Journal of Exposure Science and Environmental Epidemiology (2014) 24, 405–411

www.nature.com/jes

ORIGINAL ARTICLE

Atmospheric particulate matter size distribution and concentration in West Virginia coal mining and non-mining areas

Laura M. Kurth[1], Michael McCawley[2], Michael Hendryx[3] and Stephanie Lusk[4]

People who live in Appalachian areas where coal mining is prominent have increased health problems compared with people in non-mining areas of Appalachia. Coal mines and related mining activities result in the production of atmospheric particulate matter (PM) that is associated with human health effects. There is a gap in research regarding particle size concentration and distribution to determine respiratory dose around coal mining and non-mining areas. Mass- and number-based size distributions were determined with an Aerodynamic Particle Size and Scanning Mobility Particle Sizer to calculate lung deposition around mining and non-mining areas of West Virginia. Particle number concentrations and deposited lung dose were significantly greater around mining areas compared with non-mining areas, demonstrating elevated risks to humans. The greater dose was correlated with elevated disease rates in the West Virginia mining areas. Number concentrations in the mining areas were comparable to a previously documented urban area where number concentration was associated with respiratory and cardiovascular disease.

Journal of Exposure Science and Environmental Epidemiology (2014) **24,** 405–411; doi:10.1038/jes.2014.2; published online 19 February 2014

Keywords: epidemiology; particulate matter; personal exposure

INTRODUCTION

Atmospheric particulate matter (PM) originates directly from natural sources (i.e., pollen, dust, forest fires) and anthropogenic sources (i.e., motor vehicles, factories, agriculture) and indirectly from chemical reactions converting atmospheric precursors into secondary PM.[1] Exposure to increased PM is associated with excess respiratory and cardiovascular hospital admissions, morbidity, and mortality.[2–4] Current US Environmental Protection Agency (EPA) standards monitor PM with partially size-classified, mass-based standards of PM_{10} (particulates with an aerodynamic diameter of $\leq 10\,\mu m$) and $PM_{2.5}$ (particulates with an aerodynamic diameter of $\leq 2.5\,\mu m$).[5]

PM_{10} and $PM_{2.5}$ provide few details on the complete size distribution, number concentration, and health risks of PM sampled. Mounting evidence suggests a need to account for ultrafine particles (UFPs) when evaluating health risks of PM.[6] UFPs (particles with a diameter of $\leq 0.1\,\mu m$) are more toxic to humans than larger PM ($> 5\,\mu m$) because they are more efficient at penetrating deep into the alveolar region.[6] UFPs have a greater surface area per unit mass, larger number concentration, and greater alveolar deposition efficiency compared with larger particles, resulting in greater inflammation capabilities.[6–9] Delivery of equal masses of UFPs and larger PM results in increased inflammatory effects and bronchoalveolar inflammation from exposure to UFPs compared with the larger PM.[6] UFPs are nearly indistinguishable by PM_{10} and $PM_{2.5}$ mass measures because UFPs contribute only a small fraction of mass to PM_{10} and $PM_{2.5}$ concentrations but contribute a large fraction to number concentration.[6,10] Health risk is more effectively measured with complete particle size distributions that account for UFPs to determine deposited respiratory dose.[6,11] *Deposition* or *dose* is the proportion of inhaled particles that deposit or adhere to the lung surface.[10] Deposited particles generate a physiological response, including inflammation and oxidative stress. Deposition is dependent upon respiratory tract and breathing characteristics, as well as particle properties.[10,12] This study utilized mass-based and number-based particle size distributions to calculate deposited lung dose during incremental periods at two sampling areas in West Virginia (WV) with relatively higher or lower disease rates.

Coal mining, including mountaintop coal mining (MTM), was prominent around the higher disease rate area and no mining occurred in the lower disease rate area. The MTM method uses explosives and excavation equipment to remove vegetation, rock, and dirt from mountaintops to expose coal seams.[13] Residents of coal mining areas have significantly higher mortality from chronic heart, respiratory, and kidney diseases, and elevated morbidity from chronic cardiopulmonary, cardiovascular, and kidney diseases.[14–16] Age-adjusted total mortality rates in MTM areas are significantly greater compared with non-mining areas in central Appalachian states.[17] Residents of MTM areas have an increased prevalence of congenital anomaly births compared with residents of other Appalachian coal mining areas.[18] In addition, age-adjusted chronic cardiovascular rates were greater in MTM Appalachian counties compared with non-mining Appalachian counties.[19] It is possible that mining activities contribute to local air pollution, but assessments of number concentrations and deposited lung dose have not been undertaken in mining communities.

MATERIALS AND METHODS

Sampling

Ambient PM sampling was conducted from June 2011 to May 2012 at three rural residential areas in WV. The two MTM sampling sites, Twilight

[1]Department of Epidemiology, School of Public Health, West Virginia University, Morgantown, West Virginia, USA; [2]Department of Occupational and Environmental Health, School of Public Health, West Virginia University, Morgantown, West Virginia, USA; [3]Department of Health Policy, School of Public Health, West Virginia University, Morgantown, West Virginia, USA and [4]School of Public Health, West Virginia University, Morgantown, West Virginia, USA. Correspondence to: Dr. Laura M. Kurth, Department of Epidemiology, School of Public Health, West Virginia University, Morgantown, West Virginia 26505, USA.
Tel.: +1 30 4293 8059. Fax: +1 30 4293 3755.
E-mail: lesch@mix.wvu.edu
Received 29 July 2013; accepted 17 December 2013; published online 19 February 2014

Table 1. Summary of sampling site characteristics.

Nearest town	Latitude	Longitude	Elevation (m)	Distance to state road[a] (m)
Edwight	37°53′0.4″ N	81°32′2.5″ W	303	160
Twilight	37°55′31″ N	81°37′25″ W	333	143
Green Bank	38°25′27″ N	79°49′43″ N	821	125

[a]Distance from the site to the nearest main road.

Figure 1. The locations of sampling sites in West Virginia.

and Edwight, were located in valleys surrounded by mountains in two adjacent counties (Raleigh County and Boone County) where active MTM and other coal mining activities (rail and truck transportation, underground mines, and coal processing facilities) were prominent. A comparison site in eastern WV (Green Bank, Pocahontas County) did not have any mining activity. Figure 1 illustrates the locations of the sampling sites.

In June 2011, brief sampling was conducted at additional sites dispersed throughout each county (three sites per county) to select one sampling site that was representative of each area. The sites were selected based on representativeness of mining activity and particle parameters in the area as well as site accessibility.[20] The three selected sites are summarized in Table 1.

A sample of residents surveyed from the areas around the MTM and non-mining sites indicated that a similar proportion of residents were female, current or former smokers, and smokeless tobacco users in the two areas.[21] Previous research on the health status of residents living around the MTM sites indicated increased reports of poor health outcomes, including significantly higher reports of cancer, compared with residents around the non-mining site.[21] Residents around the MTM areas were significantly more likely to have an occupational history as a coal miner and to have resided in the area for a longer period ($M = 45.1$ years, $SD = 23.0$) as compared with those surveyed around the non-mining site ($M = 39.8$ years, $SD = 24.3$).[21]

Edwight, Raleigh County. Edwight is an unincorporated area ~12.9 km south of Whitesville, WV, and 48.3 km northeast of the county seat, Beckley, WV. Raleigh County ranks among the lowest 25% of all 55 WV counties by overall county health ranking. County health ranking was determined based on the mortality rate of those ≤75 years of age, self-reported fair or poor health, self-reported physical and mental health, and rate of low-birth-weight infants.[22] In 2011, Raleigh County had 13 surface mines (5.7% of total state surface mines) and 23 underground mines (8% of total state underground mines) and contributed 6.5% to the state's total coal production.[23]

The Edwight sampling site was located in a resident's yard along the Coal River ~160 m off state road WV 3. The site was surrounded by MTM activity, including an expansive MTM site to the northwest and a smaller MTM site ~4.8 km northeast.

Twilight, Boone County. Twilight is an unincorporated area with a population of 90 people.[24] Twilight is located ~32.2 km from the county seat of Madison, WV (population of 3000).[24] Boone County ranks in the lowest 10% of all WV counties and has a higher than state average premature death rate.[22] In 2011, Boone County had 31 surface mines (13.5% of total state surface mines) and 68 underground mines (24.3% of total state underground mines) and, as a county, contributed the greatest percent (15%) to the state's total coal production.[23]

The Twilight sampling site was located on a dead end road ~143 m off county highway 26 in a Twilight resident's yard. The Twilight site was at a slight elevation from the main road. An expansive MTM site stretched from the southwest to the southeast of the Twilight valley.

Green Bank, Pocahontas County. Green Bank (population 143) is in a county where ~60% of the land is federal or state owned and no coal mining is present.[24,25] Pocahontas County ranks in the lowest 40% of WV counties by health outcome; however, the health rankings of the surrounding counties are high.[22] The Green Bank site was in the yard of a rural library, ~125 m off state road 28/92. Visits to this site were predominately when the library was closed or at times when there was minimal, if any, vehicle traffic in the library parking lot. This site was unique from the MTM sites because it was located in an open area rather than in a contained valley with the topography reflecting increased wind speeds and air movement patterns.

Instrumentation

Total suspended particle (TSP) mass concentration was measured gravimetrically with a polytetrafluoroethylene (PTFE)-coated glass fiber filter (37 mm diameter 5.0 μm pore- size, SKC) connected to a low-volume vacuum pump (Model 1531-1078-G288x, Gast Manufacturing, Benton Harbor, MI, USA). The vacuum pump was permanently placed at each site to collect TSP mass concentration for ~3 to 6 weeks (average time of 29 days) before exchanging the used filter for a clean filter.

A PTFE filter was placed in a two-piece plastic filter cassette on top of a support pad to prevent filter fibers from rupturing. Cassette pieces were sealed together with tape to eliminate particles from leaking into the cassette or bypassing the filter. The filter cassettes were ~0.5 m above the ground surface and the vacuum pump was placed in a covered water-resistant structure. Each filter was equilibrated for 24 h at constant relative temperature and humidity before pre-weighing. TSP mass concentration was calculated gravimetrically from the change in pre- and postexperiment filter weights, volume of air passing through the filter, and sampling time. The difference between the pre- and post-experiment filter weight was divided by the average volume of air passing through the filter. The volume was calculated by averaging the sampling period start and end pump flow rates (overall average flow rate of 17.4 l/min) during the sampling period. Field blanks were prepared to check for the precision of the weighing method, resulting in an average error of 0.00075 mg.

Particle size distributions were determined with two near-real-time instruments to obtain size distributions for two particle size ranges. The Aerodynamic Particle Sizer (APS) (Model 3321, TSI, Shoreview, MN, USA) measured size-classified, mass concentrations for 52 size categories based on the aerodynamic diameter of particles 0.5–20 μm. The Scanning Mobility Particle Sizer (SMPS) included both an Electrostatic Classifier (Model 3080, DMA, Model 3081, TSI) and Condensation Particle Counter (Model 3788, TSI) to size particles 0.01–0.4 μm based on thermodynamic equivalent diameter into 105 size categories by particle number. The Electrostatic Classifier sized and separated particles based on their electrical mobility before they were counted in the Condensation Particle Counter. The SMPS calculated total number concentration for particles 0.01–0.4 μm by averaging the number-weighted concentration per channel for all size categories. The SMPS was equipped with an X-ray aerosol neutralizer as opposed to krypton neutralizer, making it suitable for fieldwork.

The APS and SMPS were transported to each site every 3 to 6 weeks. At each site visit, the APS recorded mass-based size distributions for two, 10-min sampling sessions, and the SMPS recorded number-based size distributions for 3–8 2-min and 15 s sessions. The instruments sampled from a height of ~0.5–0.8 m above the ground surface. Currently, the EPA does not have a State and Local Air Monitoring Station set up in Pocahontas County to conduct PM_{10} and $PM_{2.5}$ ambient air monitoring.[26] A $PM_{2.5}$ monitor was operating in Raleigh County but was located in the county's major population center and distant from MTM activity. Therefore, we obtained estimates of PM_{10} and $PM_{2.5}$ at each site visit using gravimetric measurements and APS size distributions. The proportion of the total mass in the PM_{10} or $PM_{2.5}$ APS size distribution was applied to the

TSP mass concentration sample to estimate what we denote as PM_{10} or $PM_{2.5}$ from the TSP mass concentration.

Respiratory Deposition Calculations

Mass-based and number-based size distributions were used to estimate total deposited respiratory dose to adult humans. Respiratory deposition was calculated with a mathematical model based on the International Commission on Radiation Protection standards and formulas for healthy adult males from Stahlhofen et al.[27] to determine regional deposition efficiency values in four areas of the respiratory tract (nose, extrathoracic region, tracheobronchial region, and alveolar region). Inertial impaction and gravitational sedimentation were the primary deposition methods in the nasal and extrathoracic regions. Brownian diffusion was the primary mechanism of deposition in the alveolar and tracheobronchial regions.

The arithmetic mass-based particle size distributions from the APS and number-based particle size distributions from the SMPS were converted to lognormal distributions for empirical purposes. The product of the fraction of PM in a specific particle size range and the respiratory deposition fraction for that size range determined the percent deposition for the nose, extrathoracic, tracheobronchial, and alveolar regions.[27] Deposition in a region is affected by deposition in the preceding regions, and total deposition is deduced by accounting for deposition in preceding regions. After determining respiratory deposition for all size ranges, the regional geometric means and SDs were integrated to estimate total lung deposition for 50% oral and 50% nasal breathing with the parameters: volumetric flow rate $=116.7\,cm^3/s$, volume $=500\,cm^3$, functional residual lung capacity $=3{,}300\,cm^3$, and breaths per min $=7$. The deposited fraction measurement converts a multimodal size distribution with multiple means and SDs (because of a polydisperse sample) to a single number that has utility for comparing size distributions across sites.

The following are the basic deposition fractions simplified from Stahlhofen et al.[27] by Hinds[1]. Here, d_p is the particle diameter, *IF* is the inhalable fraction, DF_H is the deposition fraction for the head airways, DF_{TB} is the deposition fraction for the tracheobronchial region, DF_A is the deposition fraction for the alveolar region, and the total deposition, *DF*, is the sum of all region depositions.

$$IF=1-0.5\left(1-\frac{1}{1+0.00076\,dp^{2.3}}\right) \quad (1)$$

$$DF_H = IF\left(\frac{1}{1+\exp\left(6.84+1.183\ln d_p\right)}+\frac{1}{1+\exp\left(0.924-1.885\ln d_p\right)}\right) \quad (2)$$

$$DF_{TB}=\left(\frac{0.00352}{d_p}\right)\left[\exp\left(-0.234\left(\ln dp+3.40\right)^2\right)+63.9\exp\left(-0.819\left(\ln dp-1.61\right)^2\right)\right] \quad (3)$$

$$DF_A=\left(\frac{0.0155}{d_p}\right)\left[\exp\left(-0.416\left(\ln d_p+2.84\right)^2+19.11\exp\left(-0.482\left(\ln d_p-1.362\right)^2\right)\right)\right] \quad (4)$$

$$DF=IF\left(0.0587+\frac{0.911}{1+\exp\left(4.77+1.485\ln dp\right)}+\frac{0.943}{1+\exp\left(0.508+2.58\ln dp\right)}\right) \quad (5)$$

Data Analysis

Wilcoxon signed-rank tests were carried out between MTM and non-mining particle parameters. This method of analysis was selected because of small sample sizes and non-normal distributions of the particle variables. Median values were compared between the MTM and non-mining sites by season and month. Seasonal labels were assigned to groups of months and were not specific to the actual astronomical season dates. Seasonal labels included: summer (June, July, August), fall (September, October, November), winter (December, January, February), and spring (March, April, May).

Multiple particle size distributions were obtained at each site visit, so respiratory deposition was calculated by obtaining a dose for each size distribution, then finding the arithmetic mean of all site doses. Results were consistent with respiratory deposition measures calculated by fitting a deposition curve to the site's overall average size distribution.

RESULTS

Meteorology

Meteorological conditions influence particle concentration and were reported for each sampling area (Table 2). Greater average wind speeds were recorded around the non-mining Green Bank site compared with the MTM sites in Edwight and Twilight. Light winds were present at the MTM sites during July, November, and April sampling dates, but periodic wind gusts were more characteristic of the MTM sites. Temperature fluctuated with the season and was generally lower at Green Bank, where elevation was higher compared with the MTM sites. The observed relative humidity fell within the normal monthly range for the county, except on the January and February 2012 sampling dates at Edwight and Twilight when the humidity was above the normal monthly range.[28] Humidity was above the normal monthly range at Green Bank on the March and May sampling dates, but lower in April.[28]

Seasonal Particle Concentrations

Sampling sites were divided into two subgroups with the Edwight and Twilight sites grouped together as MTM. The Green Bank site was the non-mining site. Figure 2 shows the median, interquartile range, maximum, and minimum TSP, $PM_{10,}$ and $PM_{2.5}$ mass concentrations across seasons at the MTM and non-mining sites. A Wilcoxon signed-rank test indicated no significant effect of mining activity for TSP and $PM_{2.5}$ mass concentrations when compared for all four seasons. A Wilcoxon signed-rank test showed a significant effect of mining activity for PM_{10} mass concentration during June ($n=12$, $Z=2.8$, $P=0.0172$) and July ($n=20$, $Z=4.0$, $P=0.0008$) (monthly results not shown) but the effect of mining activity for PM_{10} mass concentration was not significant during the combined summer season ($Z=-1.7$, $P=0.1014$). $PM_{2.5}$ mass concentration was significantly greater at the MTM site ($Mdn=1.8$) than at the non-MTM site ($Mdn=0.54$) during July ($n=20$, $Z=4.0$, $P=0.0008$).

The sized-mass distributions for particles 0.5–20 μm provided by the APS were used to estimate respiratory deposition, and the deposition percent was applied to the TSP mass concentrations to obtain deposited mass concentrations. Wilcoxon signed-rank tests (not shown) indicated significant effect of mining activity for deposition percent ($n=39$, $Z=-4.3$, $P=0.0001$) and deposited mass concentration ($n=39$, $Z=-2.5$, $P=0.0158$) during the summer. Monthly comparisons (not shown) indicated a significant effect of mining activity for deposition percent during July ($n=15$, $Z=-3.0$, $P=0.0095$) and for deposited mass concentration for June ($n=12$, $Z=2.8$, $P=0.0172$) and July ($n=15$, $Z=4.0$, $P=0.0095$) for particles 0.5–20 μm.

Figure 3 shows the median, interquartile range, maximum, and minimum respiratory deposition percent, number concentration, and deposited respiratory number concentration at the MTM and non-mining sites by season for particles 0.01–0.4 μm. There was a significant effect of mining activity for number concentration ($P=0.0015$) and deposited respiratory number concentration ($P=0.0015$) in winter and when collapsing all measurements across seasons. At the mining and non-mining sites, deposition percent was lowest in the summer. Deposition percent was significantly greater at the MTM site compared with the non-mining site during summer ($P=0.0018$) and fall ($P=0.0325$), representing a greater deposition percent at the MTM areas in August ($n=19$, $Z=-2.6$, $P=0.0171$) and October ($n=10$, $Z=-2.5$, $P=0.0105$) (not shown). The remaining months and seasons had similar deposition percents, indicating similar particle distributions. The median particle count was highest at the mining sites during the winter season and highest at the non-mining site in summer. Deposited number was elevated in the winter and spring seasons.

Table 2. Daily meteorological conditions for each site visit.

Site	*Date*	*Wind direction*	*Average wind speed, km (max)*	*Average temperature °C*	*Relative humidity (%)*
Edwight	6/6/11	NW	0 (4)	26	77
Edwight	6/7/11	NNW	0 (12)	23	77
Twilight	6/8/11	NNW	0 (6)	26	80
Twilight	6/9/11	NW	0 (7)	27	82
Green Bank	6/10/11	NWN	7 (20)	21	75
Green Bank	6/11/11	W	6 (13)	21	79
Edwight, Twilight	7/11/11	NNW	1 (7)	28	71
Green Bank	7/11/11	SW	9 (22)	24	80
Edwight, Twilight	8/5/11	SE	0 (6)	26	80
Green Bank	8/5/11	SSE	13 (17)	21	93
Edwight, Twilight	9/12/11	N	0 (6)	19	86
Green Bank	9/11/11	W	6 (14)	17	71
Edwight, Twilight	10/8/11	ESE	0 (5)	16	80
Green Bank	10/8/11	ESE	5 (8)	13	81
Edwight, Twilight	10/26/11	NW	0 (7)	13	77
Green Bank	10/25/11	WNW	12 (21)	10	65
Edwight, Twilight	11/14/11	SW	1 (9)	17	65
Green Bank	11/14/11	WSW	16 (26)	13	68
Edwight, Twilight	12/4/11	SSE	0 (5)	8	73
Green Bank	12/4/11	S	9 (15)	40	72
Edwight, Twilight	1/16/12	SW	0 (5)	3	82
Green Bank	1/16/12	SSW	9 (18)	−3	72
Edwight, Twilight	2/19/12	SE	0 (5)	3	87
Green Bank	3/2/12	SSE	11 (16)	3	100
Edwight	4/3/12	NNW	1 (7)	16	68
Green Bank	4/3/12	W	7 (12)	14	30
Edwight	5/7/12	N	–	22	79
Green Bank	5/6/12	SE	4 (12)	17	97

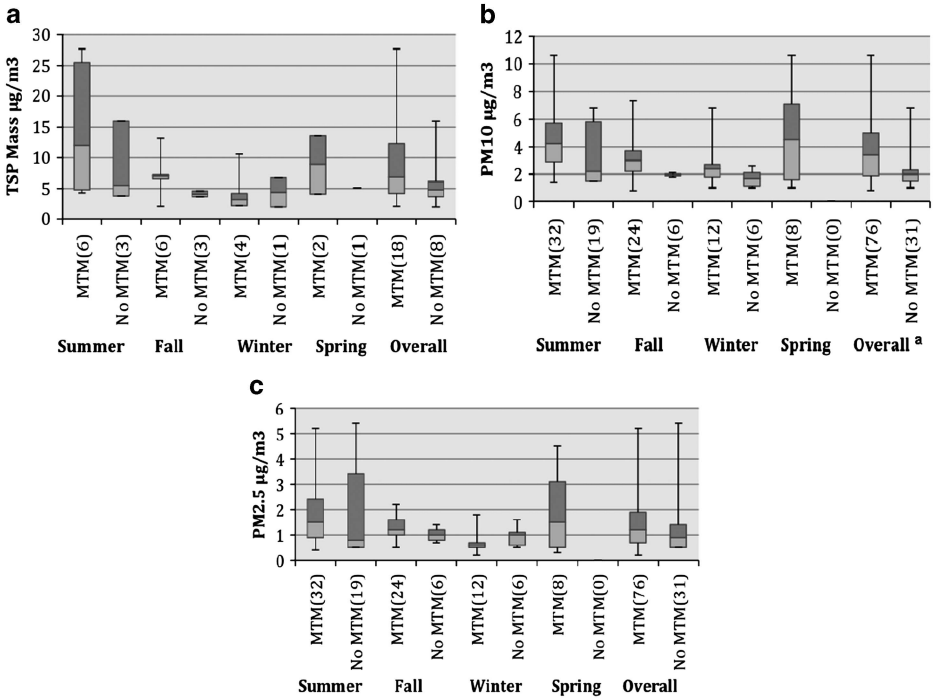

Figure 2. Comparison of MTM and non-mining sites by season (*N*) for (**a**) TSP, (**b**) PM_{10}, and (**c**) $PM_{2.5}$ mass. [a]Significantly different mass between MTM and non-mining sites based on a Wilcoxon signed-rank test (two-tailed *t*-approximation *P*-value <0.05).

DISCUSSION

This study applied repeated sampling to examine seasonal behavior of mass concentration, number concentration, and deposited lung dose at limited sampling sites around MTM and non-mining areas. Despite a statistically significant difference in combined PM_{10} mass concentration between the MTM and non-mining areas (Figure 2), PM_{10} concentrations were substantially less than the EPA limit of 150 $\mu g/m^3$. A higher PM_{10} mass indicated a greater contribution from localized coarse particles that is not toxicologically likely to account for the health outcome differences

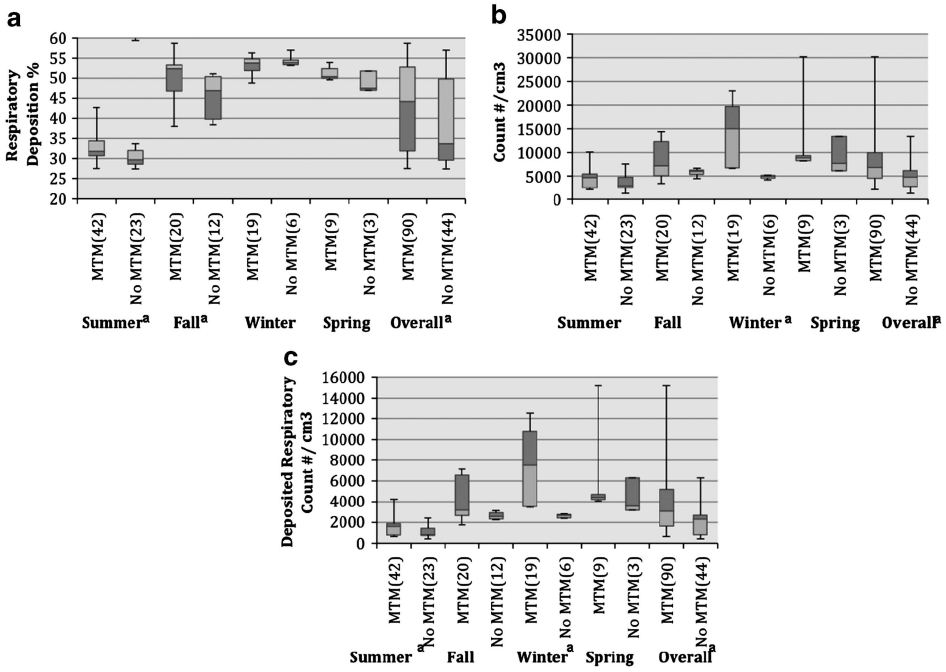

Figure 3. Comparison of MTM and non-mining sites by season (*N*) for (**a**) respiratory deposition percent, (**b**) count, and (**c**) deposited respiratory count for particles 0.01–0.4 μm. [a]Significantly different mass between MTM and non-mining sites based on a Wilcoxon signed-rank test (two-tailed *t*-approximation *P*-value <0.05).

Table 3. Summary of significant (+) and non-significant (−) results between MTM and non-mining sites by season.

Parameter	*Summer 2011*	*Fall 2011*	*Winter 2011–2012*	*Spring 2012*	*Overall*
TSP mass μg/m^3	−	−	−	−	−
PM_{10} μg/m^3	−	−	−	−	+
$PM_{2.5}$ μg/m^3	−	−	−	−	−
Respiratory deposition % 0.5–20 μm	+	−	−	−	+
Deposited respiratory mass 0.5–20 μm	+	−	−	−	+
Respiratory deposition % 0.01–0.4 μm	+	+	−	−	+
Count 0.01–0.4 μm	−	−	+	+	+
Deposited respiratory count 0.01–0.4 μm	−	−	+	+	+

between the two areas. The chemical constituents of TSPs and elevated numbers of UFPs are more plausible explanations for the difference in health outcomes between the two areas. The clinical significance of the small PM_{10} differential (~ 1.4 μg/m^3) between the two areas overall is, therefore, questionable.

Respiratory deposition percent values were similar across the seasons; however, lower summertime deposition percents suggested fewer particles in the smaller size ranges compared with other seasons (Table 3). Particle growth by absorption and condensation or elevated concentrations of larger, aged PM during the summer months may contribute to this seasonal difference. In addition, a PM source may have been absent or at an increased distance from the sampling sites during the summer, resulting in a decreased contribution of smaller PM. The respiratory deposition values account for particle size and the relatively comparable values for the remainder of the year indicate consistent PM sources at both sites. However, the number concentrations around the MTM area were significantly higher compared with the non-mining site, alluding to an increased number of particles from MTM area sources.

Particle size distributions had multiple modes indicating multiple particle sources; however, the equipment used in this study did not directly identify PM source. Size distributions consistently specified a mode in the distribution around 0.02–0.03 μm, suggesting the presence of secondary PM manufactured through photochemical reactions converting oxides of nitrogen and sulphur into particulates. These secondary PMs may be the result of coal-fired power plant emissions that travel across state boundaries from plants in Cincinnati, OH, Columbus, OH, Lexington, KY, and the Ohio River Valley area located northwest and south of sampling areas. Coal-fired power plant emissions cause health problems among populations living near and at a distance from plants.[29,30] A peak in the size distributions around 0.05–0.07 μm indicated coagulated soot and accumulation mode particles.[31] Larger, coarse particles were also prominent in the size distributions and were likely mechanically generated or secondary long-range transported particles. At MTM sites, coarse particles were speculated to be primarily coal dust or crustal dust generated during overburden removal.[32]

Number concentrations taken with similar sizing equipment as that used in the present study were not available for nearby regions. Number concentrations at the mining and non-mining areas were found in higher concentrations in comparison with number concentrations from Copenhagen, Denmark, where similar sizing equipment was used.[11] Anderson et al.[11] reported an average number concentration of 8116 particles/cm^3 in Copenhagen, Denmark, from May 2001 to December 2004 for particles from 29 size channels between 0.006 and 0.7 μm accounting for the width of the size channels. The number concentrations in Copenhagen were associated with hospital admissions for cardiovascular disease and respiratory conditions in

the elderly and children. Number concentrations in the present study were approximately equal or greater than the average in Copenhagen, suggesting if health reactions at the WV sites were similar to those in Copenhagen, pediatric asthma hospital admissions and cardiovascular and respiratory disease hospital admissions in the elderly should also be a problem in the WV sampling areas.[11] In addition, health effects are associated with elevated number concentrations in the mining areas as ambient PM collected from WV MTM areas altered normal microvascular function after being intratracheally instilled in rats.[33]

Mining activity is a possible explanation for the greater number concentrations in the vicinity of the MTM area sites and seasonal differences in particle parameters. During mining processes, particulate matter is generated from explosive blasting, dragline operations, feeder breakers that crush coal, and wind erosion of coal storage piles.[34] Fossil fuel combustion sources, including motor vehicles, diesel engines, and heavy equipment around MTM sampling sites, are major sources of organic compounds and secondary PM. Unidentified sources of PM may contribute to a substantially elevated background particle number concentration, but local sources around MTM areas seem to exceed that contribution and account for significant differences between the MTM and non-mining areas.

The elevated number concentrations relative to Copenhagen and the relatively higher mining area number concentrations may be explained by meteorological conditions affecting the transport and dispersion of PM in the atmosphere. Atmospheric processing changes PM size, shape, composition, and weight through coagulation, absorption, and evaporation.[32] Increased airflow, as was present at the non-mining, Green Bank area, aided particle transport to the area, but also improved dispersion of particles in the atmosphere. The valleys in southern WV mining areas trap the air and shield circulation patterns. Warmer air flowing up the valley during the daytime reverses direction at night, trapping contaminants in the valley. Particle number may also be increased from inversions of warm air over cooler air, suppressing vertical mixing of PM at MTM areas, but inversion occurrences were not recorded in this study.

Limitations of this study include shortcomings of the sampling equipment. Low-volume pump flow calibration measures were only obtained at pump start and stop times because of limited equipment resources as well as the rural locations of sampling sites. The difference between low-volume start and stop flows indicated flow decreased during sampling periods as a result of diminishing filter collection efficiency and filter overload. In some instances (September 2011 and May 2012), the low-volume pumps stopped altogether and no mass concentration was recorded because the pump shut-off time and stop flow rate were unknown. In addition, the filters became overloaded and encrusted with impacted particles, making them less efficient at collecting PM. Therefore, the TSP mass concentration measure was an underestimate and seemed to be a better estimate of PM_{10} because of the entry efficiency of the 35 mm filter for 25 μm particles. Still, the filter cassettes were used because they were a more practical and feasible method for obtaining TSP mass concentrations at multiple sites in the field compared with other sampling methods.

Measurements were conducted with the same instrumentation to obtain comparable data between sites. The findings from this study are based on measurements from three sites with a small number of brief observations for each month and season, limiting statistical power and generalizability to other areas. The deposition fractions were calculated for healthy adults under idealized laboratory conditions and are not representative of all individuals in the mining and non-mining areas of interest. Further examination of the magnitude of PM concentrations must be conducted with additional sites, specifically MTM sites. Although data were collected from an area with significantly more documented health problems where MTM was occurring, particles were not necessarily from a MTM source, and no biological assessments were made of PM exposure in humans.

In conclusion, significant differences in the number concentration and deposited number concentration between the MTM and non-mining areas are correlated with previously documented differences in population health outcomes including mortality, cardiovascular disease, birth defects, and cancer.[17–19,21] The levels of particle number concentrations around the MTM sites were larger than those associated with increased paediatric asthma hospital admissions and respiratory and cardiovascular disease hospital admissions in the elderly in a major urban area.[11] PM concentration varied over the sampling period, but PM sources were relatively constant. Statistically significant differences between the two areas for PM_{10} were present for a small number of sampling periods, but the clinical and toxicological significance of this small differential is unlikely. This study indicated that measuring the deposited fraction of dose is now technically feasible for mass-based and number-based size distributions. Accounting for the fraction of particulate that deposits in the lung, thus directly contributing to respiratory dose, is a more sensitive indicator of epidemiological disease patterns than mass measures such as PM_{10} and $PM_{2.5}$.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

ACKNOWLEDGEMENTS

We acknowledge Bean Chen (National Institute of Occupational Safety and Health, Morgantown, WV, USA) Matthew Gurka (School of Public Health, West Virginia University), and Timothy Nurkiewicz (School of Medicine, West Virginia University) for their assistance and support.

REFERENCES

1 Hinds WC. *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*. John Wiley & Sons: New York, NY, 1999.
2 Dockery DW. Epidemiologic evidence of cardiovascular effects of particulate air-pollution. *Environ Health Perspect* 2001; **109**: 483–486.
3 Dominici F, Peng RD, Bell ML, Pham L, McDermott A, Zeger SL *et al.* Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. *JAMA* 2006; **295**: 1127–1134.
4 Pope CA, Burnett RT, Thun MJ, Calle EE, Krewski D, Ito K *et al.* Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *JAMA* 2002; **287**: 1132–1141.
5 US EPA. Particulate Matter (PM). US EPA Air & Radiation 2013. Retrieved from www.epa.gov/pm/index.html.
6 Donaldson K, Stone V, Clouter A, Renwick L, MacNee W. Ultrafine particles. *Occup Environ Med* 2001; **58**: 211–216.
7 Kim CS, Jaques PA. Analysis of total respiratory deposition of inhaled ultrafine particles in adult subjects at various breathing patterns. *Aerosol Sci Technol* 2004; **38**: 525–540.
8 Oberdorster GE, Oberdorster E, Oberdorster J. Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles. *Enviorn Health Perspect* 2005; **113**: 823–839.
9 Sioutas C, Delfino RJ, Singh M. Exposure assessment for atmospheric ultrafine particles (UFPs) and implications in epidemiologic research. *Environ Health Perspect* 2005; **113**: 947–955.
10 Lippmann M, Yeates D, Albert R. Deposition, retention, and clearance of inhaled particles. *Br J Ind Med* 1980; **37**: 337–362.
11 Anderson ZJ, Wahlin P, Raaschou-Nielsen O, Ketzel M, Scheike T, Loft S. Size distribution and total number concentration of ultrafine and accumulation mode particles and hospital admissinos in children and the elderly in Copenhagen, Denmark. *Occup Enviorn Med* 2008; **65**: 458–466.
12 Hogg JC, van Eeden S. Pulmonary and systemic response to atmospheric pollution. *Respirology* 2009; **14**: 336–346.
13 US EPA. What is Mountaintop Mining? Mid-Atlantic Mountaintop Mining 2013. Retrieved from http://www.epa.gov/Region3/mtntop/.
14 Hendryx M. Mortality from heart, respiratory, and kidney disease in coal mining areas of Appalachia. *Int Arch Occup Enviorn Health* 2009; **82**: 243–249.

15 Hendryx M, Ahern MM. Relations between health indicators and residential proximity to coal mining in West Virginia. *Am J Public Health* 2008; **98**: 669–671.
16 Hendryx M, Zullig KJ. Higher coronary heart disease and heart attack morbidity in Appalachian coal mining regions. *Prev Med* 2009; **49**: 355–359.
17 Hendryx M. Poverty and mortality disparities in central Appalachia:mountaintop mining and environmental justice. *J Health Disparities Res Pract* 2011; **4**: 44–53.
18 Ahern MM, Hendryx M, Conley J, Fedorko E, Ducatman A, Zullig KJ. The association between mountaintop mining and birth defects among live births in central Appalachia, 1996–2003. *Environ Res* 2011; **111**: 838–846.
19 Esch L, Hendryx M. Chronic cardiovascular disease mortality in mountaintop mining areas of central Appalachian states. *J Rural Health* 2011; **27**: 350–357.
20 Esch LM, Lusk S, Hendryx M, McCawley M. Comparison of particle size distribution and concentration between MTR and non-MTR areas in West Virginia. Prepared for the American Association for Aerosol Research 30th Annual Conference, Orlando, October 2011.
21 Hendryx M, Wolfe L, Luo J, Webb B. Self-reported cancer rates in two rural areas of West Virginia with and without mountaintop coal mining. *J Commun Health* 2011; **37**: 320–327.
22 University of Wisconsin Population Health Institute. West Virginia Rankings. County Health Rankings and Roadmaps 2012, Retrieved from www.countyhealthrankings.org/#app/west-virginia/2012.
23 West Virginia Office of Miners' Health Safety and Training. 2011 Calendar Year Statistical Report. West Virginia Office of Miners' Health Safety and Training 2012. Retrieved from www.wvminesafety.org/2011statisticalreport.htm.
24 United States Census Bureau. State & County QuickFacts. 2011. Retrieved from. http://factfinder2.census.gov/faces/nav/jsf/pages/index.x.html.
25 WV Division of Tourism Research. Quick County Facts:Pocahontas County. WV Department of Tourism 2006, Retrieved from www.pocahontascountywv.com/files/quick_county_facts.pdf.
26 Division of Air Quality. Air Monitoring. West Virginia Department of Enviornmental Protection 2011. Retrieved from http://www.dep.wv.gov/daq/air-monitoring/Pages/dataprocessing.aspx.
27 Stahlhofen W, Rudolf G, James AC. Intercomparison of experimental regional aerosol deposition. *J Aerosol Med* 1989; **2**: 285–308.
28 National Weather Service Forecast Office. Climate Data. National Oceanic and Atmospheric Administration 2009, Retrieved from http://www.nws.noaa.gov/climate/local_data.php?wfo=rlx.
29 Levy JI, Spengler JD. Modeling the benefits of power plant emissions controls in Massachusetts. *J Air Waste Manage Assoc* 2002; **52**: 5–18.
30 MacIntosh DL, Levy JI, Spengler JD. Testimony before the Wisconsin Public Service Commission: Matter of a Pollution Control Construction Permit, Case No. IH-04-03 2003.
31 Wilson WE, Suh HH. Fine and coarse particles:concentration relationships relevant to epidemiological studies. *J Air Waste Manage Assoc* 1997; **47**: 1238–1249.
32 Kolker A, Egle M, Orem W, Tatu C, Hendryx M, McCawley M *et al.* Atmospheric particulate matter in proximitiy to mountaintop coal mines. Prepared for the VM Goldschmidt Conference, Montreal, June 2012.
33 Knuckles T, Stapleton PA, Minarchick VC, Esch L, McCawley M, Hendryx M *et al.* Air pollution particulate matter collected from an Appalachian mountaintop mining site induces microvascular dysfunction. *Microcirculation* 2013; **20**: 158–169.
34 Ghose MK, Majee SR. Characteristics of hazardous airborne dust around an Indian surface coal mining area. *Environ Monit Assess* 2007; **130**: 17–25.