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Characterization of Residential Woodsmoke $PM_{2.5}$ in the Adirondacks of New York

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ABSTRACT

Although woodsmoke from residential wood heating can be the dominant source of winter $PM_{2.5}$ in rural areas, routine monitoring is done primarily in urban or suburban areas. To obtain data on elevated woodsmoke concentrations from nearby sources, the $PM_{2.5}$, black carbon at 880 and 370 nm, particle-bound polycyclic aromatic hydrocarbons (PAHs), and wind speed and direction were measured during winter at three residential locations in Saranac Lake, New York. A paired-site design enabled the identification of local sources relative to larger spatial scales. With the exception of occasional regional PM events, the hourly measurements of this pollutant between the paired sites exhibited poor correlations, suggesting that local woodsmoke was responsible for the observed increases in PM values. One location that was adjacent to a residence with a wood stove, which was 40 meters from the monitoring site, experienced repeated episodes of elevated $PM_{2.5}$ concentrations, with a maximum 3-hour average of 150 µg m^{-3}, a maximum 24-hour rolling average of 64 µg m^{-3}, and a maximum midnight-to-midnight average of 46 µg m^{-3}. Despite these PM events, the data indicated that this location was likely in compliance with the current U.S. EPA National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$. The daily $PM_{2.5}$ concentration peaked and troughed during the nighttime and the daytime, respectively, at all of the sites, which is consistent with local ground-level pollution sources, such as woodsmoke; this diel pattern was also confirmed by Aaethalometer *Delta-C* (DC) data, a woodsmoke PM indicator. The particle-bound PAH data was less specific than the DC data to the PM in the woodsmoke, partly because the instrument for the former also responds to traffic pollution. One site repeatedly displayed the influence of 2-cycle engine snowmobile exhaust during the early evening hours, with very high PAH concentrations but only modestly elevated DC concentrations. Subsequent tests showed that fresh 2-cycle small engine exhaust produces a somewhat weaker response than woodsmoke in the DC in terms of the concentration per unit of PM.

Keywords: Biomass burning; Black carbon; Carbonaceous aerosols; Optical properties; Polycyclic aromatic hydrocarbon.

INTRODUCTION

Woodsmoke from residential wood heating contains a large number of toxic compounds (Zelikoff *et al.*, 2002; Danielsen *et al.*, 2011; Bølling *et al.*, 2012) and accounts for 15% of U.S. PM emissions, which is five times more than U.S. petroleum refineries, cement manufactures, and pulp and paper mills combined (U.S. EPA, 2018a). In rural areas, PM emissions from residential wood heating often dominate emissions from all other source sectors combined (EPA, 2018a), can account for 50% of total area source air toxics cancer risk (EPA, 2018b), and may be trending upwards in rural New York (Masiol *et al.*, 2018; Blanchard *et al.*, 2019). Woodsmoke PM tends to be higher when temperatures are colder as indicated by heating degree days (Zhang *et al.*, 2017) due to the increased use of residential wood heating appliances. Exposure to woodsmoke has been shown to have a range of adverse health effects (Naeher *et al.*, 2007; Weichenthal *et al.*, 2017). Elevated levels of PM well below the current U.S. Environmental Protection Agency (EPA) National Ambient Air Quality Standards (NAAQS) of 35 µg m^{-3} daily average and 12 µg m^{-3} annual average have been associated with increased mortality (Schwartz *et al.*, 2015; Schwartz *et al.*, 2016; Achilleos *et al.*, 2017; Di *et al.*, 2017a, b; Maker *et al.*, 2017; Vodonos *et al.*, 2018). As part of the EPA 2015 revisions to the New Source Performance Standards for New Residential Wood Heaters (2015 NSPS), PM emission standards for new residential wood heating devices were recently made more stringent to reduce exposure to woodsmoke. While this regulation may reduce emissions and exposure from new installations of wood burning appliances, it does not apply to existing appliances. The majority of residential appliances in use today have either older control technologies or are uncontrolled, such as pre-1988 NSPS stoves and indoor or outdoor wood furnaces installed before 2015 (Congressional Research Service, 2018).

Routine monitoring of $PM_{2.5}$ for determining compliance with the National Ambient Air Quality Standards for $PM_{2.5}$

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(PM-NAAQS) is done by state and local air agencies. These monitoring sites are usually located in urban areas or near large industrial point sources, and do not characterize the potential for elevated woodsmoke PM exposures from nearby individual residential wood heating devices in small rural towns. Several studies have measured ambient PM concentrations in residential areas using fixed monitoring sites or mobile monitoring platforms (Glasius *et al.*, 2006; Larson *et al.*, 2007; Robinson *et al.*, 2007; Hellén *et al.*, 2008; Jeong *et al.*, 2008; Krecl *et al.*, 2008; Bari *et al.*, 2010; Allen *et al.*, 2011; Bari *et al.*, 2011; Wang *et al.*, 2011; Grange *et al.*, 2013; Loeppky *et al.*, 2013; Thatcher *et al.*, 2014; Su *et al.*, 2015; Ranasinghe *et al.*, 2016). Most of these studies characterized woodsmoke PM in the local airshed but were not designed to characterize PM from individual nearby woodsmoke plumes. One study explicitly chose sites to avoid large impacts by a single source (Yli-Tuomi *et al.*, 2015). Some of these studies used integrated filter PM sampling, and some had hourly or shorter average PM measurements. Time-resolved (on-line or continuous) measurements are useful in identification of specific sources; Snyder (2012) and Zhang (2017) measured PM in individual plumes using forward light scattering methods on mobile monitoring platforms.

A study by the New York State Department of Health (NYS-DOH, 2013) was specifically designed to identify and assess the impact of large individual PM sources (outdoor wood boilers) using a paired-site design, with one site near the source of interest and a second site to measure background PM that had little or no influence from local PM sources. The work presented here uses the NYS-DOH paired-site concept of characterizing nearby (micro- to mid-scale) sources separately from an elevated background on the neighborhood scale, but only requires a "background" site to be reasonably free (relative to the near-source site) from large and very local PM sources instead of being uninfluenced by local sources. This allows the background site to be located in the same airshed as the near-source site so that airshed-wide influences of local woodsmoke (e.g., a valley entrapping woodsmoke overnight) are properly accounted for and not attributed solely to the source of interest.

Previous measurements and modeling of woodsmoke in several Adirondack communities in northeast New York observed strong diurnal variations and large differences in woodsmoke within villages compared to locations just outside of the same villages (Allen *et al.*, 2011; Su *et al.*, 2013). This study facilitates an improved understanding of the spatial and temporal variation of woodsmoke concentrations in a rural valley community, as well as the range of exposures to woodsmoke PM in these valleys where routine PM monitoring is not being done. These data may serve as a baseline for comparison as old technologies are switched out with new, more efficient and less polluting wood heating appliances. This study was designed specifically to characterize woodsmoke PM concentrations in various settings, especially in areas that are affected by relatively close sources as well as "valley background" concentrations (i.e., elevated concentrations that are not strongly influenced by large nearby sources). This information may be used to inform air planning and public health efforts or new initiatives such as woodstove changeout programs.

METHODS

Both mobile platform and fixed-site $PM_{2.5}$ monitoring were performed during the winters of 2013–2014 and 2014–2015 in Saranac Lake, NY, a village in the Adirondacks 400 km north of New York City with a population of approximately 5,000 and no significant local industrial or traffic $PM_{2.5}$ sources. While not in a traditional valley, the town is surrounded by hills on several sides. Fig. 1 shows the topography of the town and the approximate location of the three fixed monitoring sites.

For the first winter, PM and wind speed and direction were only measured at one site (A). For the second winter, PM, wind, BC, and particle-bound PAH were measured at Site A for the full winter, and at Sites B and C for six or seven weeks each. PM monitoring using a mobile platform was performed during both winters in the evening on nights when forecasted wind speed and ambient temperature were low and would allow local woodsmoke to accumulate. Mobile platform results are presented here only for the first winter when these data were used to identify possible locations for fixed-site woodsmoke measurements.

$PM_{2.5}$ was measured for both the mobile platform and fixed-site monitoring with a Thermo Scientific (Franklin, MA) pDR-1500 monitor that uses 70° forward light scattering at 880 nm as a highly time-resolved surrogate for PM (Wang *et al.*, 2016; Zhang *et al.*, 2018).

A BGI (Waltham, MA) SCC0.732 cyclone was used for the pDR-1500 inlet with a sample flow of 1.0 L min^{-1}, giving a D_{50} size cut of 2.5 μm. The instrument's relative humidity correction was turned off, since the f(RH) correction curve it uses is based on hygroscopic aerosol such as sulfate and is not necessarily appropriate for woodsmoke PM (Martin *et al.*, 2013). The data recording interval was 1 second for the mobile platform and 1 minute for fixed-site monitoring.

The pDR-1500 has been shown to agree well with Federal Equivalent Method (FEM) monitors when sampling ambient PM that is predominantly woodsmoke from residential space heating. A Met One (Grants Pass, OR) BAM 1020 $PM_{2.5}$ FEM for $PM_{2.5}$ and a pDR-1500 run in the same configuration as used in this study were collocated at the Vermont Department of Environmental Conservation's central monitoring site in Rutland, VT, for the winter of 2011–2012. Elevated levels of $PM_{2.5}$ at this site during the winter are primarily from residential wood heating appliances (Allen *et al.*, 2004). Agreement was very good over a wide range of $PM_{2.5}$ concentrations (Zhang *et al.*, 2017), with a slope of 1.08 and R^2 of 0.90 for 24-hour average values.

For the mobile platform, the PM inlet probe was positioned towards the front of the car 30 cm above the car roof. For fixed-site outdoor monitoring, the pDR-1500 was inside a heated enclosure (approximately 10℃ above ambient temperature), with the PM inlet 2 meters above the ground. Wind speed, wind direction, and sonic temperature were measured every 10 seconds at fixed sites using a Gill (Lymington, Hampshire, UK) heated WindObserver II sonic anemometer. Sonic temperature was used as a quality

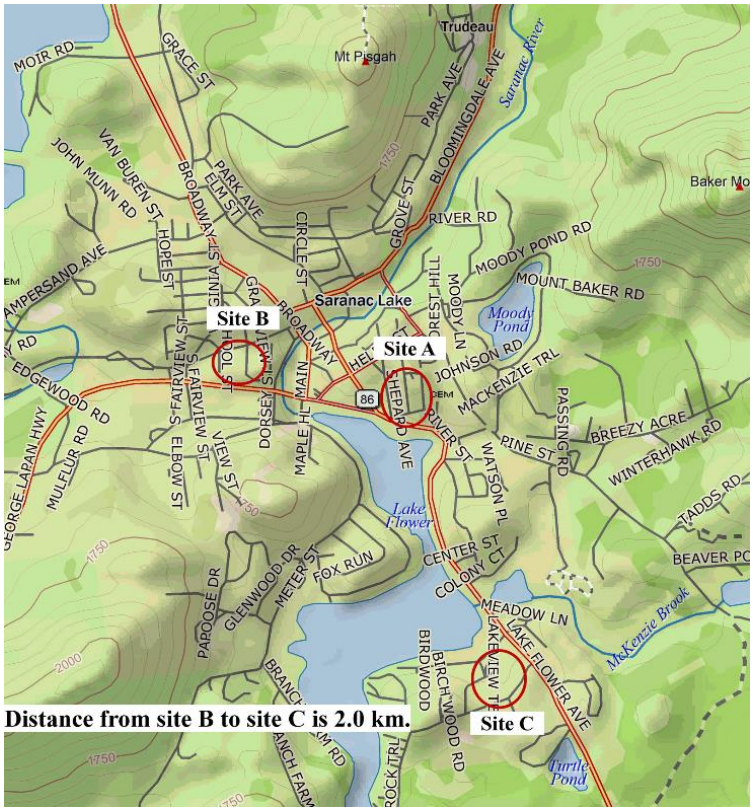

Fig. 1. Saranac Lake, NY; topography; and monitoring site locations.

control check for the sonic wind data. 10-second raw wind measurements were processed up to 5-minute and 1-hour values for scalar wind speed and resultant wind vectors.

In addition to $PM_{2.5}$ and wind speed and direction, real-time fixed-site measurements for the second year included BC and PAHs. A 2-channel Aethalometer® Model AE-42 (Magee Scientific, Berkeley, CA) sampling at 2 L min^{-1} with a $PM_{1.0}$ inlet cyclone (BGI SCC0.732) was used to measure black carbon (BC) at 880 and 370 nm at 1-minute intervals. Aethalometer data were corrected for filter loading artifacts using the binned approach described by Park (2010) and based on a method described by Virkkula (2007). The concentration difference between these two channels is called *Delta-C* (DC). This derived parameter can be used as a semi-quantitative measurement of the organic carbon (OC) component of woodsmoke PM, which dominates primary PM emissions from conventional cordwood stoves (Bertrand *et al.*, 2017). DC is used here to demonstrate that elevated levels of $PM_{2.5}$ were predominantly from woodsmoke (Allen *et al.*, 2004; Wang *et al.*, 2012; Olson *et al.*, 2015; Zhang *et al.*, 2017). In locations where woodsmoke dominates local $PM_{2.5}$ concentrations, DC has been shown to be well correlated with levoglucosan, an organic molecular marker for woodsmoke PM (Hedberg *et al.*, 2006; Wang *et al.*, 2011). Fig. 2 shows a robust comparison between 24-hour average DC and levoglucosan measured by the Monterey Bay Air Resource District (MBARD) in the San Lorenzo Valley, CA, during the winter of 2014–2015 (data courtesy of MBARD). This result demonstrates that DC is as good a woodsmoke marker as levoglucosan in areas that are dominated by woodsmoke. It has the advantage of being highly time-resolved, which allows additional analysis such as time-of-day patterns that can identify woodsmoke PM. It should be noted that while levoglucosan is very specific to PM from wood combustion, there can be situations where

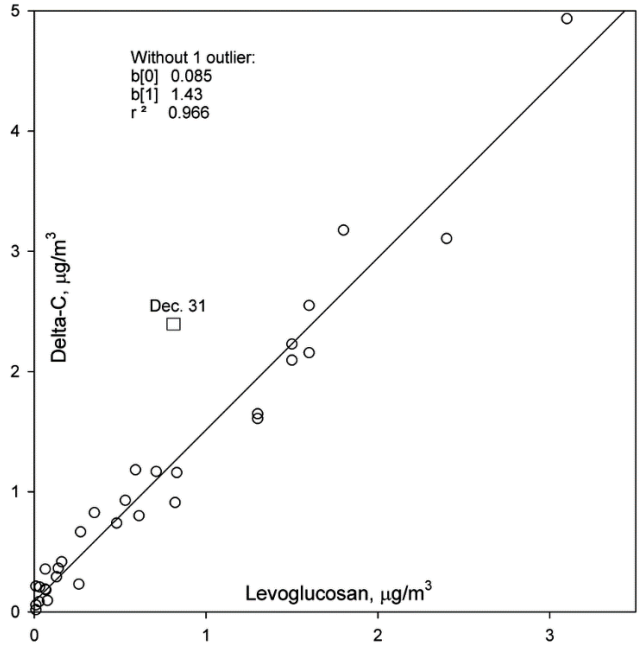

Fig. 2. DC vs. levoglucosan in the San Lorenzo Valley, CA, winter of 2014–2015.

other combustion sources of OC aerosol can contribute to the DC signal. Thus caution should be used when using DC as a woodsmoke indicator when there may be other large local sources of OC aerosol present. Domestic coal burning for heating has been shown to have levoglucosan and light-absorbing "brown carbon" or BrC (Li *et al.*, 2018; Yan *et al.*, 2018) but it is unlikely that coal is used much for domestic space heating in New York. In this paper we show that exhaust from 2-cycle snowmobile engines also contains BrC as measured by the Aethalometer DC.

Continuous particle-bound PAHs were measured at 1-minute intervals with EcoChem (League City, TX) PAS-2000 analyzers with $PM_{1.0}$ inlets at the fixed sites to investigate if this measurement was of use in identifying woodsmoke PM. This method is a semi-quantitative measurement of total particle-bound PAHs (Wilson *et al.*, 1995), since the response depends on the kinds of PAHs present in the sample (Kelly *et al.*, 2003). Ultraviolet light from a 222-nm excimer lamp ionizes particle-bound PAH molecules, and an electric field then removes the free electrons. The resulting positively charged particles are collected on a filter, generating a current that is measured by an electrometer. The PAH monitors were overhauled and calibrated by the manufacturer prior to use in this study. The instrument output is in femtoamperes (fA), and although this can be approximately scaled to mass of PAHs, fA is used as the measurement unit here to avoid confusion with quantitative measurements of PAHs. 1 fA is approximately 1–3 ng m^{-3} PAH mass (Wilson *et al.*, 1995).

RESULTS

The mobile platform monitoring during the first winter showed multiple sites with elevated $PM_{2.5}$ above the local background $PM_{2.5}$ on many trips. An example of a typical evening's measurements in Saranac Lake is shown in Fig. 3 for January 9, 2014. The three fixed sites are noted, as are measurements made at the top of Mt. Pisgah, a 100-meter (above local terrain) hill just to the north of downtown Saranac Lake that is used as a background PM measurement away from local woodsmoke sources. $PM_{2.5}$ in town varied over a wide range over short distances and peaked at over 400 µg m^{-3} (5-second average). Such high concentrations in this rural area are indicative of local woodsmoke sources. Background $PM_{2.5}$ (at Mt. Pisgah, on the north edge of the site map) was very low—less than 5 µg m^{-3}.

First Winter Fixed-site Monitoring

During the first winter, a single fixed site (A) was deployed based on preliminary mobile platform results. Site A is located near the center of town in the backyard of a single-family residence in a residential neighborhood. The residence used a pellet stove, but the monitor was sited away from the

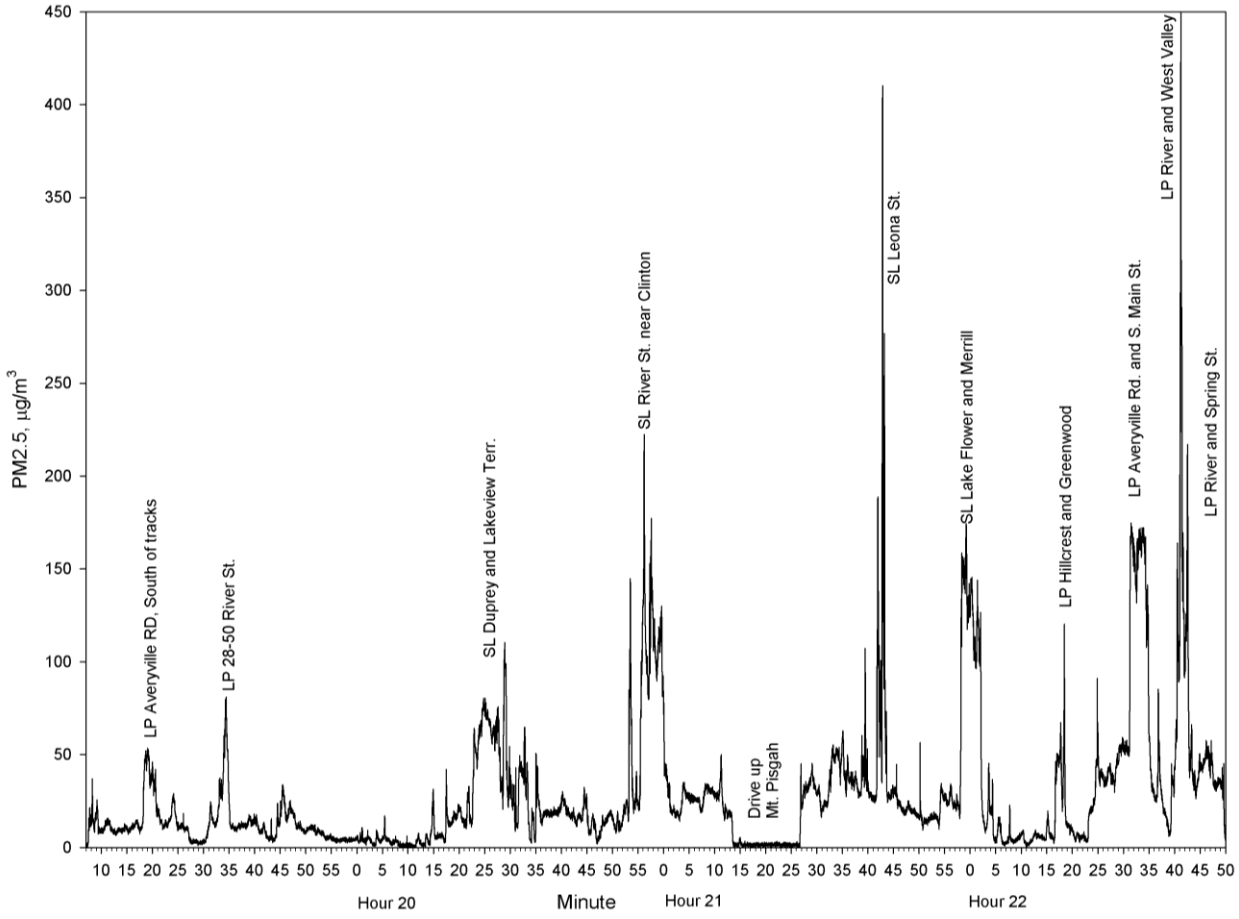

Fig. 3. 1-second mobile platform $PM_{2.5}$ in Saranac Lake, January 9, 2014.

house by approximately 20 meters to the southeast to avoid being routinely impacted by that local woodsmoke source. $PM_{2.5}$ was monitored from January 28 to May 7, 2014.

Regional PM events were identified from local woodsmoke events by the daily temporal pattern of PM. Local woodsmoke has a strong diel pattern with peaks overnight and mid-day minimums (Jones *et al.*, 2011). Regional PM events do not show these diel patterns. While there were many short-term (sub-daily duration) periods of elevated PM during this period, the running 24-hour PM average from several regional PM events was higher than most 24-hour average peaks from local woodsmoke, because woodsmoke PM is typically elevated for only about half the day (overnight), while regional PM can be elevated for one to two days at all times of the day. Fig. 4 shows an example of a multi-day regional $PM_{2.5}$ event during mid-February 2014, with brief PM spikes from local woodsmoke superimposed on the regional $PM_{2.5}$ that varies very slowly.

The diel pattern of $PM_{2.5}$ at Site A during the first winter (not shown) had the typical pattern of local woodsmoke PM (as shown in Fig. 11 from the second winter), with the highest PM in the evening to overnight and in the morning between 6 and 8 a.m. A mid-day minimum of 3 µg m^{-3} occurred between 10 a.m. and 4 p.m. when atmospheric mixing and dispersion is greatest, and is an estimate of regional background $PM_{2.5}$.

The average $PM_{2.5}$ concentration from January 28 to March 31, 2014, at Site A was 6.2 µg m^{-3}. The median and 95th percentile values for 1-hour average $PM_{2.5}$ were 3.0 and 23 µg m^{-3} respectively.

Second Winter Fixed-site Monitoring

During the second winter, fixed-site monitoring was again performed at Site A for the entire winter. Since this site had more than 4,000 hours of PM data when both winters are combined, weekend PM was compared to weekday PM to detect potential differences resulting from occupants usually being at home more on weekends. A Mann-Whitney rank sum test on hourly average values indicated the weekend and weekday median PM (6.5 and 5.9 µg m^{-3} respectively) were not significantly different ($p = 0.07$). This suggests that space heating, not weekend recreational burning such as fireplaces, is the dominant use of wood burning. This is consistent with the lack of natural gas infrastructure in Saranac Lake, and in contrast to day-of-the-week patterns in Rochester, NY (Wang *et al.*, 2011), an urban area where residential wood combustion is more for recreational use than for domestic space heating due to the wide availability of natural gas. Fixed-site monitoring was also done at two additional sites (labeled *B* and *C* in Fig. 1) in Saranac Lake for at least six weeks each to provide information on spatial PM patterns and to characterize woodsmoke PM from sites near local sources. The residences at Sites B and C did not have any wood burning appliances. All three sites had the same instrumentation: pDR-1500 for $PM_{2.5}$, 2-channel Aethalometer for BC and DC, EcoChem PAS-2000 for particle-bound PAH, and WindObserver II sonic anemometer for wind speed and direction.

While no location in town was completely free from local woodsmoke impact, Site A was chosen to avoid large local sources. The two additional sites (B and C) were chosen for having significant local woodsmoke impacts based on mobile platform monitoring from the previous winter. Site B is 0.93 km WSW from Site A, and Site C is 1.5 km SSW of Site A. The distance between Sites B and C is 2.1 km. All three sites are in the same valley airshed and within the town limits.

Table 1 lists the dates that monitoring was performed for each of the second-winter sites.

Site B was adjacent to a residence with a wood stove 40 meters to the south-southeast. While there were other sources of woodsmoke in that neighborhood, winds were

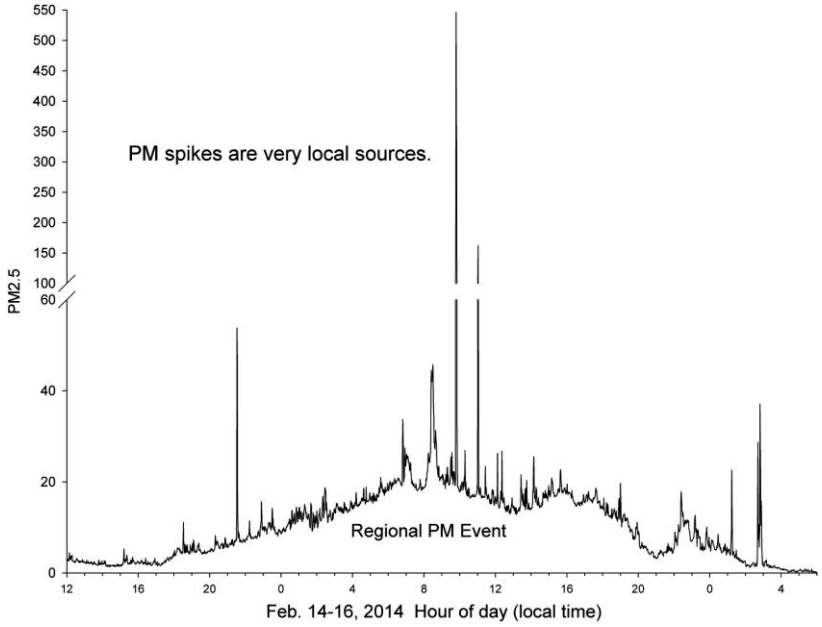

Fig. 4. Regional PM event, February 14–16, 2014.

Table 1. Monitoring periods for second winter sites.

Site Name	Start Date	Stop Date	Number of Days
A: full winter	16 Dec 2014*	13 Apr 2015	118 (104 to 31 Mar 2015)
B: first half	9 Dec 2014**	26 Jan 2015	48
C: second half	27 Jan 2015	12 Apr 2015	75 (63 to 31 Mar 2015)

* Site A wind data start: 22 Dec 2014; ** Site B wind data start: 16 Dec 2014.

predominately from the south to west, so this source dominated the measured PM at this site. Site C had several local sources of woodsmoke, but none that dominated the site as with Site B.

The distributions of 1-hour $PM_{2.5}$ for the two site pairs are shown in Fig. 5. While median concentrations are similar across all three sites, Site B had higher PM than the other two sites.

Note that although the average $PM_{2.5}$ for Site B exceeds the level of the annual NAAQS value of 12 µg m^{-3} for the 48 winter days when sampling was performed, it is likely that the annual average $PM_{2.5}$ at this site would be in compliance with the annual NAAQS since much of the year is free from woodsmoke influence. The 24-hour ("daily") PM concentration for comparison to the NAAQS is calculated on a midnight-to-midnight basis; this tends to reduce the average 24-hour concentration since it splits an overnight PM event across two calendar days. Although the highest 24-hour running average concentration was 64 µg m^{-3}, when using this midnight-to-midnight daily metric, there were just two 24-hour periods that exceeded the daily PM-NAAQS of 35 µg m^{-3} for this 48-day period (46 and 37 µg m^{-3}). Extrapolating that regulatory metric to the 4-month heating season of 120 days suggests that the site would experience five days with $PM_{2.5}$ greater than 35 µg m^{-3}. Since the form of the daily PM-NAAQS is the 98th percentile value, or seventh-highest for 365 days, this suggests that Site B would also likely be in compliance with the daily PM-NAAQS. Thus currently, neither the annual nor the daily $PM_{2.5}$ NAAQS provide protection from these kinds of seasonal episodic elevated PM concentrations.

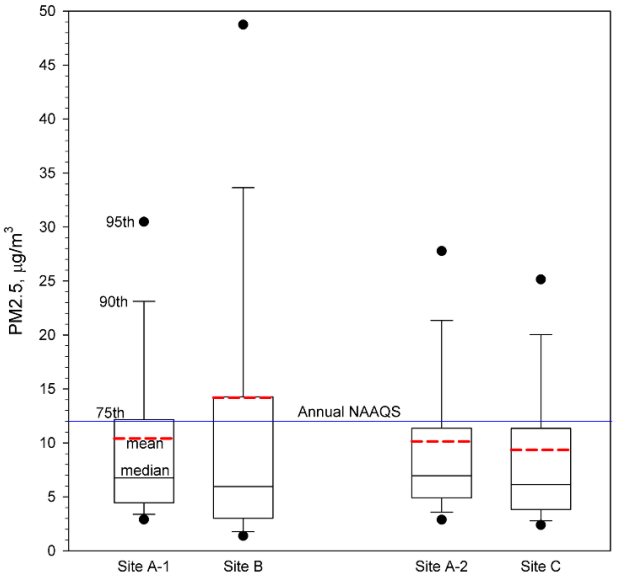

Fig. 5. Distributions of 1-hour $PM_{2.5}$ for two site pairs.

The maximum 1-hour and 3-hour $PM_{2.5}$ concentrations at Site B were 400 and 150 µg m^{-3}, respectively. Fig. 6 shows the 3-hour running average $PM_{2.5}$ at Site B, with EPA Air Quality Index (AQI) breakpoints for the 2012 $PM_{2.5}$ NAAQS indicated by colored lines. While the 24-hour PM standard is 35 µg m^{-3}, EPA uses the Nowcast method for real-time AQI reporting (U.S. EPA, 2018c). When hourly average $PM_{2.5}$ levels are high and changing rapidly, the real-time PM AQI is based on a 3-hour running average, and concentrations greater than 150 µg m^{-3} are reported as "very unhealthy" (purple AQI color).

Hourly $PM_{2.5}$ for Site B vs. Site A (N = 981, R^2 = 0.10) and Site C vs. Site A (N = 1402, R^2 = 0.33) during the second winter of monitoring is not well correlated. While there are some hours below 20–25 µg m^{-3} where PM for site pairs is similar (due to regional transport), there are relatively few hours where both sites are elevated. This decoupling at high PM concentrations demonstrates that very local PM sources dominate when PM is elevated. Since Site C does not have the large impact (next door, upwind) of a single local source that Site B has, the divergence at elevated PM levels is not as large at Site C as Site B. The influence of more spatially uniform concentrations is greater on the neighborhood scale (1–4 km) where PM is elevated due to regional transport, but not from a very local specific source or a regional event. This results in a higher R^2 for Site C vs. Site A. Fig. 7 shows a 3-week interval in March 2015, where there are four overnight or early morning cases where PM measurements from Sites A and C track very closely. These cases are different from the regional PM event scenarios since the woodsmoke PM source is local (not regional transport) and peaks only during overnight or morning periods when wind speed is low and the valley entraps local woodsmoke. These examples also demonstrate that the PM measurements between these two sites are closely matched.

Aethalometer DC Woodsmoke Indicator

Aethalometer DC, a semi-quantitative measurement of woodsmoke PM, was used in this study to confirm that elevated levels of $PM_{2.5}$ were primarily from woodsmoke. A useful approach to comparing DC and $PM_{2.5}$ is shown in Fig. 8. Regression of the 24 hourly diel data pairs (PM vs. DC) shows strong correlation and a slope of 16 for Sites A and C. The slope for Site B is 25 (indicating less DC per unit PM) and correlation is not as strong, possibly because of the extremely high PM concentrations from a single wood stove where PM emissions are likely dominated by organic carbon (poor burn conditions that result in high PM are mostly

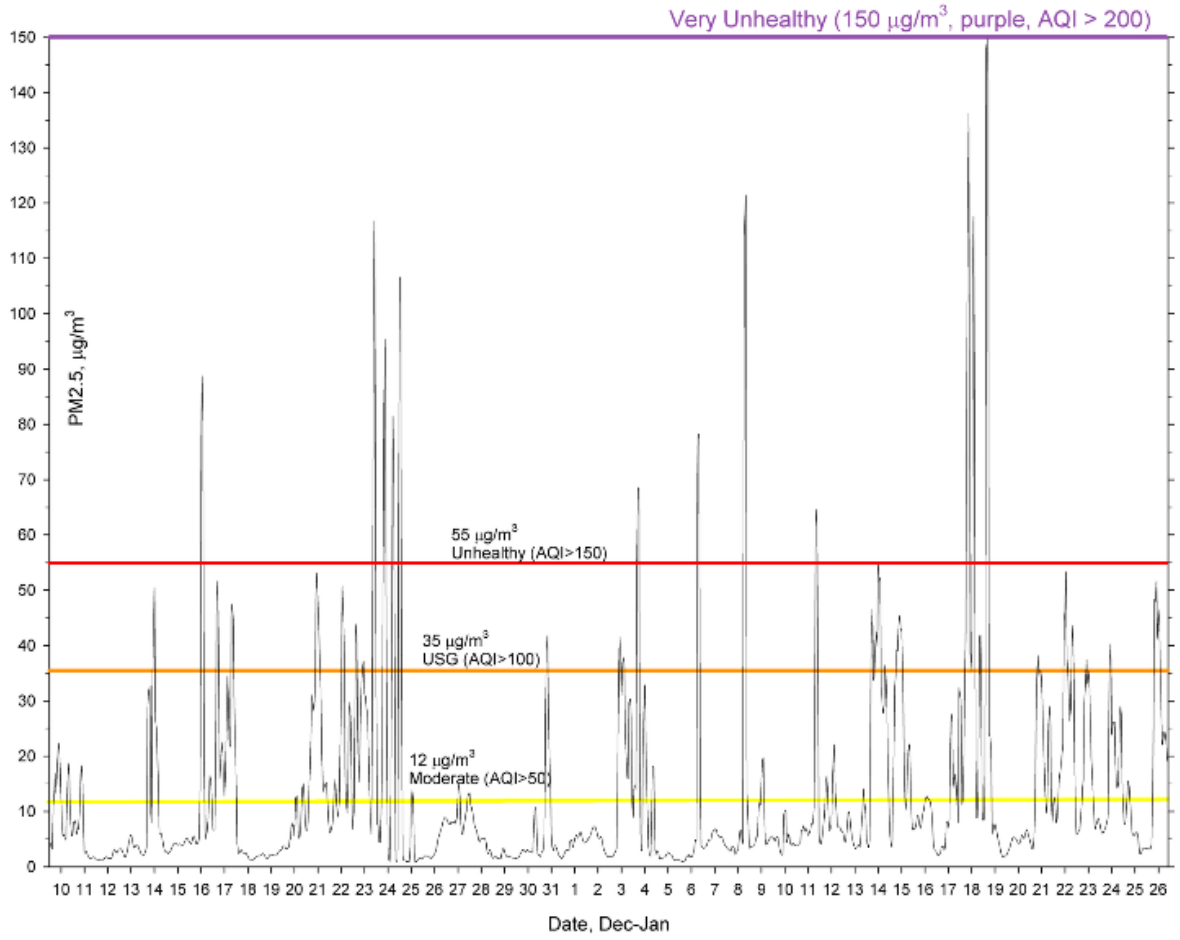

Fig. 6. Running 3-hour average $PM_{2.5}$ at Site B. Colored lines indicate EPA AQI breakpoints for the 3-hour Nowcast.

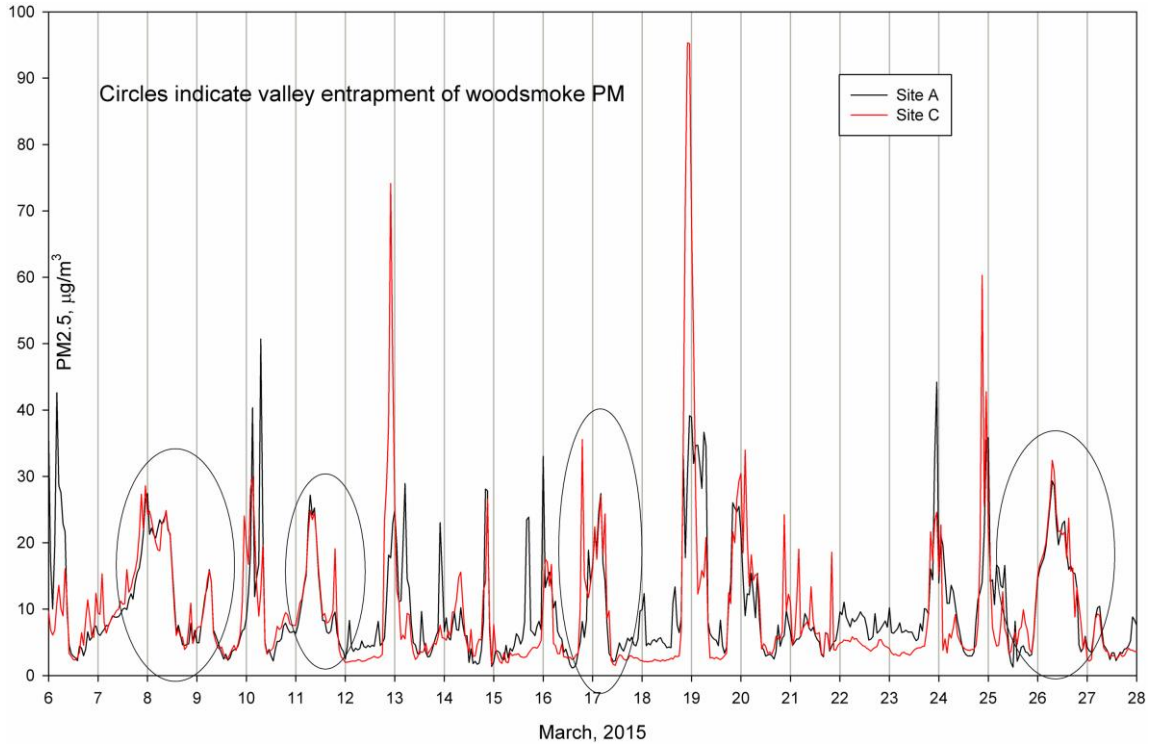

Fig. 7. 1-hour $PM_{2.5}$ for Sites A and C, March 6–28, 2015.

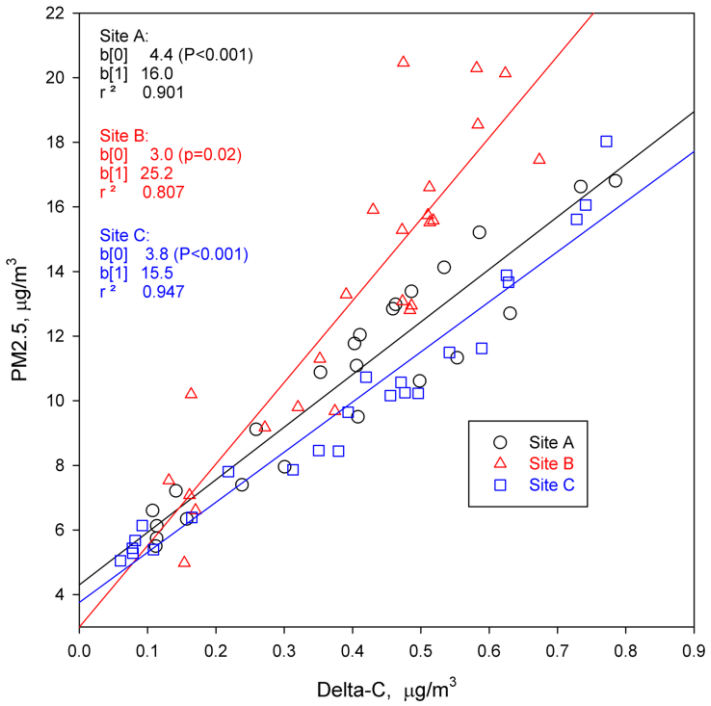

Fig. 8. Regression of diel data pairs from Sites A, B, and C during the second winter of monitoring.

organic carbon) instead of a mix of various woodsmoke sources that would be expected at most monitoring locations. The intercept for all three sites is between 3.0 and 4.4 µg m^{-3}, and represents the average PM concentration when there is no local contribution from woodsmoke.

Non-parametric Wind Regression Analysis for PM and DC

Since Aethalometer DC is a reasonably specific marker for woodsmoke in this rural winter case, it can be used to directionally identify sources using 1-dimensional (wind direction only) non-parametric wind regression (NPWR) analysis (Henry *et al.*, 2002; Henry *et al.*, 2009). A scalar wind speed threshold of 0.45 m s^{-1} and a smoothing window of 8° was used. 2-dimensional (wind speed and direction) NPWR was not used due to the relatively small data set available for the analysis. The additional utility of 2-dimensional wind regression may include characterization of the distance to local sources (Henry *et al.*, 2002) and distinguishing elevated sources from those close to ground level (Henry *et al.*, 2002; Yu *et al.*, 2004); these were not of primary interest for this analysis.

The NPWR approach to identifying the direction of major local sources of $PM_{2.5}$ was validated using data from Site B, where a wood stove was next door, 40 meters to the south of the monitoring site. Site B had 984 hours with both wind (speed and direction) and PM data; 5-minute values were used to obtain a larger sample size (and thus smaller uncertainty estimates) in this analysis. Fig. 9(a) shows PM, BC, and DC NPWR analysis for Site B, as well as a wind rose for these data. Note that there were very few hours with winds from NW through N to SE, and thus NPWR results for those directions have very large uncertainties. The large next-door source shows a sharp peak at 160° in both $PM_{2.5}$ and DC, but not for BC. This would be consistent with a source of poor combustion where most of the PM is organic carbon. There is a suggestion of a small peak at ~212°, which is the direction of a commercial wood pellet boiler 150 meters away, used to heat an elementary school. There are suggestions of other sources in directions with very few hours of data, and thus large uncertainties. An example of bootstrapped uncertainty estimates from this regression is shown in Fig. 9(b).

Similar analysis was conducted for DC at Sites A and C, where local sources were not identified. Fig. 10 shows that for Site A there may have been a source to the east and southeast, but the uncertainty related to those directions is large since the wind did not come from that direction very often. For Site C, there does not appear to be any distinct source influencing PM at this site, consistent with a number of smaller woodsmoke sources at that location.

Particle-bound PAHs from 2-Cycle Snowmobile Engines

The particle-bound PAH data were not well correlated with PM or DC, and thus are not useful as a woodsmoke indicator. The diel pattern for PAHs at Site C showed an unusual temporal pattern that is not typical of woodsmoke or traffic PM emissions. Fig. 11 shows the diel pattern at this site for PM, BC, DC, and PAHs from January 27 to March 31, 2015, and a scatter plot of PAHs vs. DC for the diel values where the highest PAH concentrations have a much higher PAH-to-DC ratio. Examination of the raw data showed frequent elevated PM, BC, DC, and PAHs in the early evening, typically between 6 p.m. to 9 p.m. The Site C homeowner was queried about this pattern and reported that there were snowmobiles frequently used on an adjacent lot

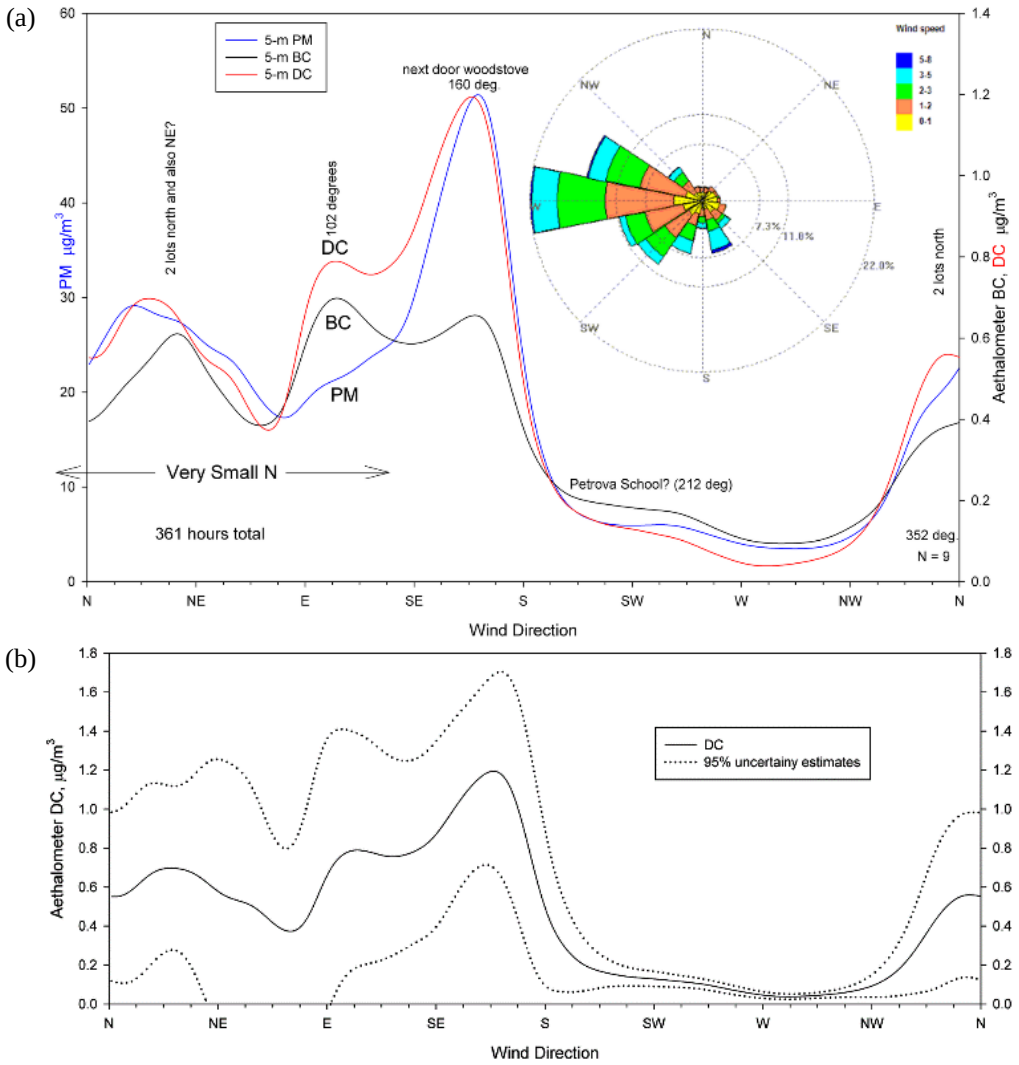

Fig. 9. (a) PM, BC, and DC non-parametric wind regression analysis for Site B, December 16, 2014–January 26, 2015. (b) Non-parametric wind regression analysis 95% bootstrap confidence intervals for DC at Site B, December 16, 2014–January 26, 2015.

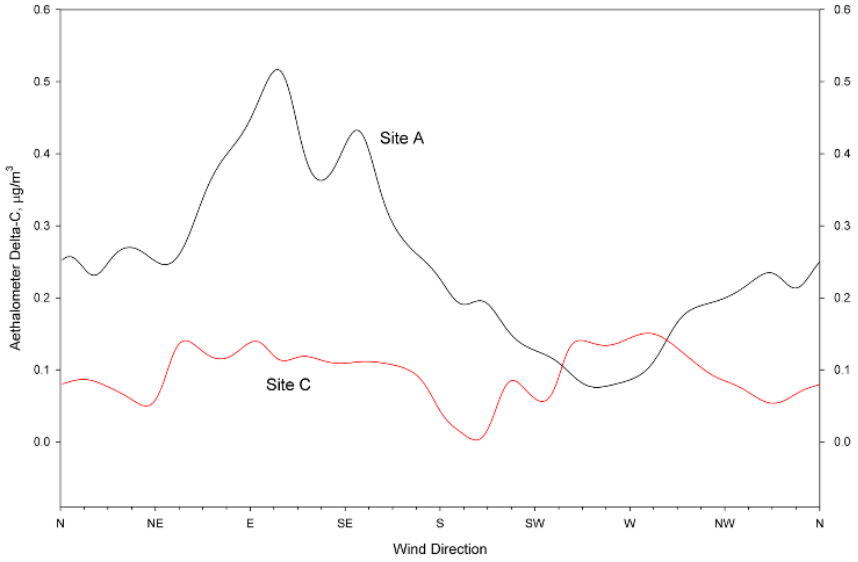

Fig. 10. DC non-parametric wind regression analysis for Sites A and C.

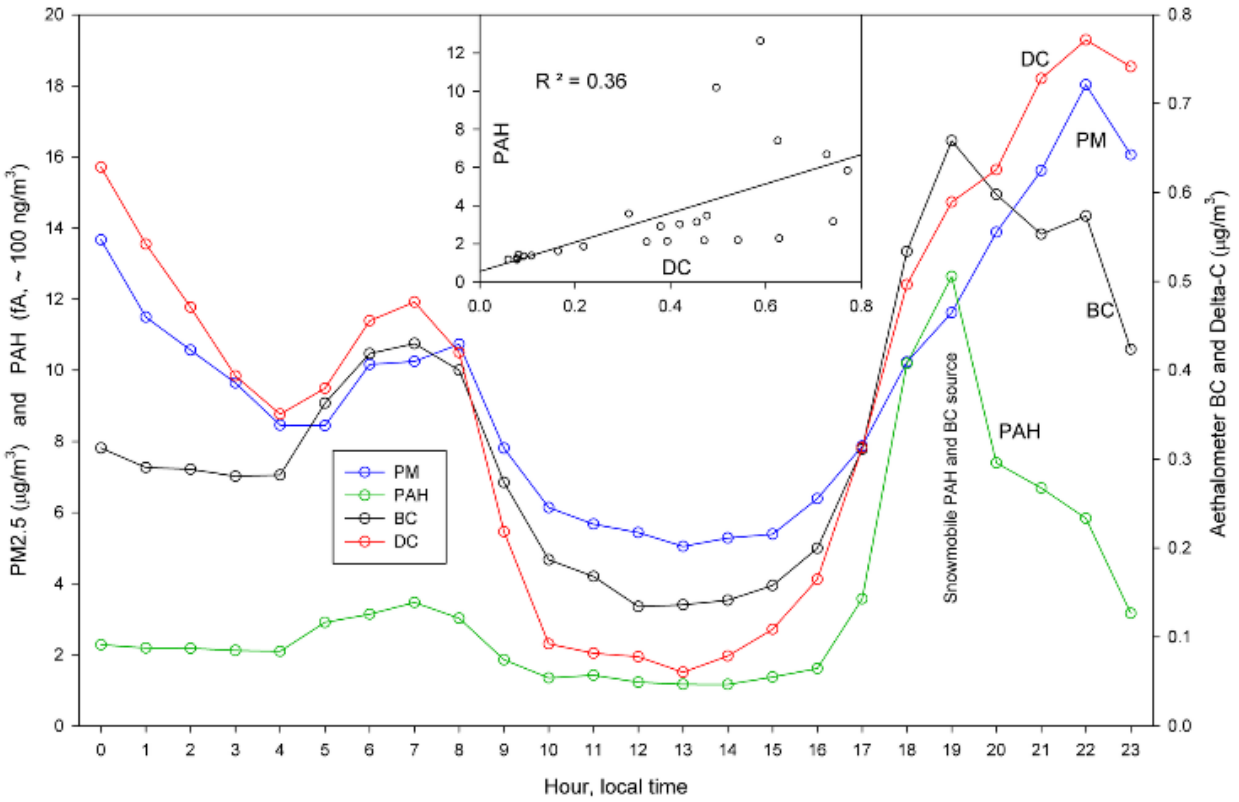

Fig. 11. Influence of snowmobile engine exhaust on PAHs and PM at Site C, and PAHs vs. DC scatter plot.

at that time of day. The snowmobile-emitted PAH and BC peak is very evident for the three hours from 6 p.m. to 9 p.m., superimposed on the expected woodsmoke diel pattern for DC and PM with minimums mid-day and peaks in the morning (at 7 a.m.) and late evening.

DC is a measure of enhanced optical absorption at shorter wavelengths, a property usually associated primarily with biomass combustion as noted above. These results suggest that particle-bound PAHs from incomplete combustion of fossil fuels can have similar optical properties. To better characterize this response, a controlled test was performed to measure PM, BC, and DC for smoke from a small 2-cycle engine. A Thermo Scientific Model TEOM 1400ab and a Magee Scientific Aethalometer AE33 were used to measure the exhaust in an enclosed space from a pair of hand-held hedge trimmers that had been used by a local landscaping company. The engine was run for a few minutes until a concentration of approximately 1,000 µg m^{-3} of PM was reached, at which point the engines were turned off. Measurements continued for 55 minutes as infiltration, volatilization, and other particle losses reduced concentrations to ~20% of the peak. Fig. 12 shows PM, BC, and DC from this test. The PM-to-DC ratio for the 2-cycle engine exhaust testing is approximately 20, compared to a ratio of 15–25 for PM to DC for woodsmoke in Saranac Lake. While these ratios are similar, it was the PAH data that suggested that some combustion particle source other than woodsmoke was routinely impacting this site for three hours from 6 to 9 p.m., activity that was confirmed by contemporaneous conversations with the property owner for this site.

There are some limitations of this controlled 2-cycle small engine experiment to quantitatively measure the PM-to-DC ratio from this source. These measurements were performed with much higher engine exhaust PM concentrations compared to the ambient measurements, and at a much higher temperature (+15℃ compared to ~–17℃ during the hours of interest). These competing differences in temperature and concentration, and thus vapor pressure, will affect the gas/particle partitioning of volatile organic compound (VOC) species and make a quantitative assessment of PM-to-DC ratio from this controlled testing difficult. In addition, other particle species present (wood smoke or regional PM) could provide condensation sites for the fresh semi-volatile organic compounds (SVOCs) from the snowmobiles.

CONCLUSIONS

Large spatial gradients in $PM_{2.5}$ exist within the town of Saranac Lake in the Adirondacks in upstate New York. These gradients were characterized using a mobile monitoring platform, and three areas with elevated $PM_{2.5}$ concentrations were subsequently selected for fixed-site monitoring of PM in woodsmoke. A paired-site monitoring approach demonstrated the influence of highly localized (micro- to mid-scale) sources of woodsmoke PM on neighborhood-scale concentrations. The time of the day and event duration patterns differentiated transported PM (associated with regional events) from local emissions.

Short-term PM concentrations from woodsmoke were observed that may pose risks to human health. For example,

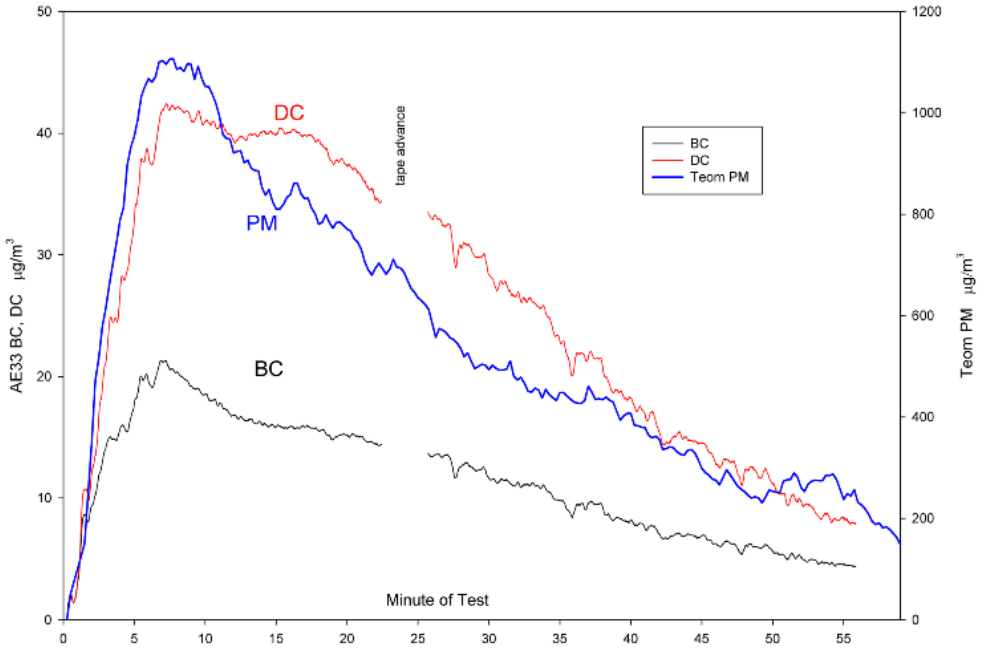

Fig. 12. PM, BC, and DC from 2-cycle engine exhaust.

one monitoring site that was situated next to a house with a wood stove repeatedly experienced periods of elevated PM, with the 1-hour and rolling 24-hour average concentrations reaching 400 and 64 µg m^{-3}, respectively. Despite these high concentrations, we estimated that the PM levels at this site were in compliance with both the current daily and annual $PM_{2.5}$-NAAQS. Based on the Aethalometer DC data, a biomass combustion indicator that displays good correlations with elevated levels of $PM_{2.5}$, we estimated a non-woodsmoke winter background PM concentration of 3–4 µg m^{-3}. Although the particle-bound PAH measurements were a poor indicator of the woodsmoke PM, at one of the sites they frequently exhibited distinct increases during the early evening because of the contribution from 2-cycle engine snowmobile exhaust. Controlled testing revealed that the PM-to-DC ratios per unit of PM for this exhaust were similar to those of fresh woodsmoke, suggesting that caution should be exercised when basing source apportionment of biomass burning PM on optical measurements of brown carbon in urban areas where motorized scooters with 2-cycle engines may be commonly used for intra-urban transportation.

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