Characterization and Nonparametric Regression of Rural and Urban Coarse Particulate Matter Mass Concentrations in Northeastern Colorado

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Title

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Running Title

Coarse Particulate Matter in Northeastern Colorado

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Authors

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Abstract

The Colorado Coarse Rural Urban Sources and Health study (CCRUSH) is an ongoing study of the relationship between coarse particulate mass concentrations (PM$_{10-2.5}$, particulate matter with diameter between 2.5 and 10µm) and selected health effects. For two urban monitoring sites in Denver, CO and two comparatively rural sites in Greeley, CO, hourly mass concentrations of PM$_{10-2.5}$ and fine particulate matter (PM$_{2.5}$, diameter less than 2.5 µm) are being measured using dichotomous tapered element oscillating microbalances (TEOMs) with Filter Dynamics Measurement Systems (FDMS). This paper presents air quality results from just over a year of PM$_{2.5}$ and PM$_{10-2.5}$ measurements.

Average PM$_{2.5}$ concentrations ranged from 8.5 to 10.1 µg m$^{-3}$ across the four sites, with higher concentrations in Denver than Greeley. Average PM$_{10-2.5}$ concentrations ranged from 8.9 to 15.3 µg m$^{-3}$, with the highest values at the site in northeast Denver. Temporal variability in PM$_{10-2.5}$ was higher than that in PM$_{2.5}$ concentrations at all four sites. The two Greeley sites displayed moderate spatial correlation for PM$_{2.5}$ and high correlation for PM$_{10-2.5}$, whereas the two Denver sites showed lower spatial correlation for both PM sizes. PM$_{10-2.5}$ concentrations in Denver were highest with winds from the direction of the city’s urban core. PM$_{10-2.5}$ concentrations in Greeley were moderately elevated with winds from the southwest, the direction in which Denver lies. Wind speed regressions for PM$_{10-2.5}$ at the Denver sites primarily exhibited resuspension effects, while PM$_{10-2.5}$ concentrations in Greeley showed relatively complex wind speed dependence.

1. Introduction

Coarse particulate matter in the size range from 2.5 to 10 microns (PM$_{10-2.5}$) is believed to be important for human health because particles in this size range are capable
of penetrating to the thoracic region of the lungs when inhaled (Chan and Lippmann, 1980).

Size resolved and chemically speciated data indicate that compared to particulate matter less than 2.5 microns in diameter (PM$_{2.5}$), PM$_{10-2.5}$ is more likely to contain crustal elements such as aluminum, iron, and calcium, but may also contain ions, transition metals, organic and biological material (e.g., Milford and Davidson, 1985; 1987; Boreson, et al., 2004; Heuglin et al., 2005). PM$_{10-2.5}$ is commonly derived from abrasive mechanical processes, including construction and agricultural activities, resuspended road dust, vegetative debris, and sea spray (Patterson and Gillette, 1977; Duce et al., 1976), with emissions from many of these processes depending strongly on wind speed (Harrison et al., 2001). PM$_{10-2.5}$ is also produced from incomplete combustion of solid fuels such as coal and biomass.

Over the past two decades, health studies of particulate matter have focused primarily on PM$_{2.5}$, with less attention given to PM$_{10}$ or PM$_{10-2.5}$. However, Brunekreef and Forsberg (2005) reviewed nearly 60 studies that evaluated health effects of short-term exposure to PM$_{10-2.5}$ and concluded that for some endpoints, including chronic obstructive pulmonary disease, asthma, and respiratory admissions, PM$_{10-2.5}$ could have as strong or a stronger effect than PM$_{2.5}$. Short-term increases in PM$_{10-2.5}$ have also been positively associated with mortality in several studies (e.g., Castillejos et al., 2000; Mar et al., 2000; Ostro et al., 2000; Villeneuve et al., 2003; Zanobetti and Schwartz, 2009). In a recent review of studies of the health effects associated with short-term exposure to ambient PM$_{10-2.5}$, the U.S. Environmental Protection Agency (EPA) concluded that existing evidence is suggestive of a causal relationship between exposures and mortality, cardiovascular effects, and respiratory effects (U.S. EPA, 2009). EPA also recognized several critical uncertainties in epidemiology studies of PM$_{10-2.5}$ impacts, including relatively high exposure error.
compared to PM$_{2.5}$, due to greater expected spatial variability in PM$_{10-2.5}$ concentrations, and limitations in characterization of spatial distributions. Many epidemiologic studies published to date have used differences between PM$_{10}$ and PM$_{2.5}$ concentrations measured at co-located monitors and in some cases monitors located at different sites within the same county to estimate PM$_{10-2.5}$, which contributes further uncertainty in exposure estimation. Furthermore, epidemiologic studies of PM$_{10-2.5}$ have mostly focused on urban areas, where large populations result in greater power for detecting statistically significant effects. Because sources of PM$_{10-2.5}$ may be different in urban compared to rural regions, it is unclear whether exposure to PM$_{10-2.5}$ in smaller communities or rural areas is also linked to adverse health effects.

Because of their size, coarse particles are removed from the atmosphere more quickly than fine particles. As a consequence of both deposition velocity and the intermittent nature of many source processes, concentrations of PM$_{10-2.5}$ are expected to be more spatially and temporally variable than PM$_{2.5}$ concentrations. Wilson et al. (2005) reviewed prior studies and found that reported correlation coefficients between sites within several cities ranged from 0.14 – 0.60 for 24-h average PM$_{10-2.5}$ concentrations; these values are generally lower than those observed for PM$_{2.5}$ or PM$_{10}$ (Wilson et al., 2005). Chen et al. (2007) found an average correlation coefficient of 0.75 between 24-h average PM$_{10-2.5}$ concentrations measured on about 70 days at a central monitor in Chapel Hill, NC and monitors placed at residences within about a 60 km radius. With a year of weekly monitoring at 10 sites across the Los Angeles basin, Pakbin et al. (2010) found pairwise correlations ranging from 0.04 for 24-h average PM$_{10-2.5}$ concentrations from an
industrial site in Long Beach and concentrations at suburban monitors, to 0.80 for PM$_{10-2.5}$ concentrations at a pair of coastal sites located within a few kilometers of each other.

Most studies of seasonal variability in PM$_{10-2.5}$ concentrations have observed the highest concentrations in summer, but exceptions occur due to specific source activity patterns (Thornburg et al., 2009; Pakbin et al., 2010). Harrison et al. (2001) measured PM$_{10-2.5}$ continuously at five sites in England over a three-year period. They observed higher PM$_{10-2.5}$ concentrations on weekdays than on weekends, and found the fraction of PM$_{10}$ contributed by PM$_{10-2.5}$ was highest in the spring and summer. Moore et al. (2010) reported correlation coefficients of 0.1 – 0.4 for continuous hourly PM$_{10-2.5}$ concentrations measured at three sites across the Los Angeles basin. In their study, the most pronounced diurnal variation in PM$_{10-2.5}$ concentrations was observed at a site near Riverside, CA, with less diurnal variability in concentrations measured near downtown Los Angeles and at a desert location about 110 km NW of downtown. Daytime or evening maxima were observed at all three locations.

This paper presents just over a year of mass concentration data from continuous PM$_{10-2.5}$ and PM$_{2.5}$ sampling conducted in Denver and Greeley, Colorado, as part of the Colorado Coarse Rural Urban Sources and Health (CCRUSH) study. CCRUSH is a multi-year study of the relationship between PM$_{10-2.5}$ mass concentrations and adverse health effects, including cardiopulmonary emergency department visits and adverse birth outcomes. Denver and Greeley were selected for the study to allow comparison of the composition and relative health effects of coarse PM in urban and rural communities. For two sites in Denver and two sites in Greeley, hourly mass concentrations of PM$_{10-2.5}$ and PM$_{2.5}$ were measured using dichotomous tapered element oscillating microbalances (TEOMs) with
Filter Dynamics Measurement Systems (FDMS). The TEOM sampling began in January 2009 and will continue for three years. At the end of the sampling period, the mass concentration data will be analyzed with local data on birth outcomes and emergency department visits to assess and compare associations between the two communities.

This paper examines spatial and temporal variations in hourly and 24-h average concentration values for PM$_{10-2.5}$ and PM$_{2.5}$. The paper also examines the influence of hourly wind speed and wind direction on the mass concentrations. Nonparametric regression (NPR; Henry et al., 2002, 2009; Yu et al., 2004; Kim and Hopke, 2004) was used to characterize the wind speed and wind direction relationships, and help understand differences in mass concentrations across sampling locations.

2. Methods

2.1 Sampling Locations

Continuous particulate mass concentrations were measured at two locations in Denver and Greeley, CO (Table 1). Greeley is located in Weld County, which has a population of 254,759 (U.S. Census, 2009a), an area of 10,417 km$^2$, and is roughly 50 miles northeast of Denver. Greeley has a population of 92,625 (U.S. Census, 2009b) and an area of 77.7 km$^2$. Agriculture and oil and gas extraction are among the county’s leading economic activities. In contrast, the City and County of Denver has a population of 610,345 (U.S. Census, 2009c) and an area of 401.3 km$^2$, with a highly mixed economy (the urban area$^1$ of Denver-Aurora has a population of 1.98 million and an area of 1291.9 km$^2$). Denver is transected by major interstate highways and experiences much greater traffic volumes.

$^1$An urban area consists of core census block groups or blocks that have a population density of at least 1,000 people per square mile (386 people/km$^2$) and surrounding census blocks that have an overall density of at least 500 people per square mile (193 people/km$^2$) (U.S. Census, 2000).
than Greeley. Correspondingly, PM$_{10-2.5}$ concentrations in Denver are expected to be dominated by resuspended urban road dust, while agricultural activities (e.g., feedlots, soil preparation, and ditch burning) are expected to be relatively important sources in Greeley.

Monitors were located on the roofs of two elementary schools in Denver: Alsup and Edison (11.1 km apart). Monitors were located in Greeley on the roof of Maplewood elementary school and in the HVAC system enclosure at McAuliffe elementary school (4.5 km apart).

<Table 1. Monitoring Sites for the CCRUSH Study>

### 2.2 Particulate Matter Monitoring Methods

Tapered element oscillating microbalance (TEOM), model 1405-DF (ThermoFisher Scientific, Waltham, MA), ambient PM monitors were located at each site. Three monitors (Alsup, Edison, and Maplewood) were located outside on roofs and housed in enclosures (Complete Outdoor Enclosure for TEOM Series 1405, ThermoFisher Scientific, Waltham, MA) designed to maintain appropriate instrument conditions. At extreme high and low ambient temperatures the enclosures failed to maintain appropriate instrument operating conditions, which resulted in data removal. The monitor located at McAuliffe was located just below the roof in a HVAC system crawl space and was equipped with an in-house designed foam enclosure equipped with a commercial air conditioner/heater unit set to 21.1°C.

The TEOM 1405-DF is dichotomous and is equipped with a Filter Dynamic Measurement System (FDMS) to correct for semi-volatiles and water losses from mass measurement filter surfaces. The mass measurements are made by a vertical oscillating
tapered glass element with a TX-40 TEOM filter placed on the end. Particulate mass is
deposited as aerosol passes through the filter, which changes the natural oscillating
frequency of the tapered glass element. The frequency change is related to filter mass
change by simple vibration theory. The ambient mass concentrations are calculated by the
change in mass and volumetric air flow rates.

The instrument operates by sampling in two modes that alternate every six
minutes. In the “Base” measurement mode, the sample stream is held at 30 °C, with the
aerosol passing directly to the mass measurement filter. The effect of water is reduced in
the TEOM 1405-DF by the use of a Nafion™ membrane diffusion dryer in each particulate
channel. In the Base mode, mass can be both lost and gained from the filter, depending on
the amount of semi-volatiles present. Thus in this mode it is the net mass change that is
recorded. In the “Reference” mode, after the dryer, the sample is diverted through the
cooled FDMS filter, which is held at 4°C. This filter removes material that will condense at
4°C or below. This filtered air stream is then directed through the TEOM filter and the mass
change on the filter recorded. Reference mode values are commonly negative due to mass
loss from the TEOM filter, but adsorption or absorption of organic gases may also occur,
resulting in mass gain (Green, 2009). The mass change during the Reference mode due to
gas-phase sampling artifacts is assumed to be equal to the mass change that occurred
during the previous Base measurement. The time series of Reference mass concentrations
are thus subtracted from the Base measurements, correcting for sampling artifacts and
approximating the true aerosol mass concentration. This provides a total mass
concentration for each 12-minute time step, with the first 6 minutes providing the Base and
the second 6 minutes the Reference concentration. The instruments were operated at flow
rates prescribed by the manufacturer: 1.67 lpm (PM$_{10-2.5}$), 3 lpm (PM$_{2.5}$), and 12 lpm (bypass).

TEOM instrument maintenance was performed monthly at each site and consists of changing TEOM and FDMS filters; cleaning the PM$_{10}$ inlet, virtual impactor, and FDMS valve; checking for seal leaks in the mass transducer, FDMS valve and FDMS filter holder; flow audit and calibration; and an instrument leak check. Operators ensured the instrument was operating properly before leaving the site. Other regular maintenance was performed as needed and included exchanging Nafion diffusion dryers, pump maintenance, and replacing mass transducer, FDMS valve and FDMS filter holder seals. Ball valves were installed between the virtual impactor and diffusion dryers to increase ease of access to sample lines for flow audits, which were performed at a higher frequency than prescribed by the manufacturer. A single external filter on the bypass flow line was used to extend pump life.

To assure the highest quality data were used for analysis, extensive quality assurance protocols were developed. Upon arriving at a monitoring site, an instrument status log, maintenance log, comment log, and flow audit/leak check log were completed. The status log was filled out before and after maintenance to assure the instrument conditions did not change due to operator intervention. The TEOM data were downloaded manually each month prior to instrument maintenance. The discrete section of data from the last site visit to the current visit was downloaded via the available USB port on the front of the instrument. This process closed the previous section of data before the operator interfered with instrument operation. Using the ePort software provided by Thermo Scientific, the entire TEOM database was also downloaded. Data were transferred from a
field laptop or flash drive to a desktop computer immediately upon arriving back at the University of Colorado. All data files were backed-up weekly on university servers, as well as monthly on a portable external hard drive.

Discrete data sections downloaded via USB flash drive were processed by a code developed in-house. Log files for each data section were created that specified data filenames, whether maintenance occurred, whether to output hourly averages, saved data interval, number of hours to remove after maintenance occurred, and number of hours to shift the time stamp into Mountain Standard Time (MST). Rows were flagged as missing data if the status code reported the following errors: power failure, database failure, FDMS valve failure, mass transducer failures, any channel flow deviating from set flow rate more by than 10%, either channel reading filter loading above 90%, or heater tube temperatures deviating from set temperature by more than 2%. Instrument problems were flagged as well and included: vacuum pressures above 40.5 kPa, temperatures more than 0.5°C below the specified set point, or if channel relative humidity was above 98%.

Equations 1 – 6 were applied to correct for PM$_{2.5}$ mass depositing in the PM$_{10-2.5}$ channel due to the virtual impactor. In the following equations, $Q$ represents the volumetric flow rate through the indicated channel. $PM$ represents the mass concentration, with the TEOM label indicating raw TEOM data. It was assumed that both Base and Reference channels followed the same correction, i.e., that semi-volatile mass loss was proportional to the amount of total mass in each channel. It can be shown that PM$_{10}$ is conserved with and without the applied corrections.

$$\frac{Q_{PM_{10-2.5}}}{Q_{Total}} = \frac{1.67 \text{lpm}}{16.67 \text{lpm}} = 0.1$$ (1)
\[
\frac{Q_{PM \, 2.5}}{Q_{Total}} = \frac{15 \, lpm}{16.67 \, lpm} = 0.9 \quad (2)
\]

\[
PM_{2.5 \, Base} = \frac{PM_{2.5 \, Base \, (TEOM)}}{Q_{PM \, 2.5}} \quad (3)
\]

\[
PM_{2.5 \, Ref} = \frac{PM_{2.5 \, Ref \, (TEOM)}}{Q_{PM \, 2.5}} \quad (4)
\]

\[
PM_{10-2.5 \, Base} = PM_{10-2.5 \, Base \, (TEOM)} - \frac{Q_{PM \, 10-2.5}}{Q_{Total}} \left( \frac{PM_{2.5 \, Base \, (TEOM)}}{Q_{PM \, 2.5}} \right) \quad (5)
\]

\[
PM_{10-2.5 \, Ref} = PM_{10-2.5 \, Ref \, (TEOM)} - \frac{Q_{PM \, 10-2.5}}{Q_{Total}} \left( \frac{PM_{2.5 \, Ref \, (TEOM)}}{Q_{PM \, 2.5}} \right) \quad (6)
\]

The hourly average and standard error (i.e., the standard deviation divided by the square root of the number of measurements in the hour), of all downloaded variables were calculated and exported, excluding data flagged as missing. Logs used to process data were accessed to compile full data sets, filling in missing sections of data between discrete data sets with missing data flags or combining same-hour measurements with a weighted average based on the number of measurements made in that hour in each separate data set. Three scenarios were identified that required further data processing: major events of mass loss from filter surfaces, instances of highly variable noise due to temperature aliasing from rapid or oscillating changes of enclosure temperature or other sources, or instances of elevated standard error when a non-removal status code had been triggered. The mass loss incidents were identified if the calculated mass concentration was less than
the 1st percentile of the time series and the standard error of the measurement was above the 95th percentile. Incidents of induced highly variable noise were identified if the calculated concentration was below the 1st percentile and the subsequent measurement was greater than the 99th percentile or vice versa. The third scenario was triggered when a non-zero status code was recorded and the calculated hourly mass concentration standard error was above the 95th percentile. Each occurrence of one of these three scenarios was assessed manually to determine if data should be removed for final hourly average data sets. Data were then filtered for hours with less than 75% completeness. Daily averages were calculated from cleaned hourly average data sets and days missing more than 75% of completed hours were also removed. Completeness statistics for each site are shown in Table 2.

The data set reported in this paper has been labeled Phase 1, which is a result of cutting off the currently validated results when instruments were updated to a new version of the TEOM 1405-DF firmware. This update required exchanging a physical flash card; after the update instrument settings were unintentionally reset to defaults. The start and end dates and hours of each site’s Phase 1 data are listed in Table 2. Sampling began on different dates at each site, and completeness varies by site based on instrument maintenance issues.

<Table 2. Sampling Period and Completeness for Phase 1 Hourly Average PM Concentration Data>

The TEOM 1405-DF is a relatively new instrument and correspondingly posed numerous challenges in our effort to produce continuous time series of mass concentration data. Through collaboration with Thermo Scientific, solutions were found for most
problems, but they nonetheless led to substantial gaps in our time series. Denver and Greeley experience significant seasonal temperature variations. The air heating and cooling systems incorporated into the Thermo Scientific TEOM 1405-DF enclosures were unable to adequately condition the space within the enclosures when ambient temperatures were very high or low. Numerous measurements from midday throughout the summer were suspect and hence censored due to large hourly variability associated with increased TEOM mass transducer temperatures. This high measurement variability mostly originated in the Reference channel, where hourly standard errors sometimes exceeded 500 μg m⁻³. Cold temperature extremes were less of an issue, though the operating temperatures of the FDMS systems occasionally dropped below 4°C. These changes in FDMS operating temperature were not accompanied by significant increases in variability of mass concentration measurements, so corresponding data were not removed. A further problem with the HVAC system occurred at Alsup and Maplewood, where insulation near the blowers peeled off and either shredded or blocked the blowers.

Malfunction of the Nafion dryer assemblies and pumps also lead to gaps in the time series. Dryer assemblies had to be replaced every 7-10 months and the pumps rebuilt every 12 months, in each case about six months earlier than the manufacturer's maintenance recommendations. Premature pump failure may be partly due to low ambient atmospheric pressures in Colorado, which are typically about 85.1 kPa. In addition, the bypass flow controller of the TEOM installed at McAuliffe failed when the inlet system did not adequately dispose of water vapor in the bypass line, resulting in condensation when the air was cooled in the enclosure.
Finally, a significant gap in the McAuliffe dataset occurred due to seal leaks within the FDMS valve system that were not detected through the leak check process. The problem was only identified upon later inspection of the data. In response, we modified our monthly maintenance protocol to include disassembling the FDMS valve to verify that no seals failed, and to process and examine data on-site to verify the absence of leaks.

### 2.3 Meteorological Data

Hourly meteorological data were obtained for locations at or near each of the monitoring sites, as indicated in Table 3. The Colorado Department of Public Health and Environment (CDPHE) operates the Carriage and Alsup sites, while the National Oceanic and Atmospheric Administration (NOAA) operates the Weld County Airport site. Meteorological data summaries and wind roses are shown in the Supplemental Information. Vector averaged wind speed was used in the data analyses.

<Table 3. Characteristics of the Meteorological Stations Used in This Study>

### 2.4 Data Analysis and Nonparametric Regression

The results section presents standard descriptive statistics for $\text{PM}_{2.5}$ and $\text{PM}_{10-2.5}$ mass concentrations, along with the coefficient of divergence (COD), which is a measure of uniformity. Results of nonparametric regression of concentrations versus wind speed and direction are also presented. All data analyses used concentration data that were error code filtered. No negative censoring or replacement was performed in any of the analyses, except when calculating the COD.

The coefficient of divergence (Wilson et al. 2005) is defined by:
\[ COD = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left( \frac{X_{ij} - X_{ih}}{X_{ij} + X_{ih}} \right)^2} \]  

(7)

where \( i \) is the sample, and \( j \) and \( h \) index different measurement sites. A COD value near 0 represents perfect uniformity, and a value of 1 represents total heterogeneity. The COD loses meaning when negative values are included, so in calculating this statistic negative values in the dataset were replaced with zeros.

The data set used in the NPR was different than that used in the other analyses, as it required wind data and mass concentrations for each hour, both of which had missing data. Additionally, any data point with a corresponding wind speed value below 1 m s\(^{-1}\) was excluded from the NPR analysis. Exclusion of these periods with relatively calm winds sharply reduced the number of observations used in the NPR analyses compared to the full sets of hourly mass concentration data.

Nonparametric regression was used to estimate the expected concentration \( C_i \) from each wind direction or wind speed \( i \) by including all observations using weighting kernels, giving less weight to observations far from the point at which the estimate is being calculated and vice-versa. The Gaussian kernel:
\[
K_1(x) = (2\pi)^{-\frac{1}{2}} e^{-\left(0.5x^2\right)}, \quad -\infty < x < \infty, \quad x = \frac{\theta - W_i}{\Delta \theta}
\]  
(8)

was used for the wind direction regressions and the Epanechnikov kernel:

\[
K_2(x) = 0.75\left(1 - x^2\right), \quad -1 < x < 1
\]  
(9)

was used for wind speed regressions (Henry et al., 2002). In the kernels, \(\theta\) is the wind direction or speed for which the estimate is made, \(W_i\) is the wind speed or wind direction value at time \(i\), and \(\Delta \theta\) is the smoothing parameter. The concentration \(C(\theta)\) at a given wind speed or direction is then estimated by the Nadaraya-Watson estimator, defined as:

\[
C(\theta) = \frac{\sum_{i=1}^{n} K\left(\frac{\theta - W_i}{\Delta \theta}\right) C_i}{\sum_{i=1}^{n} K\left(\frac{\theta - W_i}{\Delta \theta}\right)}
\]  
(10)

where \(K\) references the appropriate kernel. In this work, an average value of the cross-validation derived smoothing parameters was used for all the sites and both size regimes to allow direct comparison among them. The full width at half maximum, i.e., the width of the kernel at the point where it is half of its maximum, was 28.26 degrees and 1.41 m s\(^{-1}\) for the wind direction and speed regressions, respectively. Ninety-five percent confidence intervals of the regression estimates were calculated.

3. Results

Table 4 presents summary statistics for the 24-h average PM\(_{2.5}\) and PM\(_{10-2.5}\) concentrations measured at the four study sites. Average PM\(_{2.5}\) concentrations ranged from 8.5 to 10.1 \(\mu\)g m\(^{-3}\) across the four sites. Average concentrations of PM\(_{2.5}\) were somewhat higher at the two Denver sites than at the two sites in Greeley. Average PM\(_{10-2.5}\) concentrations ranged from 8.9 to 15.3 \(\mu\)g m\(^{-3}\). PM\(_{10-2.5}\) concentrations were sharply higher...
at the Alsup site in northeast Denver than at the other three locations. Temporal variability
in PM$_{10-2.5}$ concentrations was higher than that in PM$_{2.5}$ concentrations, with COV values for
24-h average PM$_{10-2.5}$ ranging from 0.6 to 0.8 and those for PM$_{2.5}$ all near 0.5. Across thefour sites, 95$^{th}$ percentile 24-h average concentrations ranged from 16.3 to 19.9 µg m$^{-3}$ for
PM$_{2.5}$ and from 18.8 to 34.8 µg m$^{-3}$ for PM$_{10-2.5}$.

The two Greeley sites had the highest spatial correlation for 24-h average
concentrations of both PM$_{2.5}$ and PM$_{10-2.5}$, with Pearson’s R values of 0.82 for PM$_{2.5}$ and 0.97
for PM$_{10-2.5}$. Concentrations measured at the two Denver sites showed lower correlation,
with Pearson’s R values of 0.64 for PM$_{2.5}$ and 0.70 for PM$_{10-2.5}$. Concentrations at the sites in
Denver showed lower correlation compared with those measured at the Greeley sites, for
both PM$_{2.5}$ and PM$_{10-2.5}$. The COD was calculated among all site pairs. The pair of Greeley
sites displayed the most homogeneous 24-h average PM$_{2.5}$ and PM$_{10-2.5}$ concentrations
(COD=0.13 for both sizes), while the pair of Edison and Alsup was somewhat more
heterogeneous (COD=0.30 for PM$_{2.5}$ and 0.21 for PM$_{10-2.5}$).

<Table 4. Summary Statistics for 24-h Average PM$_{2.5}$ and PM$_{10-2.5}$ Concentrations>

Table 5 compares median concentrations of PM$_{2.5}$ and PM$_{10-2.5}$ between weekends
and weekdays at each site, as well as between daytime (6 am – 6 pm) and nighttime (6 pm
– 6 am) hours. Significance of differences was assessed using the Kruskal-Wallis test. For
PM$_{2.5}$, weekend concentrations were higher than weekday concentrations at all four sites,
though the difference is not statistically significant at Edison and Alsup. In contrast, for
PM$_{10-2.5}$, weekday concentrations were uniformly significantly higher than weekend
concentrations. Daytime concentrations of PM$_{2.5}$ were higher than nighttime
concentrations at Edison and McAuliffe, whereas the opposite was true for Alsup and Maplewood. Daytime concentrations of PM$_{10-2.5}$ were significantly higher than nighttime concentrations at all four sampling sites.

**Table 5. Comparison of Median 1-h Average Concentrations by Weekday/Weekend and Daytime/Nighttime**

Figure 1 shows median concentrations of PM$_{2.5}$ and PM$_{10-2.5}$ plotted by month. The four sites showed similar monthly patterns in each size regime. PM$_{2.5}$ concentrations showed relatively little monthly variation compared to concentrations of PM$_{10-2.5}$.

**Figure 1(a). Monthly median mass concentrations at the four monitoring sites for PM$_{2.5}$.**

**Figure 1(b). Monthly median mass concentrations at the four monitoring sites for PM$_{10-2.5}$.**

Figure 2 shows that the median hourly average concentrations of PM$_{2.5}$ were less variable as a function of time of day than PM$_{10-2.5}$ concentrations. PM$_{2.5}$ concentrations at all sites generally increased in the morning, around 6 am – 10 am, dropped from 1 pm – 6 pm, and once again increased in the evening, around 7 pm – 11 pm. The Alsup site showed the biggest peak, at 7 am, slightly earlier than the morning peaks at the other sites. For PM$_{10-2.5}$, concentrations at Alsup peaked at about 8 am. PM$_{10-2.5}$ concentrations at Edison peaked at about 11 am, and decreased throughout the day. The concentrations at the two Greeley sites were higher during the daytime hours, but did not show any major peaks.

**Figure 2. Median mass concentrations by time of day at the four monitoring sites for (a) PM$_{2.5}$ and (b) PM$_{10-2.5}$.**
Nonparametric regression results showing relationships of hourly PM$_{2.5}$ and PM$_{10-2.5}$ concentrations with wind speed and wind direction are presented for each site in Figures 3–6. It should be noted that in some cases, limited data in the tails of the wind speed regressions influenced the curve shapes in these regions. Also note that hourly average concentrations reach sharply higher values than the 24-h average concentrations summarized in Table 4.

The NPR results for the Edison site (Figure 3) show higher concentration estimates for both size fractions when the wind is from the northeast. Estimated PM$_{10-2.5}$ concentrations increase with wind speed, while for PM$_{2.5}$, the estimated concentrations decrease initially, then increase at the highest wind speeds.

The Alsup site had the highest median concentrations for both sizes. Estimated PM$_{2.5}$ concentrations at Alsup peak with winds from the southwest, with a general decrease in concentration with increasing wind speeds (Figure 4a). Estimated PM$_{10-2.5}$ concentrations are markedly higher with higher speed winds from the west (Figure 4b).

The Maplewood and McAuliffe sites have similar NPR results (Figures 5 and 6). The NPR results show relatively uniform PM$_{2.5}$ concentration estimates as a function of wind direction (Figures 5a and 6a). At both sites, the NPR results for PM$_{2.5}$ show a dilution effect with increasing wind speed. The NPR results for PM$_{10-2.5}$ for both sites are much more homogeneous than those for either Denver site, with only three subtle peaks corresponding to winds from the northwest, west, and southwest (Figures 5b and 6b). The PM$_{10-2.5}$ relationships with wind speed at both Greeley sites show initial decreases, then increases for speeds above 3.5 m s$^{-1}$, followed by further decreases with wind speeds above 5 m s$^{-1}$.
4. Discussion

4.1. Comparison of Mass Concentrations and Spatial Correlation with other Locations

Comparison of PM data sets across studies is complicated by the use of different instruments and measurement methods. Federal reference methods (FRMs) and federal equivalence methods (FEMs) for PM have been discussed previously (U.S. EPA 2004; 2009). The FRM for PM$_{10-2.5}$ is calculated as the numeric difference between concurrent and co-located PM$_{10}$ and PM$_{2.5}$ concentrations as measured by FRM low-volume filter samplers of the same make and model (U.S. EPA 2009). The TEOM 1400AB and 1405 for PM$_{10}$ have been designated as FRM methods for PM$_{10}$. The TEOM 1405-DF has been designated as an FRM method for PM$_{2.5}$, but not (to date) as an FRM for PM$_{10-2.5}$ (U.S. EPA, 2010).
The TEOM 1405-DF has been designed to minimize sampling artifacts, both positive and negative. Positive artifacts are a result of excess mass collection typically caused by gas-phase adsorption onto the collection media. Negative artifacts are a result of reduced mass collection typically caused by semi-volatile species that were in the particle phase but shift to the gas-phase after collection due to collection temperatures that are higher than ambient or pressures that are slightly less than ambient. For example, when PM$_{2.5}$ concentrations were measured by a pair of TEOMs, one operated at 50°C and the other operated at 30°C, the TEOM held at a higher temperature yielded consistently lower concentrations, as the higher temperature yielded only less semi-volatile mass (Zhu et al., 2006; Grover et al., 2005). The TEOM 1405-DF operates at 30°C and also utilizes an FDMS which adjusts for filter adsorption artifacts. Results from previous studies generally show that for PM$_{2.5}$ the TEOM FDMS measures higher concentrations compared to the FRM, especially as the ambient temperature increases (Grover et al., 2005; Schwab et al., 2006; Zhu et al., 2006) as the FRM does not adjust for adsorption artifacts (Solomon and Sioutas, 2008). In the end, it is important to remember that comparison across studies that have used different measurement techniques will have slight biases associated with the technique differences. Thus, the PM$_{10-2.5}$ data discussed below should be viewed as only roughly comparable across studies.

CDPHE reports mass concentrations of PM$_{2.5}$ and PM$_{10}$ from two urban monitoring sites in Denver: the CAMP site at 2105 Broadway (lat. 39.75, long. -104.99) and the Denver Municipal Animal Shelter (DMAS) site at 678 S. Jason Street (lat. 39.70, long. -105.00). Both sites house TEOM FDMS instruments for continuous PM$_{2.5}$ monitoring and TEOM-1400AB instruments for PM$_{10}$. We obtained data for these monitoring sites from CDPHE and
estimated PM$_{10-2.5}$ concentrations by subtraction. Descriptive statistics for both size ranges were then calculated for comparison with results from the four CCRUSH TEOM sites. For the time period from 1/1/09 – 2/28/10, the mean 24-h average PM$_{2.5}$ concentration at CAMP was 8.3 µg m$^{-3}$, with a 95$^{\text{th}}$ percentile value of 16.6 µg m$^{-3}$ and COV of 0.5. The mean PM$_{2.5}$ concentration at DMAS was 11.1 µg m$^{-3}$, with a 95$^{\text{th}}$ percentile value of 19.6 µg m$^{-3}$ and COV of 0.4. Mean PM$_{2.5}$ concentrations at the two CDPHE monitors thus bracket those observed at our four monitoring sites. The Pearson’s R correlation coefficient for 24-h average PM$_{2.5}$ concentrations at the two CDPHE monitors was 0.82. Correlation coefficients for 24-h average PM$_{2.5}$ concentrations at the CDPHE monitors compared with those we measured at Alsup and Edison ranged from 0.68 – 0.87.

The mean 24-h average PM$_{10-2.5}$ concentration at CAMP was 15.7 µg m$^{-3}$, with a 95$^{\text{th}}$ percentile value of 27.5 µg m$^{-3}$ and COV of 0.4. The mean PM$_{10-2.5}$ concentration at DMAS was 12.9 µg m$^{-3}$, with a 95$^{\text{th}}$ percentile value of 28.4 µg m$^{-3}$ and COV of 0.7. PM$_{10-2.5}$ concentrations at the CDPHE sites are thus comparable to those we observed at Alsup, and higher than those observed at our other study sites. The Pearson’s R correlation coefficient for 24-h average PM$_{10-2.5}$ concentrations at the two CDPHE monitors was 0.61. Correlation coefficients for PM$_{10-2.5}$ concentrations at the CDPHE monitors compared with those measured at Alsup and Edison range from 0.60 to 0.83.

U.S. EPA (2009) presents distributions of 24-h average PM$_{2.5}$ and PM$_{10-2.5}$ mass concentrations measured from 2005 – 2007 from FRM monitors across the country that report to the agency’s Air Quality System. For PM$_{2.5}$, the national mean 24-h average concentration was 12 µg m$^{-3}$ and the 5$^{\text{th}}$ and 95$^{\text{th}}$ percentile values were 4 µg m$^{-3}$ and 28 µg m$^{-3}$, based on nearly 350,000 observations. For Denver, U.S. EPA (2009) reports a mean 24-
h average PM$_{2.5}$ concentration during 2005–2007 of 9 µg m$^{-3}$ and 5$^{th}$ and 95$^{th}$ percentile values of 3 µg m$^{-3}$ and 18 µg m$^{-3}$, respectively, based on 4192 observations. Results from the CDPHE monitors discussed above and from the TEOM sampling conducted in this study during 2009–2010 show similar mean PM$_{2.5}$ concentrations to those EPA reports for Denver, but with greater variability, as indicated above in Table 4.

For PM$_{10-2.5}$, U.S. EPA (2009) reported concentrations estimated from co-located monitors using low-volume FRM filter samplers. The national mean 24-h average PM$_{10-2.5}$ concentration was 13 µg m$^{-3}$, with 5$^{th}$ and 95$^{th}$ percentile values of 1 µg m$^{-3}$ and 33 µg m$^{-3}$, respectively, based on just over 12,000 observations. For PM$_{10-2.5}$ in Denver, EPA reported a mean concentration of 20 µg m$^{-3}$ with 5$^{th}$ and 95$^{th}$ percentile values of 4 and 42 µg m$^{-3}$, based on 353 observations. In comparison to the values U.S. EPA (2009) reports for Denver, the results from the CDPHE monitoring discussed above and results from our study suggest lower mean PM$_{10-2.5}$ concentrations. Differences could be due to differences in sampling methods and monitoring locations or changes in pollutant levels over time.

Spatial representativeness is a critical concern when air quality monitoring data are used in epidemiological studies, as intended with data from CCRUSH. As mentioned in the introduction, concentrations of PM$_{10-2.5}$ have generally been expected to be more variable than those of PM$_{2.5}$. Results from this study indicate that 24-h average PM$_{10-2.5}$ concentrations are somewhat more temporally variable than those for PM$_{2.5}$, with coefficients of variation ranging from 0.6-0.8 for PM$_{10-2.5}$ and near 0.5 for PM$_{2.5}$. Data from CDPHE show a comparatively low COV for PM$_{10-2.5}$ concentrations at CAMP. For both size classes, spatial correlation was relatively strong for the two monitors in Greeley, compared to the monitors located in Denver. The correlation coefficient of 0.97 for PM$_{10-2.5}$...
concentrations from the two Greeley locations, which are located 4.5 km apart, is also relatively high compared to correlation coefficients reported for pairs of PM$_{10-2.5}$ monitors in other cities (Wilson et al., 2005; Pabkin et al., 2010; U.S. EPA, 2009).

**4.2. Temporal patterns in PM$_{2.5}$ and PM$_{10-2.5}$**

This study indicates temporal patterns in PM mass concentrations vary substantially by size regime. PM$_{10-2.5}$ generally had higher weekday, daytime concentrations, while PM$_{2.5}$ generally had higher weekend, nighttime concentrations. These temporal patterns likely resulted from local sources as well as meteorology. For example, relatively high PM$_{2.5}$ concentrations in the morning hours might have been due to temperature inversions in addition to source activity.

The day of week patterns showed relatively high median PM$_{10-2.5}$ concentrations during weekdays, likely corresponding to higher anthropogenic activity on weekdays than weekends. Harrison et al. (2001) likewise reported higher PM$_{10-2.5}$ concentrations on weekdays than weekends, for two sites in London and across all seasons. Although median PM$_{2.5}$ concentrations were relatively constant across the week, compared to PM$_{10-2.5}$ concentrations, the median PM$_{2.5}$ concentrations in this study were highest on the weekends at each site (although the difference was not statistically significant at two sites). This result was unexpected. In comparison, the Denver Aerosol Sources and Health study (DASH; Vedal et al., 2009), which performed daily PM$_{2.5}$ filter sampling for 4.5 years at Palmer Elementary School in Denver, found a significantly higher weekday median (7.1 µg m$^{-3}$) than weekend median (6.5 µg m$^{-3}$) (unpublished statistics).

Compared to PM$_{2.5}$ levels, concentrations of PM$_{10-2.5}$ exhibited relatively strong diurnal variability. This finding is consistent with the shorter residence time of PM$_{10-2.5}$ in
the atmosphere. For both PM$_{2.5}$ and PM$_{10-2.5}$, the Alsup site showed a relatively pronounced morning peak in concentrations, which might reflect the influence of local industrial activity and traffic. Harrison et al. (2001) and Moore et al. (2010) similarly found elevated PM$_{10-2.5}$ concentrations during daytime hours, at monitoring sites in London and the Los Angeles area, respectively.

4.3. Source Identification and Physical Processes

In this study, source regions of greater PM$_{2.5}$ and PM$_{10-2.5}$ impact were identified with NPR for wind speed and direction. For the Denver sites, NPR results for both pollutants point towards the more densely populated, highly travelled, and industrialized core of the city as a significant source area. There are no known major point sources near the Edison site, but the westerly peak in the NPR results for PM$_{10-2.5}$ at Alsup is likely influenced by a sand and gravel operation 1 km west of the monitor, in addition to a major interstate highway junction (I-76 and I-270) just west of that.

NPR results for wind direction for the two Greeley monitoring sites are similar to each other, with moderately elevated concentrations of PM$_{2.5}$ and PM$_{10-2.5}$ with winds from the south and southwest, respectively. These wind directions and the similarity across the two Greeley sites are consistent with expectations for a regional-scale PM contribution centered on the Denver metropolitan area, which is south-southwest of Greeley. We are not aware of nearby point sources that are south or southwest of the Greeley monitoring sites.

The NPR results for PM$_{2.5}$ dependence on wind speed show general agreement across all four sites, showing a dilution effect with increasing wind speed. Results for PM$_{10-2.5}$ are more complicated. The PM$_{10-2.5}$ wind speed regressions for the two Denver sites in
this study primarily suggest resuspension effects, with concentrations increasing with wind speeds up to 8 m s\(^{-1}\) at Edison and 10 m s\(^{-1}\) at Alsup. The PM\(_{10-2.5}\) concentrations at the two Greeley sites showed relatively complex wind speed dependence. They decreased for wind speeds up to about 3 m s\(^{-1}\), increased with wind speeds between 3 and 6 or 7 m s\(^{-1}\), and then decreased again at higher wind speeds. For comparison, Harrison et al. (2001) found a U-shaped curve for the wind speed dependence of PM\(_{10-2.5}\) mass concentration measurements taken near a roadway in Birmingham Hodge Hill, England, suggesting dilution at wind speeds below about 4 m s\(^{-1}\) and resuspension at higher wind speeds.

Moore et al. (2010) found positive correlation between PM\(_{10-2.5}\) concentrations and wind speed for three Los Angeles area sites during the dry seasons, but negligible or negative correlation in winter. Once additional data are available for our study, seasonal analysis and consideration of additional meteorological variables related to resuspension could assist in interpreting the relationship between wind speed and PM\(_{10-2.5}\) concentrations at our monitoring sites.

**Acknowledgments**

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References


December 27, 2010.


Supporting Information Figure SI.1

Supporting Information Figure SI.2

Supporting Information Figure SI.3
<Supporting Information Figure SI.4>
Table 1. Monitoring Sites for the CCRUSH Study

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Table 2. Sampling Period and Completeness for Phase 1 Hourly Average PM Concentration Data

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### Table 3. Characteristics of the Meteorological Stations Used in This Study

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Table 4. Summary Statistics for 24-h Average PM\textsubscript{2.5} and PM\textsubscript{10-2.5} Concentrations

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Table 5. Comparison of Median 1-h Average Concentrations by Weekday/Weekend and Daytime/Nighttime

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<td>0.000*</td>
<td>0.000*</td>
</tr>
<tr>
<td>Daytime (µg m$^{-3}$)</td>
<td>8.3</td>
<td>11.9</td>
<td>6.8</td>
<td>6.8</td>
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<tr>
<td>Nighttime (µg m$^{-3}$)</td>
<td>5.2</td>
<td>8.6</td>
<td>5.1</td>
<td>4.7</td>
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<tr>
<td>p-value</td>
<td>0.000*</td>
<td>0.000*</td>
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<td>0.000*</td>
</tr>
</tbody>
</table>

*Statistically significant at the 0.05 level
Figure 1(a). Monthly median mass concentrations at the four monitoring sites for PM$_{2.5}$. 
Figure 1(b). Monthly median mass concentrations at the four monitoring sites for PM$_{10-2.5}$. 
Figure 2. Median mass concentrations by time of day at the four monitoring sites for (a) PM$_{2.5}$ and (b) PM$_{10-2.5}$. 
Figure 3. Nonparametric regression results for (a) PM2.5 (n = 4028) and (b) PM10-2.5 (n = 4028) mass concentrations at Edison, showing relationships with wind direction and wind speed.
Figure 4. Nonparametric regression results for (a) PM2.5 (n = 7626) and (b) PM10-2.5 (n = 7496) mass concentrations at Alsup, showing relationships with wind direction and wind speed.
Figure 5. Nonparametric regression results for (a) PM2.5 (n = 2930) and (b) PM10-2.5 (n = 2930) mass concentrations at Maplewood, showing relationships with wind direction and wind speed.
Figure 6. Nonparametric regression results for (a) PM2.5 (n = 2262) and (b) PM10-2.5 (n = 2262) mass concentrations at McAuliffe, showing relationships with wind direction and wind speed.