Variability of Natural Dust Erosion from a Coal Pile*

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ABSTRACT

A study of fugitive dust emissions from a pile of crushed coal revealed that, in addition to dust being emitted into the atmosphere during periods of pile-management (human) activity, it is also emitted during periods without human activity. This “natural” emission in itself is not surprising given past work on wind erosion of particulate matter from aggregate piles. However, hourly downwind measurements of fine particle (PM_{10}) mass concentrations at two sites revealed that excessive dust was present in the air even when wind speeds were below the erosion threshold estimated from nearby wind speed measurements and regulatory guidance on coal pile aerodynamic characteristics. During periods of natural emissions with higher wind speeds, downwind concentrations were strongly associated with μ^2 —the squared excess of 1-min maximum wind speed above the erosion threshold—consistent with previous work on wind erosion potential. However, 88% of hourly concentrations coincided with lower winds for which wind speed was not a good predictor of airborne dust levels. Evidence was found that natural low-wind PM_{10} concentrations varied significantly with relative humidity, air temperature, and turbulence parameters (σ_u and σ_w). Smoke from coal combustion was ruled out as a significant factor in PM_{10} levels, but statistical evidence along with visual observation suggests that microscale turbulent airflows, including dust devils, were a significant source of PM_{10} during low wind speeds over the pile. The localized behavior of the turbulence makes it very difficult to develop a strong statistical model of natural downwind concentrations on the basis of off-pile meteorological measurements.

1. Introduction

Fugitive emissions are emitted gaseous or aerosol materials that do not pass through vents, stacks, or other openings. These emissions can significantly affect air quality near a source and, by their nature, are difficult to quantify. Various indirect techniques have been used to estimate fugitive particulate emissions—from sources such as bare soils (Gillette et al. 1980, 2004; Axtell and Cowherd 1984; Saxton et al. 2000; Harris and Davidson 2009), aggregate storage piles (Badr and Harion 2007;

Turpin and Harion 2009), or unpaved roads (Cowherd et al. 1974)—and relate them to physical processes using empirical methods or mathematical modeling. Here we describe a study of fugitive particulate emissions from a Tennessee coal pile. The objective of the study is to characterize the impact of coal pile dust emissions on downwind air quality.

This analysis is based on measured particulate data collected upwind and downwind of the pile along with video imagery and detailed meteorological measurements. We often found enhanced particulate levels downwind of the pile in the absence of human activity even when measured wind speeds were below the threshold normally considered significant for wind erosion by way of particle saltation. Video evidence and measurements confirmed that dust clouds originated over the pile, particularly (but not exclusively) during the daytime when solar radiation levels were high. Downwind measurements indicated natural particulate concentrations that were usually less than those

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observed when human disturbances affected the pile but were, nonetheless, important for separating natural from anthropogenic influences on dust generation. In the present context dust refers to particles smaller than $10\text{ }\mu\text{m}$ in size (PM_{10}).

Coal piles are prone to the spontaneous combustion of dust content. Powder River basin subbituminous coal is dusty and especially prone to burning in storage (Nugroho et al. 2008). Combustion within piles is usually a slow smoldering process [U.S. Environmental Protection Agency (EPA) 1978] producing aerosols typically $\ll 1\text{ }\mu\text{m}$ in diameter (Perera and Litton 2011). Uptake of water vapor and other condensable gases will—along with particle aggregation—result in particle size growth but these particles generally remain in the so-called $\text{PM}_{2.5}$ size range (i.e., $<2.5\text{ }\mu\text{m}$) close to the point of emission. During one extreme case, a large coal fire produced measurable increases in both $\text{PM}_{2.5}$ and PM_{10} downwind of the coal pile. The smoke contribution was, as expected, most pronounced for $\text{PM}_{2.5}$, but this event demonstrates that under certain conditions fire emissions can influence downwind particulate levels relevant to this study.

Wind erosion of particulate matter from soils and aggregate piles has been studied previously, empirically (Cowherd et al. 1974; Saxton et al. 2000) or with models (Ono 2006; Laurent et al. 2009; Harris and Davidson 2009). Blended methods that used both modeling and observations have also been published (Gillette et al. 2004; Turpin and Harion 2009). Dust emissions from aggregate storage piles are summarized in Cowherd et al. (1974), in Watson et al. (2000), and by the EPA (1995). A common issue examined across most studies is the extent to which wind speed affects particle emissions.

Cowherd et al. (1974) did not find a particle concentration dependence on speed downwind of crushed rock and sand piles. They stated that, while measured concentrations were sensitive to the inverse of wind speed, emission rates likely had a positive dependence on speed and these two effects tended to obscure the true relationship. Saxton et al. (2000) and Gillette et al. (2004) assumed that PM_{10} emission flux from dry bare surfaces was proportional to the horizontal flux of wind-blown particles larger than $10\text{ }\mu\text{m}$ (i.e., by way of saltation or the “hopping” of large particles across the ground and the subsequent dislodging of smaller particles). Gillette et al. estimated PM_{10} wind erosion emission rates that ranged over four orders of magnitude for speeds $> 5\text{ m s}^{-1}$.

Laurent et al. (2009), summarizing experimental data published by Bagnold (1941) and Chepil (1945), describe how the threshold wind friction velocity u_t^* (a predictor of particle entrainment from the surface) has

a minimum for particles $80\text{ }\mu\text{m}$ in size. Smaller particles resist erosion due to cohesive forces—electrostatic or those associated with the surface moisture—binding them with other surface particles. For $10\text{-}\mu\text{m}$ particles $u_t^* \approx 0.5\text{ m s}^{-1}$ and exceeds 1 m s^{-1} for particles $\leq 2.5\text{ }\mu\text{m}$.

Harris and Davidson (2009) modeled physical processes resulting in the emission of $1\text{--}100\text{-}\mu\text{m}$ particles into the air by wind action. Particles $<1\text{ }\mu\text{m}$ are bound too tightly to the surface to become airborne. Modeling indicated that aerodynamic forces were responsible for particle suspension when friction velocity $u^* \leq 0.6\text{ m s}^{-1}$. Above $u^* = 0.85\text{ m s}^{-1}$ saltation was the cause of nearly all particle suspension. Harris and Davidson computed particulate emission rates that varied four to five orders of magnitude for different surface characteristics and $0.1 \leq u^* \leq 1.0\text{ m s}^{-1}$. The simulated mass median diameter of emitted particles was $6\text{--}9\text{ }\mu\text{m}$.

Turpin and Harion (2009) examined wind erosion from aggregate material piles using a computational fluid dynamics model of airflow over flat-topped piles of different configurations relative to the mean wind direction. The key parameter in analyzing this process is $\rho_u = u_s/u_r$, where u_s is wind speed at 0.25 m above the pile surface and u_r is a reference speed at 10 m . The value of ρ_u increases bottom to top along the windward edge of the pile and varies along lateral edges because of localized accelerations. Results also indicated the fraction of pile surface area subject to wind erosion due to variations in ρ_u . Pile heights varied from 13 to 16 m , and piles were about 75 m in length. With $u_r = 5\text{ m s}^{-1}$ and wind gusts of up to 10 m s^{-1} , the fraction of total surface area of a pile theoretically subject to wind erosion varied between 46% and 55% across the different test configurations. These fractions decreased to 19% – 30% for the same mean wind but with wind gusts reduced by half.

Regulatory guidance on estimating wind-generated fugitive particulate emissions (EPA 1995; Watson et al. 2000) is consistent with the studies previously described. Specifically, wind emissions (i) are estimated to increase greatly with wind speed and gustiness (turbulence), (ii) are sensitive to the dimensions and orientations of material stockpiles, and (iii) tend to produce the greatest emission rates for particles near or just below $10\text{ }\mu\text{m}$ in size. Based on the EPA AP-42 Emissions Handbook (EPA 1995, with subsequent updates), wind erosion potential P_w from a dry aggregate storage pile is given by

$$P_w = 58(u^* - u_t^*)^2 + 25(u^* - u_t^*), \quad (1)$$

with $P_w = 0$ for $u^* \leq u_t^*$. The nonlinear relationship between P_w and u^* requires that the former be computed considering the spatial and temporal variation of u^* on the pile. AP-42 provides guidance on how to estimate u^*

variations due to pile geometry and orientation relative to wind direction and recommends using the peak wind speed measured at a short time interval to determine temporal variation in u^* . Given a peak wind speed at 10 m (u_{10}^+), it is possible to estimate a peak surface wind speed u_s^+ for air flowing over a pile as

$$u_s^+ = \rho_u u_{10}^+, \quad (2)$$

where ρ_u is the wind speed ratio described by [Turpin and Harion \(2009\)](#). For a typical storage pile, $\rho_u = 0.2$ near the pile base, rises to 0.9 just below the pile top, and reaches 1.1 at the edge of the top where the wind accelerates as it crosses the pile. Assuming a pile roughness length of 0.5 cm and a log wind profile,

$$u^* = 0.10 u_s^+. \quad (3)$$

AP-42 lists u_t^* as ranging from 0.55 m s^{-1} (at the base of a crushed coal pile) to 1.12 m s^{-1} (uncrusted pile) giving a factor-of-2 difference in u_t^* on a coal pile. The former value is a lower limit in estimating wind erosion and is the threshold (along with its wind speed equivalent u_t) referenced later in this paper. This value is not absolute because small variations in pile structure and composition can change the response of surface particles to wind action at the surface ([Watson et al. 2000](#)). Thus, u_t^* is used here as a benchmark for separating hours with higher and lower wind speeds that are more or less likely to experience particle entrainment by way of ambient airflow across the coal pile.

Direct wind erosion of particles from soils and aggregate storage piles requires wind speeds with sufficient force to overcome binding forces, but dust can be lifted from a surface when mean speeds are lower than the entrainment threshold. This occurs when a surface is subjected to the action of turbulent eddies that are likely to form on a sunny day over a dark-colored surface such as a coal pile. An extreme example of turbulent eddies occurs in the form of dust devils. Even at night it is possible that dust may be entrained into the air above a storage pile by the action of air moving across the pile. This is somewhat counterintuitive given our expectation that nighttime winds and turbulence are lower than during the daytime.

[Renno et al. \(1998\)](#) describe dust devils as “heat engines” driven by sensible heat flux at the surface and formed from vorticity created by ambient wind shear. A dark coal pile is a strong absorber of solar radiation and the sloped pile surface warms fastest when the solar angle of incidence is optimally aligned to receive the maximum radiation exposure. Airflow over a pile can force localized wind shear depending on wind direction, pile shape, and orientation. The potential exists during daytime for a coal pile to experience conditions suitable

for forming rotating convective turbulence and, in the extreme, dust devils. These microscale circulations potentially enhance localized wind speed near the surface and drive particle entrainment into the boundary layer.

2. Field measurements

A study of fugitive dust emissions from a coal storage pile was conducted June–November 2012 at the Gallatin electric power generating station [see the online supplementary material (SM) that accompanies this paper for details]. The 6.6-ha elliptical coal pile has a long axis (northwest–southeast) of 380 m and a short axis (northeast–southwest) of 220 m. Pile topography is represented in [Fig. 1](#) using approximate 5-m height contours. The actual height and contours change as coal is added or removed. However, the pile is maintained roughly in a constant configuration as shown with a narrow flat top. During the latter part of the study (after 14 October 2012) there was a net removal of coal from the southern end of the pile but the maximum pile height (~ 15 m) did not visibly change. The northwest and southwest faces of the pile were steep such that bulldozers did not drive across them. Bulldozers did drive across the rest of the pile, including its top. The conditions examined here are for hours when airflow passed over the upwind face of the pile from southerly and westerly directions. According to AP-42, the highest wind accelerations under such conditions occur along the leading and lateral edges of the pile top, shown by the shaded region in [Fig. 1](#). These are the locations where direct wind removal of coal dust is most likely. A few infrequent minor sources of fugitive dust exist near the studied pile as described in SM but their impacts on downwind measurements are considered insignificant. The pile was viewed continuously using a time-lapse camera and this information was critical in detecting both human activity and natural phenomena occurring around the coal pile.

Measurement systems deployed for this study are identical to those described by [Mueller et al. \(2013\)](#) for a study of fugitive dust emission from fly ash disposal. Particulate measurements ([Table 1a](#)) were made at one upwind site—140 m south-southeast of the pile’s eastern end (not seen in [Fig. 1](#))—and two downwind sites labeled site 2 and site 3 in [Fig. 1](#). Hourly particle measurements were made at each site using a pair of beta attenuation monitors (BAMs) with separate 2.5- and 10- μm selective inlets. The particle monitors are an EPA-approved federal equivalence method for monitoring fine particles. Pre- and poststudy tests quantified the measurement precision of each BAM under clean air conditions such that they varied within $\pm 3.6 \mu\text{g m}^{-3}$ when the mean concentrations of input airstreams were

FIG. 1. Approximate topographical representation of the coal pile. White lines denote approximate 5-m elevation contours (contour intervals not necessarily to scale). Pile base elevation and pile boundary are marked by the outermost contour line. The innermost closed contour represents a relatively flat pile top; but this was not always the case.

essentially zero. Thus, measurement differences of $<4 \mu\text{g m}^{-3}$ are not considered significant, consistent with previous results (Mueller et al. 2013). This creates more uncertainty in fugitive dust impacts when nonzero measured PM_{10} levels are $<4 \mu\text{g m}^{-3}$.

Meteorological measurements were made near the pile in a grass-covered field about 85 m from the coal pile edge. Instruments were mounted on or near a 10-m tower (Table 1b). The tower had cross arms mounted at 2.3 and 9.6 m above ground that held three-dimensional sonic anemometers on one end and mechanically aspirated temperature and relative humidity sensors on the other. Radiation flux was also measured at the 2.3-m tower level. Precipitation was measured with a nearby rain gauge. A nephelometer at site 2 measured light scattering from airborne aerosols and was most sensitive to particles $<1 \mu\text{m}$ in size. A soil moisture sensor was buried near the tower. All data were collected and archived at 1-min intervals.

3. Analysis

Air blew across the coal pile and past sites 2 or 3 when the 10-m wind direction was between south-southeast

and west-southwest (Table 2). Overall, these conditions occurred for about one-quarter of the study. Wind speed varied only slightly by direction for airflow crossing the pile, averaging 2.4 m s^{-1} over all of these hours. Hours were eliminated from analysis when precipitation was measured at the site. The remaining hours were classified according to whether human activity was observed on or near the pile. Here we are concerned primarily with those hours when no activity occurred.

Over 800 rain-free hours occurred when airflow passed over the pile. Excluding hours with precipitation may bias the results somewhat toward drier periods, but little dust erosion is expected when the coal surface is wet. Missing PM_{10} data slightly reduced the total number of hours analyzed. Table 3 summarizes differences between valid pairs of upwind and downwind PM_{10} concentrations. The fraction of all hours when the upwind PM_{10} measurement was significantly greater than the downwind measurement was 5.4% and 3.6% for sites 2 and 3, respectively. Upwind–downwind differences were negligible—that is, within $\pm 4 \mu\text{g m}^{-3}$ —for 20% of all hours (both downwind sites). The downwind measurements were significantly greater than those upwind during three-quarters of all hours.

TABLE 1a. Particle concentration measurements.

Instrument	Measurements	Sampling inlet height (m)	Measurement frequency
Met One Instruments BAM-1020 Beta Attenuation Monitor*	PM ₁₀ and PM _{2.5} mass concentrations	2.9/4.7**	Hourly

* Tandem instruments (for separate PM₁₀ and PM_{2.5} measurements) were deployed at each of three sites. A background site (site 1) was located south of the coal pile. Sites 2 and 3 were located north, in the prevailing downwind direction relative to the pile.

** Site 3 was in a wide shallow depression placing it 2–3 m below the elevation of the coal pile base, site 2, and the meteorological tower. The sampling inlet was raised to compensate for this difference. Inlets at the other sites were all 2.9 m high.

TABLE 1b. Meteorological measurements.

Instrument	Measurements	Measurement height (m)	Datalogger polling frequency (Hz)
R. M. Young model 81000RE sonic anemometers	3D wind components, speed, direction, turbulence (σ_θ and σ_w)	2.3 and 9.6	1*
Campbell Scientific CNR2 net radiometer	Net radiation, short- and longwave flux components	2.3	0.2
Vaisala HMI41 mechanically aspirated temperature and humidity sensors	Air temperature and relative humidity	2.3 and 9.6	0.2
Novalynx model 260–2501-A tipping-bucket rain gauge	Rainfall amount	0.5	**
Campbell Scientific CS616 water content reflectometer	Soil moisture content	~5 cm below the surface	0.2
Ecotech Aurora-1000G 525-nm nephelometer	Light scattering (b_{scat})	2.4	0.2

* Wind sensors have a 10-Hz data capture rate and data are averaged for output to the datalogger.

** Datalogger detects individual rain gauge bucket tips and stores the total number per sampling interval.

No human activity occurred near the pile for 35% and 37% of the observations at sites 3 and 2, respectively, across all PM₁₀ observations with favorable wind directions for detecting coal pile impacts. In addition, positive pile contributions to downwind PM₁₀ levels were measured during 50%–58% of hours experiencing no human activity and this high frequency was unexpected. We had assumed that the impact on excess downwind PM₁₀ levels would be minor because wind speeds were generally below the erosion threshold. Also, coal fires were thought to be primarily sources of PM_{2.5} and, thus, would not contribute substantially to PM₁₀ mass. As shown in Table 3, upwind–downwind PM_{2.5}

concentration differences were smaller relative to those for PM₁₀, implying a much smaller PM_{2.5} coal pile signal. Hours without human activity produced no significant PM_{2.5} differences about 60% of the time and showed a positive coal pile contribution only 23%–28% of the time. Clearly, PM_{2.5} levels associated with the pile are very small and represent a lower relative contribution for activity-free hours than found in the overall dataset. Coal combustion likely makes a minor but unquantifiable contribution to naturally occurring downwind PM₁₀ mass.

One-minute meteorological data provided a detailed characterization of the boundary layer moving across the pile. Data include wind speed, wind direction, the

TABLE 2. Wind data summarized for hours when airflow potentially transported coal dust from the pile toward one or both downwind monitoring sites.

10-m wind direction	Frequency (%)	Mean 10-m σ_θ (°)*	Mean 10-m speed (m s ⁻¹)	Max 10-m speed (m s ⁻¹)
SSE	5.9	32	2.2	6.2
S	7.9	30	2.6	7.0
SSW	4.4	25	2.4	5.9
SW	3.5	33	2.1	5.1
WSW	4.2	36	2.4	5.8

* Hourly standard deviation of wind direction.

TABLE 3. Total numbers of valid* upwind–downwind pairs of concentration data in each of three categories for all hours and those without human activity on or near the coal pile.

Downwind minus upwind, δ	Site 2		Site 3	
	All hours	No activity	All hours	No activity
PM ₁₀ data				
$<-4\ \mu\text{g m}^{-3}$	42	37	29	8
$-4 \leq \delta < 4\ \mu\text{g m}^{-3}$	151	105	160	111
$\geq 4\ \mu\text{g m}^{-3}$	581	142	625	165
Total	774	284	814	284
PM _{2.5} data				
$<-4\ \mu\text{g m}^{-3}$	82	53	74	36
$-4 \leq \delta < 4\ \mu\text{g m}^{-3}$	366	167	388	169
$\geq 4\ \mu\text{g m}^{-3}$	256	64	278	79
Total	704	284	740	284

* Both concentration measurements were nonmissing, hourly 10-m wind direction was between south-southeast and west-southwest, and no precipitation was measured.

standard deviation of the vertical wind component (σ_w), and the standard deviation of the horizontal wind direction (σ_θ). Other parameters were also measured or computed that had the potential to influence dust transport and downwind concentrations (see Table 4). Parameter $F(u)$ is the frequency with which the measured 1-min wind speed at level 2 (~ 10 m) exceeded the erosion threshold during an hour. This threshold, computed as the speed equivalent to the value of u_i^* previously described, was 5 m s^{-1} . For hours when u_i^* was exceeded, $F(u)$ averaged 0.14 with a maximum of 0.85. The nonlinear relationship between P_w and u^* [Eq. (1)] also suggests testing $\mu^2 = (u^* - u_i^*)^2$ as a potential predictor of downwind PM₁₀ levels.

Measured variations in 1-min wind direction indicated that airflow was seldom over the coal pile for all minutes of an hour. Therefore, from the 1-min data we computed the fraction of time f during each hour that air flowed over the pile and toward sites 2 or 3. The excess of particulate matter downwind [i.e., $C_{xs} = C_{dn} - C_{up}$, where C denotes the particulate concentration upwind (up) and downwind (dn)] was assumed to be due to pile emissions. For hours when this difference was $\leq -4\ \mu\text{g m}^{-3}$ we assumed that either a local source affected the upwind measurement or airflow variability was so large that a reliable excess concentration was indeterminate. In these cases, C_{xs} was rendered unfit for coal pile emissions analysis. An adjusted C_{xs} (C_{xs}^*) was computed to remove the effect of airflow that did not cross the pile:

$$C_{xs}^* = C_{xs}/f. \tag{4}$$

Average values of f for hours without human activity were 0.76 and 0.67 for sites 2 and 3, respectively. For cases when $f = 0$, C_{xs}^* was not computable and the hour was dropped from the analysis. This adjustment

provides for a more reliable comparison of different hours and produces a concentration value that is more easily analyzed for the dispersive influences on downwind concentrations. When C_{xs} was $0 \pm 4\ \mu\text{g m}^{-3}$ we assumed that measurement sensitivity limited our ability to confidently quantify excess concentration levels. Thus, we treated near-zero values differently depending on whether $-4 < C_{xs} < 0$ or $0 < C_{xs} < 4\ \mu\text{g m}^{-3}$ (i.e., arbitrarily setting all near-zero values to zero would introduce a bias—an excessive frequency of zero emissions—at the lower end of the C_{xs} distribution). The adjustment from Eq. (4) is meaningless for $C_{xs} < 0$ so in those cases $C_{xs}^* = 0$. However, C_{xs}^* was determined using Eq. (4) when $0 < C_{xs} < 4$ just as it was when $C_{xs} \geq 4$. Table 5 summarizes C_{xs} and C_{xs}^* for hours without coal pile activity. The maximum values listed for each site indicate a large range in excess concentrations compared to the mean, implying a long upper tail to the concentration distribution and results consistent with those previously reported for fly ash fugitive emissions (Mueller et al. 2013).

4. Results

Video images revealed that small clouds of dust occasionally formed over the pile with no human activity. This is consistent with measurements, in the absence of human activity, when downwind PM₁₀ concentrations were significantly above background. With most hours experiencing light winds it is likely that natural dust emissions occurred from the action of turbulent airflow. In one instance a dust devil was seen by researchers to form and move across the pile lofting copious amounts of coal dust into the air. Photographic evidence of dust devils was also collected during the study. During a dust-devil event captured on camera (see SM for a video of this event), the measured 10-m wind speed was $< 2\text{ m s}^{-1}$

TABLE 4. Parameters used to characterize conditions potentially affecting fugitive dust emissions from the coal pile and associated downwind particulate concentrations. All parameters are 1-h averages unless stated otherwise.

Parameter	Description	Level (m)
u	Horizontal mean wind speed	2 and 10
u^+	1-min max u	2 and 10
σ_u	Std dev of u	2 and 10
σ_u^+	1-min max σ_u	10
$F(u)$	Frequency that 1-min $u >$ wind erosion threshold during hour	10
μ^2	$(u^* - u^*)^2$	10
θ	Horizontal vector mean wind direction	2 and 10
θ_c	Angular deviation of θ from the direction of the pile center	2 and 10
σ_θ	Std dev of θ	2 and 10
σ_w	Std dev of vertical wind component	2 and 10
σ_w^+	1-min max σ_w	2 and 10
ε_{TK}	Turbulent kinetic energy	2 and 10
ε_{TK}^+	1-min max ε_{TK}	2 and 10
T	Air temperature	2 and 10
q	Relative humidity	2 and 10
ΔT	$T_{10} - T_2$	
R_{sol}	Solar radiation flux	2
R_{net}	Net radiation flux (solar and infrared)	2
b_{scat}	Light scattering at 556 nm	2
σ_{bscat}	Std dev of light scattering	2
b_{scat}^+	1-min max b_{scat}	2
P	Precipitation amount over various time intervals	Surface
M_s	Soil moisture content*	−0.05
x_f	Fetch of airflow across the pile	
x	Distance from the pile edge to measured downwind concentrations	

* Measured below the surface adjacent to the meteorological tower.

and was from the north. The dust-devil motion vector had an upwind component when compared with tower measurements. Such extreme turbulent behavior implies complex airflow over the pile superimposed on the mean ambient wind. The most probable cause of this localized airflow is solar heating of the sloping pile surface and intense surface heat flux. When mixed with ambient cross-flow near the top of the pile, localized wind shear can produce vortices that form extreme turbulence and occasionally dust devils as reported by Sinclair (1969).

a. Time of day variability

The nonlinear behavior of $PM_{10} C_{xs}^*$ is illustrated by mean values compared by hour of day. Generally, many meteorological parameters (e.g., wind speed, temperature, and relative humidity) vary by time of day similar to C_{xs}^* . Figure 2 plots the hourly variation in median C_{xs}^* for the two monitoring sites during hours without human activity on the pile and when the wind speed at 10 m was $< u_r$. The figure shows the tendency for PM_{10} concentrations at site 2 to be greater than values at site 3 (site 2 was nearer the coal pile). Note that there are no data for the hour ending at 1200 local standard time (LST) because of the noon meal break. Concentration values are typically lowest before 0800 LST and are highest during

0800–1100 LST, for the hour ending at 1300 LST, and—following a slight dip—plateau again through 2300 LST. We believe this pattern reflects the complex interplay of the underlying meteorological behavior: that is, 1) solar heating rate reaching its maximum around local noon, 2) coal surface temperature reaching a maximum and coal surface moisture content a minimum within about 2 h (i.e., by about 1400 LST), and 3) surface winds at a minimum overnight and increasing to a maximum during the afternoon due to vertical mixing. In addition, the highest levels of excess PM_{10} are consistent with the phenomenon of microscale convective vortices lofting dust clouds during daylight hours. Sinclair (1969) reported that dust devils in Arizona peak between 1300 and 1400 LST, which is the time of day closely associated

TABLE 5. Average measured and adjusted excess concentrations ($\mu g m^{-3}$) at downwind monitors during all hours with no human activity on the coal pile.

	Site 2		Site 3	
	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}
Concentration	Mean/max	Mean/max	Mean/max	Mean/max
C_{xs} (measured)	2.4/16	14.3/134	3.1/99	11.5/146
C_{xs}^* (adjusted)	3.5/32	19.7/235	5.9/99	21.1/442

FIG. 2. Site-specific time-of-day average values of $PM_{10} C_{xs}^*$ when winds were below the erosion threshold. Averages represent 3-h running means to smooth the data and reduce variability caused by differences in sample sizes from one hour to the next. Lines (solid = site 2; dashed = site 3) are polynomial fits to the data.

with the maximum soil surface temperature and surface heat flux and closely matches the peak in our observed natural dust concentrations.

Figure 2 also plots a 3-h running average of the median hourly C_{xs}^* . Polynomial fits (lines in Fig. 2) emphasize the primary features in the diurnal patterns and provide a smooth transition between hours. Both functions peak during midday (site 2: $48 \mu g m^{-3}$; site 3: $35 \mu g m^{-3}$) and are lowest at night. At site 2, the decline from midday through evening was more gradual than the rise from morning. Both sites also had a secondary peak of near $30 \mu g m^{-3}$ for the hour ending at 2200 LST.

Values of $PM_{10} C_{xs}^*$ when the wind exceeded the erosion threshold are plotted in Fig. 3 and indicate that the wind erosion potential was at a maximum during 1800–2300 LST. Data from both sites were combined in Fig. 3 because of the low number of observations. Typically, convection and thunderstorm activity reaches a maximum in late afternoon and early evening, leading to a greater frequency of gusty outflows reaching the surface and this could explain the evening peak in Fig. 3. At least one incidence of a late afternoon thunderstorm gust front generating large dust clouds over the pile was captured on camera.

When wind erosion was expected, the combined (sites 2 and 3) $PM_{10} C_{xs}^*$ averaged $8.5 \mu g m^{-3}$ during 0000–0500 LST, $35.2 \mu g m^{-3}$ during 1000–1600 LST, and $102.7 \mu g m^{-3}$ during 1600–2300 LST. Concentration differences between hours with and without wind erosion were not significant except for 1600–2300 LST when non-erosion events averaged $28.4 \mu g m^{-3}$, only 28% of that for the wind erosion hours. Diurnal patterns hint at other factors that may influence natural fugitive

FIG. 3. Average $PM_{10} C_{xs}^*$ by hour of day for hours without human activity on the coal pile and when the 10-m wind speed was above the wind erosion threshold. Data from sites 2 and 3 are combined.

emissions. However, time of day only accounts for 13% of the variance in C_{xs}^* .

b. Particle concentration dependencies

We computed hourly meteorological parameters (all but the last four listed in Table 4) using only those 1-min values that coincided with 10-m (i.e., level 2) wind directions aligned with the upwind location of the coal pile relative to the downwind monitoring sites. Thus, the mean hourly value $u_{10,n}$ (wind speed at 10 m) was computed for those minutes when the wind direction aligned with airflow over the pile toward site n .

The relationship of $PM_{10} C_{xs}^*$ with pile-aligned meteorological parameters was investigated using multivariate linear regression analysis with stepwise selection of the predictors. Downwind particle concentrations correlate positively with the rate of emission from the pile and negatively with parameters positively associated with diffusion (and secondarily deposition) of transported material. The general expression for concentration as a function of predictors ρ_i is

$$C_{xs}^* = a_0 + \sum_i a_i \rho_i, \quad (5)$$

where a_0 is a constant representing the value of C_{xs}^* when all predictors are zero and a_i is the set of regression coefficients for all predictors. The stepwise selection retained only predictors having significance at the 0.10 (p) level or better.

Coal moisture content M_c was derived from soil moisture content M_s measured in the ground near the coal pile. The soil measurement 5 cm below the surface represented bulk water in the topsoil layer. Hourly values of M_s varied only slightly throughout the day unless substantial precipitation occurred. Coal samples from the pile were analyzed in a laboratory to determine

FIG. 4. Comparison of sampled values of coal moisture content M_c with nearby measurements of soil moisture content M_s . Solid circles represent daytime M_c averages and M_s averaged daily for 1000–1600 LST. Vertical solid lines associated with each circle indicate ± 1 std dev of the individual data values on which the mean is based. Dashed lines represent linear fits between maximum (upper) and minimum (lower) daily M_c values vs averaged M_s . The solid line represents the linear regression of the average values, with the regression equation ($r^2 = 0.85$) included.

M_c . A plot of M_s (1000–1600 LST average for each sampling day) versus M_c (Fig. 4) shows that daily averages of the two parameters are well correlated ($r^2 = 0.85$) with M_s experiencing much larger variations than M_c . This relationship provides a means of estimating M_c using M_s as a surrogate.

The top part of Table 6 lists the results of this analysis that included 488 data points for hours when the 10-m wind speed was $< u_t$ as defined in AP-42. Wind speed (both mean and maximum measures) was not found to be a significant predictor. The eight parameters identified as being significant represented 25% of the total variance in $PM_{10} C_{xs}^*$.

The top four predictors, based on their partial r^2 contributions to C_{xs}^* , are surface (2 m) relative humidity (q_2), fetch across the pile (x_f), 2-m air temperature (T_2), and the standard deviation of wind speed at 10 m ($\sigma_{u,10}$). All of these were highly significant and together represented 20% of the overall C_{xs}^* variance. The negative association of C_{xs}^* with q_2 was not due to the moisture content of the coal surface because M_s was not identified as a significant predictor. Instead, q_2 is a surrogate for day–night differences in emissions due to the large diurnal variation in q_2 as previously described. A similar argument can be made for C_{xs}^* versus T_2 . Note that T_2 is not a surrogate for coal surface temperature T_s because T_2 was not measured directly above the coal pile and T_s responds in a more consistent manner to solar radiation flux than air temperature above the surface (Prigent et al. 2003). In addition, measured solar irradiance failed the regression significance test for a predictor.

The importance of x_f is not as a surrogate for the source surface area because x_f is negatively associated with concentration. As previously discussed, natural fugitive emissions occur at specific locations on the pile and not across the entirety of exposed coal surface. A negative association between C_{xs}^* and both x_f and x (distance from the pile edge to the measurement location) is probably due to the inverse effects of particle dispersion on distance from the point of emission.

Wind speed varies near the ground as boundary layer eddies intermittently transport momentum downward to the surface. Most turbulence metrics failed as predictors and this indicates that the tower measurements did not represent turbulence on the pile at the point where dust was generated. We postulate that the value of $\sigma_{u,10}$ as

TABLE 6. Results from a multivariate linear regression of $PM_{10} C_{xs}^*$ for hours without human activity on the coal pile.

Predictor* (ρ)	p level	Partial r^2	a	Interpretation of effect (E = emission and D = dispersion)
10-m wind speed $<$ erosion threshold (488 h, data from sites 2 and 3 combined)				
q_2	<0.0001	0.092	$-0.63 \mu\text{g m}^{-3} \%^{-1}$	Daytime E $>$ nighttime E
x_f	<0.0001	0.044	$-0.083 \mu\text{g m}^{-3} \text{m}^{-1}$	D increases with distance
T_2	<0.0001	0.034	$0.91 \mu\text{g m}^{-3} \text{K}^{-1}$	Daytime E $>$ nighttime E
σ_u	<0.0001	0.029	$69.7 \mu\text{g m}^{-3} (\text{m s}^{-1})^{-1}$	E increases with wind gustiness
ΔT	0.0007	0.021	$-11.2 \mu\text{g m}^{-3} \text{K}^{-1}$	E increases with surface heating
x	0.0014	0.019	$-0.068 \mu\text{g m}^{-3} \text{m}^{-1}$	D increases with distance
σ_{bscat}	0.049	<0.01	$0.64 \mu\text{g m}^{-3} \text{Mm}$	E increases with frequency of disturbances
ε_{TK}^+	0.098	<0.01	$-40.4 \mu\text{g m}^{-3} (\text{m}^2 \text{s}^{-2})^{-1}$	D increases with turbulence
10-m wind speed $\geq$ erosion threshold (64 h, data from sites 2 and 3 combined)				
μ^2	<0.0001	0.470	$5480 \mu\text{g m}^{-3} (\text{m s}^{-1})^{-2}$	E increases with P_w
M_s^{**}	0.0003	0.110	$-2.27 \mu\text{g m}^{-3} \%^{-1}$	E increases as coal surface dries
x_f	0.028	0.036	$-0.142 \mu\text{g m}^{-3} \text{m}^{-1}$	D increases with distance
σ_{bscat}	0.031	0.032	$4.75 \mu\text{g m}^{-3} \text{Mm}$	E increases with frequency of disturbances

* See Table 4 for an explanation of predictors. All predictors other than M_s are based on minutes when the source and downwind monitoring sites were aligned. All wind-related predictors are based on measurements only at the upper level.

** Coal moisture content was strongly associated with M_s based on data collected from the coal pile.

a predictor with positive association is in its representation of the periodicity of boundary layer eddies that acted on the pile surface to enhance dust emissions.

Concentration increased at the monitoring sites as the value of ΔT [near-ground vertical temperature difference ($T_{10} - T_2$)] became more negative. This is inconsistent with a dispersion effect because the association is in the wrong direction. Instead, ΔT is a likely surrogate for general surface heating, similar to but distinctly different from T_2 . Dust emissions show a preference for daytime conditions and ΔT is consistent with that behavior. The last two predictors (σ_{bscat} and $\varepsilon_{\text{TK}}^+$) are relatively minor and are not discussed in depth here. The low importance assigned to the 1-min maximum turbulent kinetic energy ($\varepsilon_{\text{TK}}^+$)—the only turbulence metric to show some significance as a predictor—highlights the difficulty we had using off-pile turbulence data to study dust emission rates. One reason for this is the high spatial variability in turbulence that likely occurs over the pile. Another reason is probably the fact that, while turbulence may be a factor positively influencing pile emissions, it also drives particle diffusion in a way that negatively impacts downwind concentrations. These emission and the diffusion effects oppose each other, as was noted by Cowherd et al. (1974), and the net effect is not easily discerned. Overall, the set of predictors accounted for only a small portion of the variance in downwind fugitive PM_{10} concentrations when winds were light.

A similar analysis was made of $\text{PM}_{10} C_{\text{xs}}^*$ for hours with wind erosion. As outlined in AP-42, guidance for coal piles, wind erosion is considered probable when maximum short-term (1–2-min average) wind speeds exceed roughly 5 m s^{-1} . This threshold is based on the lowest published u_t^* value (0.55 m s^{-1}) for crushed coal piles. Hour-average wind speed during the 64 h (day and night) when $u_{10,n} \geq u_t$ varied between 2.0 and 6.3 m s^{-1} . The predictors of $\text{PM}_{10} C_{\text{xs}}^*$ under these conditions (bottom part of Table 6) were μ^2 , M_s , σ_{bscat} , and x_f . Two of the predictors in the presence of wind erosion were also predictors without erosion. Wind speed as represented by μ^2 was a major factor controlling fugitive dust emissions when the erosion threshold was exceeded. Soil moisture content was also a strong predictor for the hours with more wind. This may be due to the fact that, as described by Laurent et al. (2009), moisture content in the coal surface creates stronger cohesive forces on small coal dust particles that are better able to resist erosion by wind action. The overall skill of the statistical model ($r^2 = 0.65$) when wind erosion was indicated was far greater than that for hours with low speeds.

Additional statistical modeling was performed on data for the individual monitoring sites to develop the means of estimating natural site-specific C_{xs}^* when

human activity occurred on the coal pile. Results from this work are described in SM.

c. Downwind levels of $\text{PM}_{2.5} C_{\text{xs}}^*$

Fine ($\text{PM}_{2.5}$) particle levels downwind of the coal pile were more difficult to study because of the lower levels that occurred. Downwind–upwind differences at both sites for low and high wind speed conditions are plotted in Fig. 5a, sorted into the same time-of-day categories used to examine $\text{PM}_{10} C_{\text{xs}}^*$ (Fig. 5b) in the previous section. “Invalid” denotes $\text{PM}_{2.5}$ differences that indicated upwind concentrations were significantly greater than downwind values, while “~0” denotes values that are $0 \pm 4 \mu\text{g m}^{-3}$. Most $\text{PM}_{2.5} C_{\text{xs}}^*$ values were near zero and the differences between frequency distributions for the two time-of-day datasets were small. Large day–night differences between $\text{PM}_{10} C_{\text{xs}}^*$ values (Fig. 5b) are not primarily attributable to $\text{PM}_{2.5}$ differences because daytime $\text{PM}_{2.5} C_{\text{xs}}^*$ averages $4.3 \mu\text{g m}^{-3}$ higher whereas daytime $\text{PM}_{10} C_{\text{xs}}^*$ averages $12.4 \mu\text{g m}^{-3}$ higher. This implies that unmeasured smoke plumes from coal fires—which contribute to the $\text{PM}_{2.5}$ mass fraction—were not the primary reason for the time-of-day differences in low-wind PM_{10} levels.

Given the large PM_{10} versus $\text{PM}_{2.5} C_{\text{xs}}^*$ differences, it is safe to assume that smoke did not contribute substantially to the considerable low-wind $\text{PM}_{10} C_{\text{xs}}^*$ levels measured before 0900 and after 1800 LST. To verify this conclusion we eliminated all hours when $\text{PM}_{2.5}$ and $\text{PM}_{10} C_{\text{xs}}^*$ were both $\leq 4 \mu\text{g m}^{-3}$ to focus only on periods when a valid downwind particulate excess was identified (i.e., excess concentrations were both above the minimum threshold for determining nonzero levels). We identified no significant association between $\text{PM}_{2.5}$ and PM_{10} in measurements from either site. The median $\text{PM}_{2.5}/\text{PM}_{10}$ ratios were 0.25 (site 2) and 0.37 (site 3). These ratios are far greater than the 0.10 reported by EPA (1995) for unpaved surfaces and the ratios found downwind of a dry fly ash storage facility (i.e., <0.10) where it was determined that ash disposal produced nonzero fugitive dust emissions in both the $\text{PM}_{2.5}$ and PM_c ($=\text{PM}_{10} - \text{PM}_{2.5}$) size ranges (Mueller et al. 2013).

With no statistical association between fine and coarse particle mass and given $\text{PM}_{2.5}/\text{PM}_{10}$ ratios that appear too large to represent purely fugitive sources, it appears that the presence of excess $\text{PM}_{2.5}$ mass at the downwind monitoring sites was generally not attributable to mechanically generated fugitive dust emissions from the pile. An alternate hypothesis for the few remaining unexpectedly high $\text{PM}_{2.5}$ values (i.e., $>5 \mu\text{g m}^{-3}$) measured during a few hours was smoke from coal dust combustion. Unfortunately, no coal fire database exists so this hypothesis is based on informal observations of

FIG. 5. Frequency distributions of (a) $PM_{2.5}$ and (b) PM_{10} C_{xs}^* for low speed (i.e., no wind erosion) hours sorted into two different time periods and for all wind erosion hours combined. In (a), “invalid” refers to hours with $PM_{2.5}$ $C_{xs}^* \leq -4 \mu g m^{-3}$ and ~ 0 refers to all values that are statistically 0, or $0 \pm 4 \mu g m^{-3}$. Data in (a) are generally sorted into $5 \mu g m^{-3}$ bins, whereas in (b) data are sorted into $10 \mu g m^{-3}$ bins.

small smoldering fires during infrequent site visits. However, one major fire was observed on camera, and the downwind $PM_{2.5}$ concentrations during that period were as high as $79 \mu g m^{-3}$. The $PM_{2.5}$ fraction of PM_{10} for this event was as high as 0.96. We showed earlier that relative to PM_{10} there were fewer hours for which downwind $PM_{2.5}$ levels were significantly greater than those upwind. This indicates that the signal of a $PM_{2.5}$ fugitive emission from the coal pile was usually very small. These factors made it impossible to further separate and quantify $PM_{2.5}$ fugitive dust emissions from natural mechanical processes and human activity.

5. Conclusions

Atmospheric processes frequently enhanced PM_{10} levels in air passing over a coal pile. Direct entrainment of particulate matter into the air by ambient wind was

clearly detected for the small fraction of hours when 1-min wind speed values exceeded the estimated erosion threshold. Wind erosion was not a function of the fetch across the pile, implying that it occurred at specific locations such as previously reported for the leading and lateral edges of the elevated portion of a pile. However, for most hours of this study the mean ambient wind was not sufficient to cause erosion of coal dust.

Observations identified dust devils forming and moving across the coal pile. This is evidence that the pile produces its own turbulent flows capable of raising dust from its surface even when ambient wind speeds are relatively low. In fact, the conditions listed by Sinclair (1969) as being favorable for dust-devil formation are all met over the coal pile (i.e., a surface of loose material, an area with anomalously high surface temperature, impediments to airflow capable of initiating localized vorticity formation, and a boundary between different types of terrain). The timing of measured downwind dust concentrations—with the maximum between 1100 and 1300 LST—matches closely the observed frequency of dust devils observed by Sinclair (1969) who associated dust-devil frequency with the time of day when maximum soil surface temperature and surface heat flux are most likely. Based on measurements summarized by Balme and Greeley (2006), maximum tangential wind speeds within dust devils typically range from 10 to over $20 m s^{-1}$, easily exceeding wind speed thresholds needed for entraining particles smaller than $10 \mu m$ from the surface. Thus, it is not surprising that a coal pile is an ideal location for natural dust removal and can significantly impact downwind air quality even when ambient winds are light.

Analysis during low ambient wind speeds of dust levels versus associated meteorological factors implicates the action of turbulent airflow as the primary source of excess PM_{10} mass at downwind measurement sites. We are unable to express generalities about how excess concentrations vary using a common set of specific predictors across all events. Time of day and wind erosion potential seem to be the best predictors of concentration at the two downwind sites. Excess PM_{10} levels are lowest at night when boundary layer turbulence is at a minimum, wind speeds are low, and there is no direct solar heating of the pile. These levels typically reach a maximum around midday and vary with light scattering and wind behavior measured downwind of the pile. Evidence is weak for the influence of coal surface moisture on dust erodibility in the absence of human activity, perhaps because of limited variation in its surrogate (soil moisture) during the hours examined.

Excess concentrations of $PM_{2.5}$ were not well correlated with PM_{10} mass and were usually not sufficiently high to identify nonzero levels with confidence. Excess

PM_{2.5} values were lowest at night. The difference between the low-speed (i.e., no wind erosion) midday hours ($8.7 \mu\text{g m}^{-3}$) and wind erosion hours ($4.9 \mu\text{g m}^{-3}$) was unexpected and cannot be explained. Of the processes that potentially contribute to an excess of particulate matter downwind of a coal pile, coal fires probably make an occasional contribution to PM_{2.5} levels but are unlikely to have much influence on airborne PM₁₀ mass. Overall, the data do not suggest a significant, measurable PM_{2.5} contribution to downwind particle levels from atmospheric processes, and this is consistent with previous studies that indicate most wind erosion occurs for particles in the 5–10- μm size range.

Ambient wind erosion alone cannot explain all natural particulate emissions from the coal pile because of the significant mean values of PM₁₀ C_{xs}^* across all wind speeds and hours. Localized airflow accelerations due to solar heating of the coal pile itself and vorticity formed in air flowing across the pile are the most plausible explanation for daytime hours but fail to explain nighttime values. Nighttime low-wind PM₁₀ values, though much lower, might be caused by unseen activity near the pile, could be due to a poor representation of background PM₁₀ concentrations, or they may be caused by an unidentified phenomenon. One potential explanation is that residual heat from the coal surface produces an unstable surface layer as cooler air from off the pile flows over the pile after dark. This instability could lead to localized turbulent flows occasionally lifting fine dust from the surface during brief periods. However, data from the study are insufficient for testing this hypothesis. In any event, nighttime excess dust is not associated with bulldozer action on the coal pile. Future use of these data to quantify emission factors (i.e., emission rates) from bulldozer activity must avoid including the unexplained particulate levels in emission factor calculations.

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