were then sent to the U. S. National Park Service personnel for field sampling.

[11] The TSP field sampling was conducted by installing precleaned filters onto the TSP sampler head using powder-free nitrile gloves. At the start of each sample period, the start date and time were recorded on the bag storing the filter. Filters remained on the TSP sampler for approximately two weeks during continuous sampling. At the end of each sampling period, filters were removed with nitrile gloves, folded in half to prevent sample loss, and placed in the bag that stored the filter prior to sampling. All samples were analyzed for flow restrictions (due to accumulation of dust on the filter), and no such cases were observed. Total flow was recorded for

all samples, and filters were sent to the University of Colorado Environmental Biogeochemistry lab for analysis.

[12] The TSP mass on each filter was determined by calculating filter mass (in mg) per cm^2 using presampling and postsampling filter weights. After measuring mass deposited onto the filter, the average TSP concentration in air ($\mu\text{g m}^{-3}$) per sample period was calculated from the measured TSP load and the total volume of air flow through the filter. Flow rates were corrected for site elevation and temperature using the average monthly temperatures recorded during the sampling period.

2.3. Comparisons Between TSP and IMPROVE Data

[13] IMPROVE $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ concentration was downloaded from the Views Data Wizard (<http://views.cira.colostate.edu/web/DataWizard/>). Data for 2011 were made available for use in this study by IMPROVE on a provisional basis as the 2011 IMPROVE data had not yet been released for public use. The IMPROVE samplers operate one out of every three days, and so it is not possible to directly compare the TSP and IMPROVE data on a daily or weekly basis. To compare biweekly TSP, $\text{PM}_{2.5}$, and $\text{PM}_{2.5-10}$ particle concentrations, we averaged the daily IMPROVE data across each (two week) TSP sampling period. By converting to a two-week average, we lose some of the high-frequency variation in $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ particle concentrations but are able to make a more appropriate comparison to the integrated two-week TSP measurements. The high frequency and smoothed records of $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ are shown in supporting information Figures 1 and 2. On an annual basis, the difference between an average calculated from the 24 h IMPROVE measurements versus two-week mean concentrations of $\text{PM}_{<2.5}$ and $\text{PM}_{2.5-10}$ particles is small (3.3% difference). On hourly to daily time scales, particularly during periods of dust storms, dust events, and dust haze generated by regional high winds, particle concentrations will greatly exceed the two-week mean for all particle-size classes.

[14] Below, we present TSP concentrations as a direct, biweekly measurement of total atmospheric particle concentrations. As noted above, we calculate a two-week moving average of IMPROVE $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ particle concentrations for comparison. The IMPROVE network samplers utilize a sample inlet that physically excludes particles larger than $10 \mu\text{m}$ in diameter, so the difference between the TSP measurements and the sum of IMPROVE $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ particle concentrations represents the particle-size class in excess of $10 \mu\text{m}$ (e.g., $\text{PM}_{>10}$). This calculated $\text{PM}_{>10}$ particle concentration is imperfect given the differences in sampling methodologies, but it provides insight into the relative variation of the different size classes through the sampling periods.

2.4. Particle-Size Analysis

[15] In order to determine the particle-size distribution (PSD) of the suspended particulate matter caught by the TSP filter for a small selected set of filters, we examined the particles both above and embedded in the filter. The former particles were directly suspended in an isopropanol solution (0.1 g particles per 25 mL solution), whereas the latter required extraction from the filter's silica fibers. One cm^2 was cut from the filter, put in 10 mL of isopropanol, and sonicated for 30 min. Three $15 \mu\text{L}$ aliquots from both well-mixed solutions were vacuum filtered on to a $0.2 \mu\text{m}$ Millipore

Table 1. Annual Mean, Minimum, and Maximum Particle Concentrations in $\mu\text{g m}^{-3}$ for the Canyonlands and Mesa Verde Sampling Sites^a

Sample Site	Annual Mean Concentration	Minimum Concentration	Maximum Concentration
Canyonlands 2009			
IMPROVE PM _{2.5}	2.8 (0.2)	1.8	5.3
IMPROVE PM _{2.5-10}	6.14 (0.7)	3.0	11.2
TSP	171.2 (25.7)	17.0	437.6
Canyonlands 2010			
IMPROVE PM _{2.5}	3.1 (0.3)	1.0	7.2
IMPROVE PM _{2.5-10}	7.5 (0.7)	1.6	13.7
TSP	113.3 (35.7)	10.5	700.4
Canyonlands 2011			
IMPROVE PM _{2.5}	2.4 (0.2)	1.29	4.08
IMPROVE PM _{2.5-10}	6.36(0.6)	2.36	14.5
TSP	134.1 (16.0)	15.4	277.9
Mesa Verde 2011			
IMPROVE PM _{2.5}	2.8 (1.9)	0.5	12.2
IMPROVE PM _{2.5-10}	6.7 (5.8)	1.0	36.7
TSP	90.2 (61.9)	6.2	248.3

^aNumbers in parentheses are standard errors determined from variability within the annual sampling period.

polycarbonate filter, carbon coated, and mounted on an adhesive sample stub. Samples were observed on a scanning electron microscope (SEM Model JSM-5800LV, JEOL, Tokyo, Japan), and the particle-size distributions were calculated using NSS 2.3 X-ray Microanalysis software (Thermo Fisher Scientific, Waltham, MA, USA). The filters used in this study are an imperfect means for quantitative evaluation of aerosol sizes because smaller particles are more likely to penetrate deeply into the filter, and we therefore present these data as a semiquantitative assessment of the sizes of particles collected on the filters.

[16] Seasonal variation of PSD was determined from Canyonlands TSP filters that collected particulate matter during two-week sampling periods in the late fall (10 November to 2 December 2008), spring (25 May to 8 June 2010), summer (2 June to 16 June 2009), and early fall (29 September to 13 October 2009).

2.5. Statistical Analysis

[17] To evaluate the temporal covariance of particle concentrations between PM_{<2.5}, PM_{2.5-10}, and PM_{>10} size classes, we use linear regression analyses for each study site. In these regressions, we examine whether significant correlations exist between the PM_{>10} and PM_{2.5-10}, PM_{>10} and PM_{2.5}, and PM_{2.5-10} and PM_{2.5} concentrations.

2.6. Environmental Controls on Particle Concentrations

[18] To evaluate the relations between the concentrations of TSP and wind speeds for individual sampling periods at

each site, we calculated a wind factor (Wf) for each sample period and conducted a regression between the two variables. The parameter Wf for each sample period was calculated using the Wf equation from the Revised Wind Erosion Equation (RWEQ) [Fryrear and Sutherland, 1999];

$$Wf = \left(\frac{W}{500}\right)^N$$

where Wf =wind factor (m s^{-1})³, N =number of days in the sample period, and W (W =wind value (m s^{-1})³) is calculated from the following equation:

$$W = \sum_{i=1}^N U_2(U_2 - U_t)^2$$

Where U_2 =average daily wind speed, and U_t is the threshold wind speed for wind erosion (5 m s^{-1}) commonly employed in the RWEQ model [Fryrear and Sutherland, 1999]. Sensitivity testing of this threshold value suggests minimal impact on the correlations through the use of higher or lower threshold values (data not shown).

[19] For CNP, daily wind data from the USGS Corral Pocket, Clim-Met site #2 station were used (<http://gce.cr.usgs.gov/projects/sw/clim-met/>) to calculate Wf . For the MVNP, National Climate Data Center daily wind speed from the Mesa Verde station was used (<http://www.ncdc.noaa.gov/cdo-web/datasets/GHCND/stations/GHCND:USC00055531>).

2.7. Dust-Storm Events and Back Trajectory Analysis

[20] To examine whether elevated concentrations at the CNP and MVNP sites were the result of regional scale (e.g., encompassing both sites) dust emission or more localized events, we carried out several different comparisons. The USGS maintains a dust-storm database that uses satellite

Table 2. Seasonal Mean Particle Concentrations in $\mu\text{g m}^{-3}$ for the Canyonlands and Mesa Verde Sampling Sites^a

Sample Site	Winter	Spring	Summer	Fall
Canyonlands 2009				
IMPROVE PM _{2.5}	3.3 (0.6)	2.8 (0.3)	2.5 (0.6)	2.6 (0.3)
IMPROVE PM _{2.5-10}	6.5 (2.4)	4.9 (1.2)	6.1 (1.6)	7.1 (1.5)
PM _{>10} *	74.1 (52.8)	154.2 (66.7)	181.8 (40.3)	198.0 (20.0)
TSP	78.2 (53.2)	169.6 (69.1)	192.4 (41.8)	211.6 (23.5)
Canyonlands 2010				
IMPROVE PM _{2.5}	3.3 (.4)	3.7 (1.0)	2.7 (0.5)	2.7 (0.5)
IMPROVE PM _{2.5-10}	9.8 (1.2)	7.7 (1.6)	7.2 (2.0)	6.1 (1.1)
PM _{>10} *	63.5 (6.87)	229.4 (124.6)	76.6 (23.3)	46.7 (9.7)
TSP	70.3 (7.5)	241.3 (125.1)	87.1 (23.3)	52.9 (9.5)
Canyonlands 2011				
IMPROVE PM _{2.5}	1.66(0.3)	1.9 (0.5)	0.8 (0.2)	1.0 (0.2)
IMPROVE PM _{2.5-10}	4.3 (1.3)	4.4 (1.2)	1.8 (0.5)	2.4 (0.8)
PM _{>10} *	24.5 (7.6)	186.5 (22.6)	155.4 (20.2)	116.2 (8.7)
TSP	29.7 (7.0)	198.3 (23.5)	165.9 (20.9)	123.3 (9.1)
Mesa Verde 2011				
IMPROVE PM _{2.5}	1.5 (0.7)	3.7 (2.6)	3.8 (1.6)	2.2 (0.8)
IMPROVE PM _{2.5-10}	2.9 (2.3)	9.5 (8.3)	9.5 (4.5)	4.2 (1.6)
PM _{>10} *	15.1 (5.47)	121.4 (10.7)	120.5 (21.1)	38.9 (3.2)
TSP	19.2 (14.4)	136.3 (27.7)	131.6 (59.1)	52.7 (18.4)

^aPM_{>10}* particulate concentrations are based on calculated biweekly differences between TSP and IMPROVE samplers and are not based on direct measurements of this fraction. Numbers in parentheses are standard errors determined from variability within each seasonal sampling period.

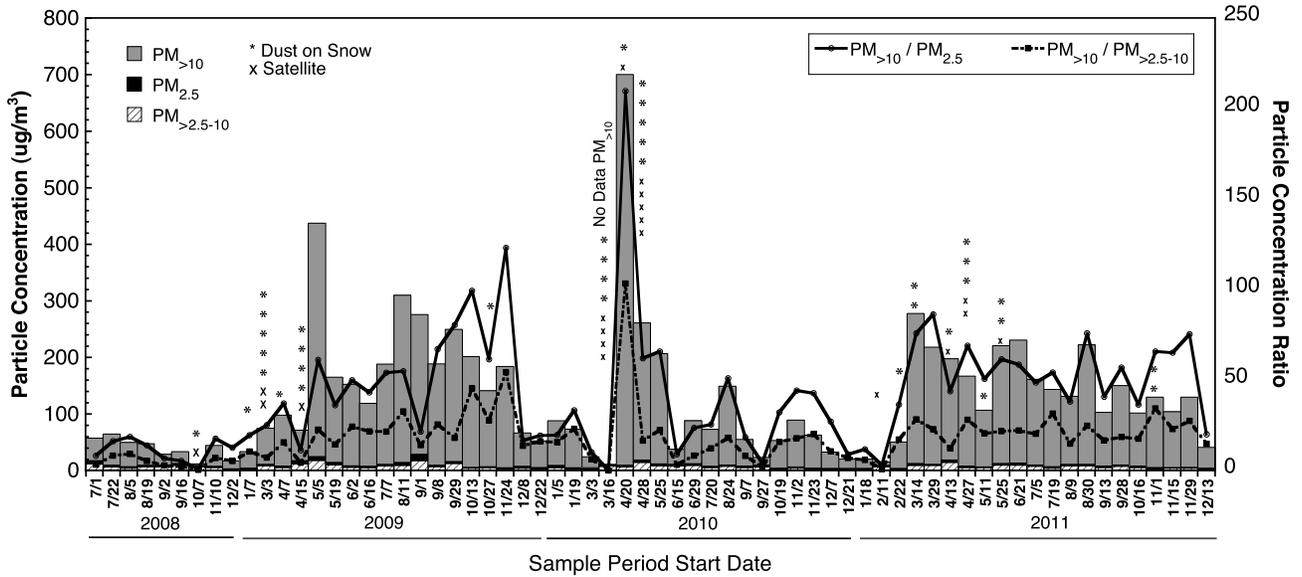


Figure 2. Average particle concentrations (in $\mu\text{g m}^{-3}$) at Canyonlands over each sampling period. $\text{PM}_{2.5}$ includes particles less than $2.5 \mu\text{m}$ in diameter, $\text{PM}_{2.5-10}$ includes particles with diameters from 2.5 to $10 \mu\text{m}$, and $\text{PM} > 10$ are larger particles calculated from the difference between TSP concentrations minus the sum of $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$.

observations to document dust storms during winter and spring seasons (http://sgst.wr.usgs.gov/dust_detection/dust-events/). A similar record of dust deposition on snowpack near Silverton, Colorado is maintained by the Center for Snow and Avalanche Studies (<http://www.snowstudies.org/CODOS/dustlog.html>). The Silverton site commonly receives storms that originate from the direction of the MVNP and CNP sampling sites and is therefore a useful location to detect regionally significant dust flux. Both of these databases provide insight into winter and spring events but unfortunately there is limited information on fall and spring events in the western U.S.

[21] To carry out a quantitative analysis of wind conditions during various periods of the year, we performed back

trajectory analysis for five two-week sampling periods across seasons in 2011 at both MVNP and CNP. For each of these two-week periods, daily Hysplit back trajectories were conducted independently for both MVNP and CNP using the web-based Hysplit Trajectory Model and archived GDAS climate data (<http://ready.arl.noaa.gov/hysplit-bin/trajtype.pl?runtime=archive>). Each daily back trajectory was run for 48 h using the vertical velocity model algorithm with a start elevation of 100 m above ground level. Each back trajectory was output into a GIS file. We estimated the average surface wind speed for each back trajectory using GDAS data downloaded from the NOAA ARL ftp server (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1>). Downloaded back trajectory GDAS time

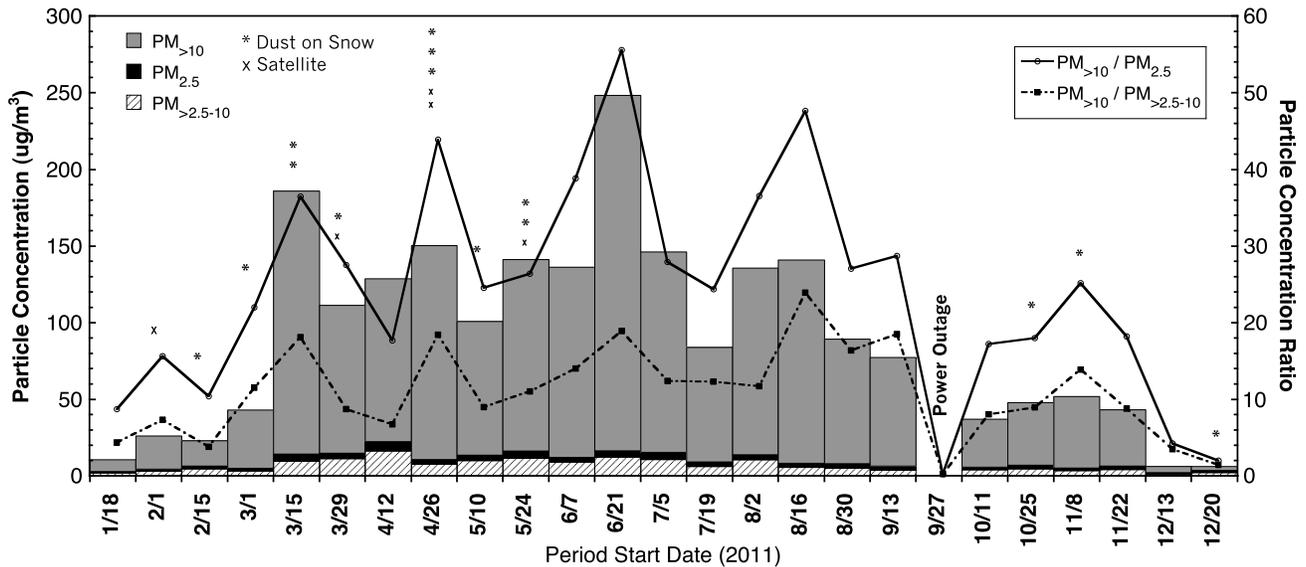


Figure 3. Average particle concentrations (in $\mu\text{g m}^{-3}$) at Mesa Verde over each sampling period. Particle size fractions as in Figure 2.

span and location specific wind data were then extracted using the Hysplit model averaged and then integrated into the back trajectory GIS outputs.

3. Results

3.1. Particle Concentrations

[22] The annual mean TSP particle concentrations at both CNP and MVNP were substantially larger than either the $PM_{2.5}$ or $PM_{2.5-10}$ concentrations. From June 2008 to December 2011 at the CNP site, the IMPROVE instrument measured annual mean concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ particles at 2.7 and 6.6 $\mu\text{g m}^{-3}$, respectively, but TSP concentrations over this same period averaged 126.6 $\mu\text{g m}^{-3}$. At MVNP, 2011 mean annual concentrations were 2.8, 6.7, and 90.2 $\mu\text{g m}^{-3}$ for $PM_{2.5}$, $PM_{2.5-10}$, and TSP fractions, respectively (Table 1).

[23] Particle concentrations of the TSP-size class tend to be higher in spring and summer than during other periods (Table 2), but the three-year record at CNP illustrates a large degree of interannual variability in concentrations and seasonal patterns. The highest seasonal TSP concentrations (three-month means) were found for CNP during spring 2010 and 2011 (241 and 198 $\mu\text{g m}^{-3}$, respectively). At CNP, summer-time TSP concentrations were lower than spring concentrations with the exception of 2009 at CNP when summer concentrations averaged 192 $\mu\text{g m}^{-3}$ and spring concentrations averaged 170 $\mu\text{g m}^{-3}$. Spring and summer TSP concentrations were nearly identical in 2011 at the MVNP site (about 130 $\mu\text{g m}^{-3}$) with lower concentrations in fall and winter. For the full multiyear record at CNP, there was substantial variation in average seasonal $PM_{2.5}$ and $PM_{2.5-10}$ concentrations (Table 2). In 2011, $PM_{2.5}$ and $PM_{2.5-10}$ concentrations at the MVNP site during the spring and summer were roughly 200–300% higher than the fall and winter concentrations, but similar relations were not found for the CNP site. Even with these seasonal inconsistencies, concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ were below 10 $\mu\text{g m}^{-3}$ for each season.

[24] A more detailed examination of the PM time series at both MVNP and CNP clearly illustrates the strong seasonality in particle concentrations and also highlights the roles that periodic windstorm-driven regional dust emission played in mineral aerosol loads at these sites (Figures 2 and 3). Through the time series 2008–2011, TSP concentrations at CNP were near to or exceeded 200 $\mu\text{g m}^{-3}$ during numerous months. At both locations in 2011, $PM_{>10}$ concentrations increased abruptly in March–April, with the onset of major dust storms in the region. In several cases, $PM_{>10}$ concentrations at CNP exceeded 300 $\mu\text{g m}^{-3}$ with the largest dust-deposition period, in 2010, having a two-week average concentration in excess of 700 $\mu\text{g m}^{-3}$. At both MVNP and CNP, particle concentrations increased substantially in late March/early April of each spring then declined (variably) through late fall.

3.2. Covariation of Particle-Size Classes Through Time

[25] During 2011, particle-size class concentrations at MVNP and CNP were strongly correlated. At MVNP, annual concentrations of $PM_{>10}$ particles were strongly correlated with $PM_{2.5-10}$ particles (linear regression analysis, $p < 0.001$, $r^2 = 0.62$) and with $PM_{2.5}$ particles (linear regression analysis, $p < 0.001$, $r^2 = 0.68$). Concentrations of $PM_{2.5-10}$ and

$PM_{2.5}$ particle were also strongly correlated (linear regression analysis, $P < 0.001$, $r^2 = 0.88$). During 2011 at CNP, significant linear relations (linear regression, all p values < 0.0001) were found between $PM_{>10}$ and $PM_{2.5-10}$ ($r^2 = 0.55$), $PM_{>10}$ and $PM_{2.5}$ ($r^2 = 0.58$), and $PM_{2.5-10}$ and $PM_{2.5}$ ($r^2 = 0.78$). In prior years at CNP, however, the relation between $PM_{>10}$ and $PM_{2.5-10}$ or $PM_{2.5}$ particles is more variable. In 2010, there were no significant correlations between $PM_{>10}$ and $PM_{2.5-10}$ or $PM_{2.5}$ particle concentrations. In 2009, $PM_{>10}$ and $PM_{2.5-10}$ particles are significantly but weakly correlated ($p < 0.001$, $r^2 = 0.43$) as are $PM_{>10}$ and $PM_{2.5}$ particles ($p < 0.001$, $r^2 = 0.31$). $PM_{2.5-10}$ and $PM_{2.5}$ particles at CNP were strongly correlated in both 2009 ($p < 0.001$, $r^2 = 0.67$) and 2010 ($p < 0.001$, $r^2 = 0.55$).

[26] The concentrations of $PM_{2.5}$, $PM_{2.5-10}$, and $PM_{>10}$ particle concentrations tend to rise and fall together seasonally and during periods of regional dust emission events. Despite the same general trend, however, dust storms led to a larger proportional increase in the $PM_{>10}$ (and TSP) particle concentrations compared to $PM_{2.5}$ or $PM_{2.5-10}$ concentrations. This behavior was illustrated in Figures 2 and 3 by the increase in the ratio of $PM_{>10}$ to $PM_{2.5-10}$ and $PM_{>10}$ to $PM_{2.5}$ concentrations during spring and summer. The larger temporal variation in $PM_{>10}$ (and TSP) compared to the $PM_{2.5}$ or $PM_{2.5-10}$ particle concentrations was also evident from comparison of the coefficient of variation (CV) values for the three particle-size classes on an annual basis. At CNP, the CV for TSP in 2011 was 0.89 compared to 0.52 for $PM_{2.5-10}$ and 0.53 for $PM_{2.5}$. At MVNP, the CV for TSP in 2011 was 0.69 compared to 0.59 for $PM_{2.5-10}$ and 0.47 for $PM_{2.5}$.

3.3. Particle Sizes

[27] Particle-size distributions (PSD) on the sampling filters at Canyonlands varied substantially by season. Samples collected in winter and summer had small mean-particle sizes (8.3 and 11.7 μm geometric diameters, respectively) and few particles $> 50 \mu\text{m}$ (Table 3). In spring and fall collections, particle sizes were larger with means of 38.8 and 20.1 μm , respectively. Maximum particle sizes ranged from 128 to 296 μm in diameter, but these sand-sized particles were rare. This may be the result of inefficient sampling of very large size particles in the TSP samplers. Most of the particulate matter embedded within the filter for all the sampling periods was less than 10 μm (mean = 2 μm , max = 30 μm) with little PSD variation across seasons. Images of the SEM analysis of filters are shown in Figure S3.

3.4. Influences of Wind Speeds and Regional Trajectories on Particle Concentrations

[28] The Hysplit analysis for events at both CNP and MVNP showed strong seasonal changes in the direction and characteristics of back trajectories from each sampling site. Both sites appeared to be influenced by similar wind directions and high wind events across each period of analysis (Figure 4). During the periods of very high TSP concentration associated with regional dust emission in March and June 2011, the back trajectories were dominated by wind flow originating from the southwestern U.S. During these events, average surface wind speeds along the back trajectories exceeded 10 m s^{-1} . During July, the winds at both sites had strong southerly components that were likely related to

Table 3. Particle-Size Parameters in Micrometers From TSP Sampler Filters From the CNP Sampling Site as Determined by SEM Analysis

	Summer 2009	Fall 2009	Winter 2010	Spring 2010
Mean	11.7	20.1	8.3	38.8
Median	7.8	14.2	5.2	14.5
Min	2.0	4.5	1.7	4.5
Max	128.2	236.7	244.0	296.0
Kurtosis	9.7	23.7	111.5	2.7
Skewness	2.6	3.8	7.9	1.8

convective storm, monsoonal flow, and that were generally weaker than earlier in the year. In October and December, there was evidence for some high wind events but with both southwesterly and northwesterly origins and lower overall wind speeds along the trajectories compared to spring and early summer periods.

[29] Local wind speeds had no statistically significant influence on particle concentrations at either the MVNP or CNP sites. The regression of biweekly TSP concentrations against the wind-factor term (related to the hours of wind speed above the erosion threshold of surrounding soils) yields r^2 values of 0.13 for MVNP and 0.18 for CNP. Variation of coefficients within the wind factor calculation had little impact on correlation coefficients.

4. Discussion

[30] The annual $PM_{2.5}$ particle concentrations from the IMPROVE sites at MVNP and CNP are amongst the lowest in the nation [Hand *et al.*, 2011; Li *et al.*, 2013; Murphy *et al.*, 2008]. $PM_{2.5-10}$ concentrations at both sites were considerably lower than annual averages for other areas of the southwestern U.S. and comparable to many other less arid regions of the country [Li *et al.*, 2013]. In contrast, annual TSP concentrations at both sites in 2011 were considerably higher than the other particle-size classes with a three-year mean TSP at Canyonlands of $126 \mu\text{g m}^{-3}$. This value is higher than 60% of the annual TSP concentrations in 115 large urban areas reported in Baldasano *et al.* [2003] and comparable to annual mean TSP concentrations in a number of Asian and African cities that are in the paths of frequent dust storms [Baldasano *et al.*, 2003; Ozer *et al.*, 2006; Xiao and Liu, 2004].

[31] The TSP concentrations at both MVNP and CNP were strongly seasonal with very low total particle concentrations during winter and with much higher concentrations during spring and summer. These patterns are similar to the seasonal cycle of dust-storm activity in northern Utah that also shows large spring and summer peaks [Hahnenberger and Nicoll, 2012]. Dust-storm events affecting the Wasatch Front of Utah typically last 1–2 days [Steenburgh *et al.*, 2012]. This type of pattern appears to occur at both MVNP and CNP where $PM_{2.5}$ and $PM_{2.5-10}$ (Figures S1 and S2) have distinct daily spikes in concentration likely associated with short-duration dust storms as discussed below. The TSP concentrations measured in this study were for two-week averages and therefore spanned periods of elevated concentrations during short-term dust storms and periods of relatively lower concentrations. For comparison, dust storms in and around the Sahara Desert result in TSP concentrations in excess of $2000 \mu\text{g m}^{-3}$ for a 24 h period [Eliasson *et al.*, 2009]. In

Beijing, China, TSP concentrations during periods of heavy dust fall associated with wind storms to the west average $2170 \mu\text{g m}^{-3}$ [Zhang *et al.*, 2010]. The systematic elevations of TSP in spring and summer months at both MVNP and CNP, coupled with the observation that most dust storms have short duration, suggest that 24 h TSP concentrations at CNP and MVNP would be much larger than the two-week averages and perhaps comparable to values commonly observed during dust storms in Asia and Africa.

[32] Few long-term measurements of TSP have been made in the U.S. in recent years, and we are aware of no published annual time scale measurements of TSP near the dryland areas of the western U.S. since the adoption of the PM_{10} air quality standard in 1988. Prior to this time period, work by Lundgren *et al.* [1984] indicated that total particle concentration could exceed $100 \mu\text{g m}^{-3}$ in urban settings for short sampling periods and particularly in those close to mineral aerosol source areas such as Riverside, CA, and Phoenix, AZ. In these prior studies, however, and in many other studies in urban areas [e.g., Marazzan *et al.*, 2002], the TSP concentration was dominated by particles in the $PM_{2.5-10}$ and $PM_{2.5}$ size classes. The results of this study indicate that TSP concentrations are considerably higher than the $PM_{2.5-10}$ and $PM_{2.5}$ particle concentrations measured at the adjacent IMPROVE monitoring sites.

[33] Several interpretations can be considered to account for these observed differences between TSP and the smaller particle concentrations. The most likely interpretation—that TSP samplers captured large quantities of $PM_{>10}$ —was supported by observed populations of large particles, commonly 10 to 40 μm in diameter, on the TSP filters. The particle-size range observed in this study was consistent with observations made from dust collected in snow cover at a site in the San Juan Mountains (~ 200 km from the Canyonlands site and 100 km from the Mesa Verde site). At this 4000 m elevation site, the particle-size distribution of collected dust indicates that particles between 10 and 50 μm make up approximately 50% of the mass fraction of dust fall [Lawrence *et al.*, 2010]. Dust-deposition measurements made using passive collectors (marble traps) in this region near CNP suggest that approximately 35% of particles are in the size range between 10 and 53 μm [Reheis, 2002]. Similarly, particles greater than 10 μm make up an estimated 61% of the total particle mass during dust storms in Beijing, China [Zhang *et al.*, 2010], so our findings are not unexpected in a global context.

[34] Another possibility that could explain the difference between TSP and the smaller particle size classes is sampling bias if either the high-volume TSP samplers were systematically biased high or the IMPROVE samplers were biased low. Neither of these possibilities seems likely as both types of samplers follow well-established sampling norms and are periodically calibrated. Continuous monitoring of flow on the TSP instruments during these measurements did not raise concern, showing very little variation in flow even during periods of high-TSP concentrations. The TSP samplers do not sample all particles with 100% efficiency, but sampling efficiency generally decreases with increasing particle size (particularly over 30 μm) thus leading to lower-than-actual total mass collection in the sampler [Watson *et al.*, 1983]. For this reason, the TSP concentrations are more likely to be an under-measurement than an over-measurement of total particle concentration in the atmosphere.

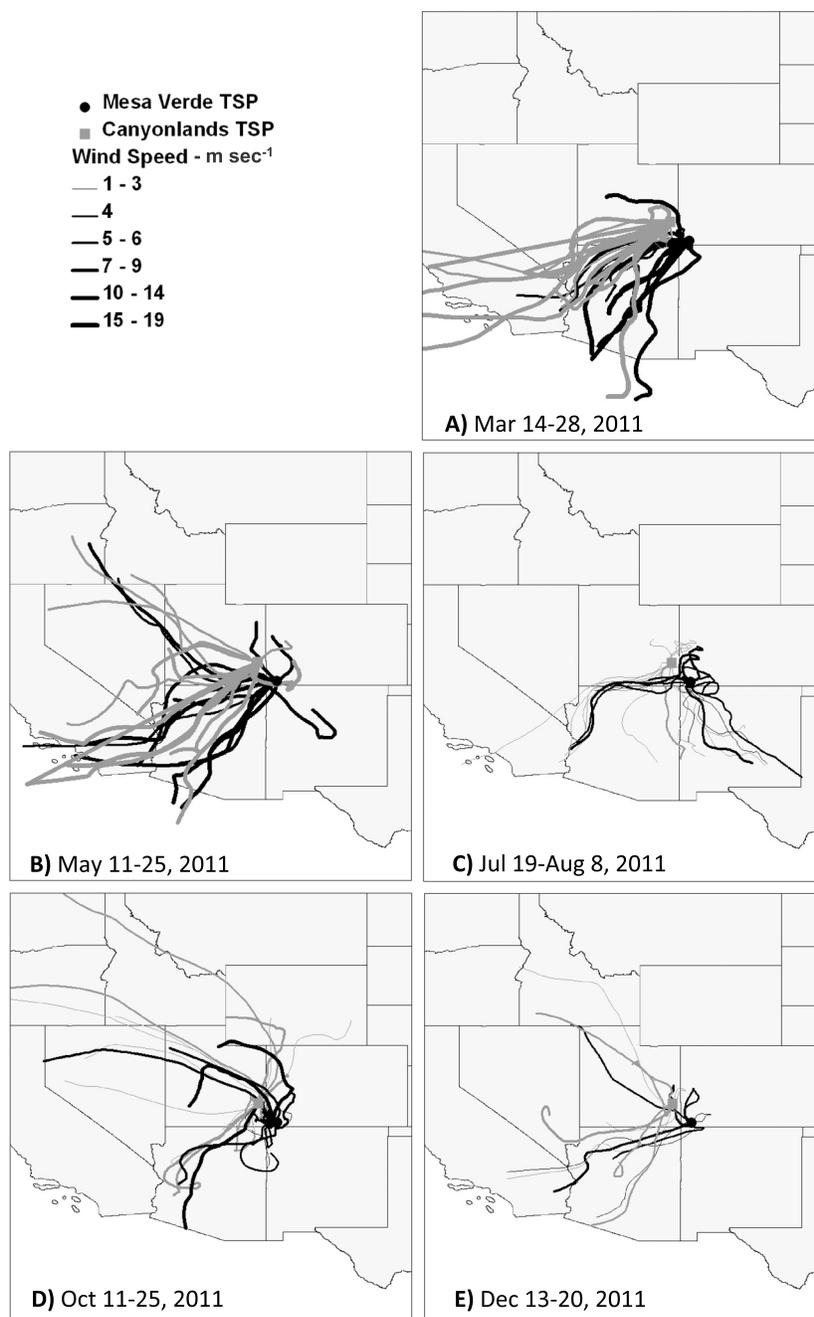


Figure 4. Forty-eight hour back trajectories from the Hysplit model for five total suspended particle (TSP) sampling periods shown in Figures 4A–E. Each back trajectory is shown for CNP (grey) or MVNP (black) and the width of the line reflects the average wind speed along the course of the trajectory.

[35] A third possibility for the discrepancy in IMPROVE/TSP concentrations is that the samplers systematically sampled different air masses with a bias toward sampling of local particles in the TSP sampler (e.g., derived from the immediate surroundings). The placement of the samplers on rock outcrops near large cliff faces with very few and very small potential dust sources nearby renders this explanation highly unlikely.

[36] We cannot completely rule out the second and third alternative scenarios as causes for some of the PM-size discrepancies. Nevertheless, our measurements of dust flux and particle sizes, integrated and compared with observations of dust emission, point strongly to the deposition of particles

larger than $10\ \mu\text{m}$ that increase and decrease seasonally in response to regional dust emission during regional windstorms.

4.1. The Role of Dust Storms in Particle Concentrations

[37] The period of elevated TSP concentration coincides with known dust-storm activity in the region. Although the National Oceanic and Atmospheric Administration maintains a storm event database (<http://www.ncdc.noaa.gov/stormevents/>) that includes ground-based observations of dust storms, data are lacking for a number of states, including Utah. Nevertheless, a valuable record of dust storms is provided from archived satellite retrievals during winter and



Figure 5. Comparison of visual conditions at the Canyonlands sampling site during a high-TSP period (right, 21 March 2011), compared with a low-TSP period (left, 23 December 2011); http://esp.cr.usgs.gov/info/regional_cams/index.html.

spring since 2009 (http://sgst.wr.usgs.gov/dust_detection/dust-events/). Moreover, continuous records of dust deposition on snow cover have been developed for the San Juan Mountains of Colorado [Lawrence *et al.*, 2010; Painter *et al.*, 2007; <http://www.snowstudies.org/CODOS/dustlog.html>]. Figure 4 illustrates a dust event at CNP in March 2011 that was subsequently measured in snowpack 200 km to the east in the San Juan Mountains. Using these records, it is clear that periods of high-TSP concentration at MVNP and CNP tend to coincide with many observed regional dust events (Figures 2 and 3). However, numerous periods of relatively high-TSP concentrations lack direct, corresponding observations of dust emission. This circumstance is not surprising given that dust storms commonly go undetected because of their lack of sufficient magnitude to be captured by satellite retrievals, or occur at night and (or) under cloud cover. Smaller scale or more localized events can also have an important influence on TSP concentrations over extended periods of time (Figure 5).

[38] The Hysplit back trajectory analyses illustrate two key points. First, the back trajectories of airflow into MVNP and CNP are similar across seasons suggesting both sites are influenced by similar large-scale airflow. In March and May, both sites experience strong southwesterly flow with average surface wind speeds along the trajectories well in excess of 7 m s^{-1} , a value sufficiently high to cause wind erosion in most soils [Gillette and Passi, 1988]. In July, concentrations of TSP at both sites were still elevated relative to the annual means but are lower than spring peak concentrations. During this time period, winds at both sites had a southwesterly or southeasterly direction but generally with much lower wind speeds. The airflow patterns for October are much more variable, including a northerly component, and these patterns coincide with lower dust concentrations at both sites. The back trajectories for December have mostly westerly origins but are lower yet in average wind speed and correspond with the low point in annual TSP concentrations at both sites. These analyses strongly imply that (1) strong winds associated with eastward movement of Pacific frontal storm systems during the spring contribute to elevated TSP concentrations at both sites, and (2) summertime events result from similar types of storms or from strong winds associated with convective activity [Steenburgh *et al.*, 2012]. Frontal storms traverse this region in the winter but typically

do not result in high concentrations of TSP at these two sites likely because of higher surface soil moisture across much of the American Southwest, along with intermittent snow cover.

[39] The three-year observation record at CNP illustrates a large degree of interannual variation in seasonal mean TSP concentrations with elevated concentrations near $200 \mu\text{g m}^{-3}$ from April to November 2009 and 2011. In comparison, 2010 TSP concentrations exceeded $700 \mu\text{g m}^{-3}$ over a two-week period in April before declining to much lower concentrations for the remainder of the year. To examine whether these interannual variations in TSP concentrations are consistent with regional drought cycles, we compared seasonal TSP concentrations at CNP to a January–September average Palmer Modified Drought Index (PMDI) estimate for the southwestern US for 2009–2011 (<http://www.ncdc.noaa.gov/temp-and-precip/ranks.php>). In 2009 and 2011 when TSP was elevated through the entire spring/summer period, the PMDI for the region was -16.0 and -17.6 indicating drought conditions. In contrast, the PMDI for 2010 was 11.9 indicative of wetter than average conditions and temporally consistent with comparatively low summer TSP concentrations at CNP. Although these comparisons are subjective, they provide one potential explanation for the strong interannual variation in TSP concentrations at the CNP site.

4.2. The Sources of Fine, Coarse, and Very Large Particles

[40] The sources of $\text{PM}_{<2.5}$, $\text{PM}_{2.5-10}$, and $\text{PM}_{>10}$ particles can be diverse and variable over time. In many urban settings, $\text{PM}_{2.5}$ is associated with industrial sources including the combustion of fossil fuels whereas the $\text{PM}_{2.5-10}$ fraction and the TSP fraction are generally associated with the wind erosion of soils [Li *et al.*, 2013; Pakkanen *et al.*, 2001]. In arid regions and downwind of large deserts, however, all three particle classes can be associated with wind erosion of soils and dust emission [e.g., Chan *et al.*, 2005]. In this study, we examined the annual correlations among the different particle size concentrations to better understand the potential sources of the different particle-size classes. A high degree of correlation between two particle size classes would provide some indication of a common source and/or transport mechanism, whereas a lack of covariation might indicate different sources and/or transport mechanisms. The results of this analysis for 2011 at both MVNP and CNP indicate a high

degree of correlation among all three-size classes. This result suggests that all three particle-size classes are associated with similar meteorological conditions and/or source regions, as has been interpreted for individual dust storms in other regions [Chan *et al.*, 2005; Eliasson *et al.*, 2009]. In 2009 and 2010 at CNP, however, the correlation between $PM_{2.5}$ and $PM_{2.5-10}$ particles was consistently robust but the correlations of $PM_{>10}$ to smaller particles were weak (2009) to nonexistent (2010). These patterns suggest a complex relation among particle-size classes at these sites and may indicate variable contributions to different PM size classes across years. The lack of any statistical correlation between the large and smaller size classes in 2010 is particularly notable given that this is the wettest year from 2009 to 2011. This raises the possibility that drought and wind erosion leads to elevated concentrations of $PM_{>10}$, $PM_{2.5-10}$, and $PM_{2.5}$ concentrations but during wetter years, these particle size classes may be the result of a different mix of sources. This conclusion is speculative but highlights the need to better understand the provenance of particles across the region and over a range of size classes during a variety of meteorological conditions.

[41] Across all years, the large dust storms in spring and summer at CNP and MVNP appear to result in large spikes of $PM_{>10}$ relative to both $PM_{2.5-10}$ and $PM_{2.5}$. Nearly all high-TSP periods are accompanied by an increase in the ratio of $PM_{>10}$ to the small size classes. Although many dust storms appear to cause spikes in both $PM_{2.5-10}$ and $PM_{2.5}$, the effect of these events appears to be much greater for the $PM_{>10}$ size class. Evidence for such increases is also seen in the analysis of particle sizes on TSP filters at CNP. The large mean-particle diameters in springtime dust collections (in contrast to a median value of 15 μm) and relative to other seasons indicate the capacity for high wind events to transport medium silt mostly less than 30 μm . Overall, the ratio of $PM_{>10}$ to both $PM_{2.5-10}$ and $PM_{2.5}$ particles is larger than observed in other U.S. locations, such as Lubbock Texas [Stout and Lee, 2003], and this ratio is highly variable through time particularly over the longer CNP record. There are extended periods of time when these ratios are relatively low followed by abrupt and pronounced changes.

[42] In prior work examining the provenance of dust storms in the San Juan Mountains of SW Colorado, we used Sr and Nd isotopic markers to identify potential sources of the minerals in large regional dust storms. These studies indicate that dust storms can have variable provenance across the season but are also suggestive of emissions across a wide area of western deserts as opposed to a single geologic setting [Lawrence *et al.*, 2010; Neff *et al.*, 2008]. More work is required to fully understand the origins of dust at these sites and the drivers of both seasonal and interannual change. Nevertheless, these data clearly highlight the complexity and variability of particle-size class relations through time and the need to better understand the origins of different particle-size classes on an annual and seasonal basis.

4.3. Future Prospects

[43] The presence of elevated concentrations of atmospheric dust in the western U.S. is relevant to both air quality and human health. Human activity, coupled with expected increasing aridity [Seager *et al.*, 2007], will influence the amount of dust in the American West and the particle sizes in the dust. In the western U.S., recent studies reveal that

disturbances of dry landscapes after European settlement [Belnap and Gillette, 1998; Belnap *et al.*, 2009; Gill, 1996; Li *et al.*, 2009; Miller *et al.*, 2012; Munson *et al.*, 2011] have increased dust activity. Lake-sediment studies indicate large and sustained increases in dust deposition to high-elevation settings that coincide with the expansion of human activity in this region between 1850 and the present [Neff *et al.*, 2008; Reynolds *et al.*, 2009]. As discussed in the foregoing, very large particles are an important component in deposited dust [Lawrence *et al.*, 2010; Reheis, 2002; Reynolds *et al.*, 2013]. Similar observations have been made for South America [McConnell *et al.*, 2007] and Asia [Chu *et al.*, 2009]. Satellite-retrieval-based analyses of dust sources yield estimates that 25% of global dust results from human activity with higher fractions of anthropogenic dust in the populated deserts in Asia, Australia, and North America [Ginoux *et al.*, 2012]. Recent work suggests a large increase in dust deposition downwind of desert regions in the U.S. over the past 17 years [Brahney *et al.*, 2013]. The resulting conclusion from evidence gathered in populated deserts around the world is that human activity can and frequently does increase the emission of mineral aerosols to the atmosphere in many arid regions. Increasing dust emission in the American West, especially associated with human activities, will likely be accompanied by continued, significant emission of very large dust particles. These expectations underscore the need to document and comprehend the full extent of contemporary dust loading to the atmosphere and environments across the region.

5. Summary

[44] This study illustrates the important role that large mineral-dust particles play in total particle concentrations at two high-elevation sites in dryland settings at the edge of Colorado Plateau in the western U.S. Although large particles travel shorter distances through the atmosphere than smaller ones, these measurements indicate that residents living on or near the Colorado Plateau experience much higher exposure to mineral aerosols than would be indicated by measurements of $PM_{2.5}$ and $PM_{2.5-10}$ size fractions. The conventional wisdom is that large particles have little impact on human health, yet recent studies in Asia and elsewhere suggest that these particles may not be entirely benign [Kwon *et al.*, 2002; Schleicher *et al.*, 2011; Zhang *et al.*, 2002]. Our observations, combined with evidence for past [Neff *et al.*, 2008] and present [Brahney *et al.*, 2013] human disturbances of dust transport and deposition in this region, indicate a need to understand better the causes and consequences of elevated TSP concentrations on the Colorado Plateau and perhaps in other regions of the American Southwest.

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