



Characterization of Valley Winter Woodsmoke Concentrations in Northern NY Using Highly Time-Resolved Measurements

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ABSTRACT

The increasing popularity of wood fired heating appliances in cold winter climates has focused attention on assessment of woodsmoke exposures. Pollution from residential wood combustion (RWC) is a major concern in areas with valley topography where nighttime inversions limit the dispersion of pollutants from ground-level sources. An intensive characterization of ambient particulate matter (PM) from RWC was performed in northern New York State during winter 2008–2009 in an area where the 2005 U.S. EPA National Emissions Inventory shows RWC to be the largest source of PM_{2.5}. Measurements of woodsmoke PM were made using optical scattering and absorption techniques during repeated night-time mobile monitoring to provide data with high spatial and temporal resolution; measurements were also made at six fixed sites for the study period to provide temporal context for the mobile measurements. The difference in optical absorption at near-infrared and near-ultraviolet wavelengths was used as a specific marker for woodsmoke PM. Woodsmoke was the only significant contributor to elevated night-time valley PM concentrations during mobile run nights; short-term (3 minute) PM concentrations frequently exceeded 100 µg/m³. Concentrations observed with mobile monitoring were consistently elevated at valley bottoms where the majority of the population lives, and approached zero outside of valleys. Data from fixed sites indicated that woodsmoke levels peaked near midnight, with a secondary peak around 7 AM and a mid-day minimum. These patterns are consistent with RWC use and diurnal patterns of atmospheric dispersion.

Keywords: Aethalometer; Nephelometer; Spatial variability; Mobile measurements; Outdoor wood boilers, Woodsmoke.

INTRODUCTION

There is ample evidence that residential wood combustion (RWC) emits significant quantities of pollutants that are known to impact health, including particulate matter, carbon monoxide, nitrogen oxides, and a number of known human carcinogens, including benzene and polycyclic aromatic hydrocarbons (Naeher *et al.*, 2007). According to the U.S. Environmental Protection Agency (U.S. EPA), exposure to fine particulate matter (particles with an aerodynamic diameter equal to or less than 2.5 micrometer, i.e., PM_{2.5}) from woodsmoke is a major health threat (U.S. EPA, 2009).

There is a large body of evidence showing cardiovascular and respiratory health effects associated with ambient PM_{2.5} concentrations (Brook *et al.*, 2010). The U.S. EPA has set

national PM_{2.5} ambient air quality standards for annual and daily (24 hour) averaging times, but sub-daily elevated periods of PM_{2.5} can also be of concern. Recent work has identified adverse respiratory impacts specifically from sub-daily (4 hour) exposures to woodsmoke (Barregard *et al.*, 2006; Barregard *et al.*, 2008; Danielsen *et al.*, 2008). Further, wood burning appliances are typically operated in residential areas so that a large fraction of emissions can result in concentrations to which people are actually exposed (Ries *et al.*, 2009).

A key challenge in the assessment of woodsmoke conditions in rural areas is the location of wood burning sources, compounded by landscape features such as valleys. These features can create significant PM_{2.5} spatial variability, including "hotspots" of elevated concentrations on top of regional PM_{2.5} contributions. Regulatory ambient air pollution monitoring networks are typically not dense enough, particularly outside of major urban areas, to effectively characterize this spatial variability.

For example, in the Adirondacks region of New York State (the focus of this work), there are no monitors in valley towns

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where maximum woodsmoke impacts would be expected. Residential wood combustion in New York is an important source for particulate matter, especially in rural counties where residential wood combustion is responsible for almost all (> 90%) of carbonaceous PM_{2.5} emissions (NYSERDA, 2008). On an annual basis, RWC is the third largest source of PM_{2.5} emissions in New York State (after road dust and miscellaneous, based on the U.S. EPA 2005 National Emissions Inventory), and is the dominant source of PM_{2.5} in the study area of Warren, Essex, Franklin, and Washington Counties (U.S. EPA, 2005). This is important from a public health perspective because even small towns and villages situated in predominately rural areas of New York can have relatively high population densities that are exposed to woodsmoke. Based on U.S. outdoor wood boiler sales, the use of wood fired heating appliances in cold winter climates has increased rapidly, from 4800 units sold in 1999 to a projected 44,600 units in 2007 (Schreiber *et al.*, 2008).

Larson *et al.* (2007) and Su *et al.* (2007) developed the first series of catchment-based land use regression models predicting the spatial variation of woodsmoke levels for three urban areas in the Pacific Northwest. There has been, however, no application of these land use regression models for non-urban areas. Given the potentially important impact of woodsmoke on PM_{2.5} spatial variability and on air quality in non-urban residential areas with high residential wood burning, there is a need to develop tools to improve exposure assessment of woodsmoke PM_{2.5} in these areas, particularly in valley terrains where high spatial variability of woodsmoke PM_{2.5} can occur due to topography.

Mobile and fixed-site woodsmoke measurements were made in the Adirondacks region of upstate New York during the winter of 2008–2009 to support the application of the land use regression models referenced above and predict the spatial variation of residential wintertime woodsmoke in a non-urban region (NYSERDA, 2010). This paper describes the measurement approach used and summarizes the observed spatial and temporal patterns of woodsmoke PM. The modeling component of this project is described in detail in Su *et al.* (2011).

EXPERIMENTAL

The woodsmoke monitoring approach for this study sought out locations with a range of expected levels based on topography and population for comparison with predicted woodsmoke spatial variability obtained from the land use modeling technique (Su *et al.*, 2007). The mobile monitoring used nephelometry as a surrogate for PM_{2.5}, supplemented with multi-wavelength optical absorption measurements as a semi-quantitative but specific woodsmoke indicator. Measurements at six fixed sites ran for the entire study period using only the optical absorption indicator method; this woodsmoke measurement approach is based upon previous work by the Northeast States for Coordinated Air Use Management (NESCAUM) in developing a real-time indicator for ambient woodsmoke (Allen *et al.*, 2004). The mobile monitoring route and location of fixed sites are shown in Fig. 1.

Nephelometer (PM_{2.5}) and Aethalometer™ (Woodsmoke Indicator) Mobile Measurements

A Thermo Scientific (Franklin, MA) model DR-4000 (DR4) nephelometer was used with a 2.5 μm inlet size cut for mobile monitoring PM_{2.5} measurements. Nephelometry (light scattering) has been shown to be a useful surrogate of PM_{2.5} when controlling for relative humidity (RH) above 40% (Molenaar, 2000; Chakrabarti *et al.*, 2004). The DR4 was run with size and relative humidity correction turned off. RH in the DR4 sensing chamber was always less than 35% without additional sample heating, since the instrument was inside a heated automobile and the chamber temperature was well above ambient dew point. The DR4 was zeroed prior to each night's run. Data were collected at one-second intervals. A correction factor of 0.70 was applied to the reported PM data based on earlier comparisons of a DR4 with federal reference method (FRM) and Thermo Scientific FDMS TEOM® samplers (Supplementary Material).

The DR4 PM_{2.5} mobile data were supplemented with one-minute data from a two-channel Aethalometer (model AE-21, Magee Scientific, Berkeley CA), which was used as a specific indicator of woodsmoke to identify the source of the elevated valley PM_{2.5}. The Aethalometer measures light absorption on a quartz filter tape at two wavelengths: 880 and 370 nm. The data are reported in micrograms per cubic meter, and are called BC (absorption by black carbon) and UVC (ultraviolet absorption) respectively. The woodsmoke indicator signal is UVC minus BC, called Delta C, or "DC". Previous work has shown that the DC signal is specific to woodsmoke (cellulose combustion) in ambient air, even in the presence of mobile sources and oil space heating (Kirchstetter and Novakov, 2004; Sandradewi *et al.*, 2008).

Aethalometer data are subject to filter spot loading saturation effects that can be minimized by data correction techniques described by Virkkula *et al.* (2007); this is an essential step for measurement of DC and thus woodsmoke PM. A fixed "Virkkula K" value of 0.01 was used to correct the mobile Aethalometer data; this is a typical value observed for fresh woodsmoke. To convert DC to an estimate of woodsmoke PM concentration for comparison with the DR4 PM_{2.5} data, a factor of 12 (DC × 12 = estimated woodsmoke PM in μg/m³) was used, based on a previous winter study in Rutland, VT that used UNMIX (Henry, 2003; US EPA, 2007) to develop PM_{2.5} source factors from three months of ambient PM and gas measurements (Allen *et al.*, 2004). That work showed a factor of 15, which was adjusted downward for this analysis since the correction for filter spot loading increases the measured DC by approximately 20%. More recent evaluation of the DC to woodsmoke conversion factor was done by the Connecticut Department of Environmental Protection, using similar modeling techniques at four sites and using Aethalometer filter spot loading correction. That study reported an average value of 7.8 for the conversion factor, although results varied substantially across sites and time periods (CT DEP, 2011).

Aethalometer Fixed-Site Woodsmoke PM Measurements

Woodsmoke particulate matter was monitored at the six fixed sites for the entire study period using Aethalometers as

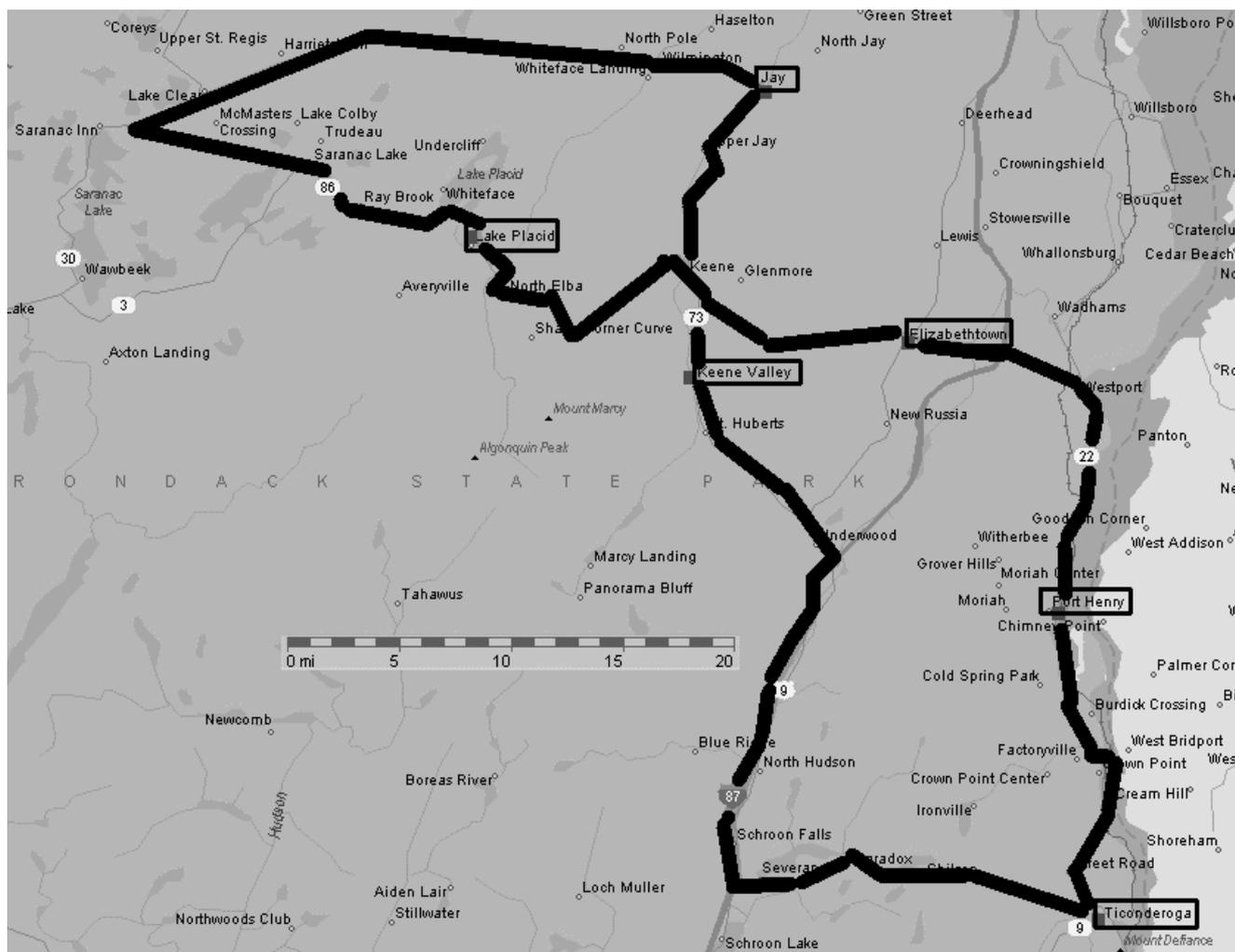


Fig. 1. Map of fixed site sample locations and mobile route.

described above to provide a specific but semi-quantitative measurement of woodsmoke PM. The fixed site monitoring network was designed using techniques described previously by Larson *et al.* (2007) and Su *et al.* (2007). All fixed monitoring sites were located within towns, generally close to the town center. Elevations ranged from 40 meters (Port Henry) to 570 meters (Lake Placid).

Fixed-site Aethalometer data were collected at 5 minute intervals and processed into 1 hour means using the WU-AQL Aethalometer processing software (version 6.0 h) (Turner *et al.*, 2007). This program also generates and applies a dynamic correction factor for filter spot loading saturation (Kirchstetter and Novakov, 2007; Virkkula *et al.*, 2007). The resulting 1-hour data were screened for outliers and negative values. Finally, the DC to woodsmoke PM factor of 12 was applied to create a database of fixed site estimated woodsmoke PM concentrations. This factor can vary widely on short (hourly or less) time scales, but is more stable when data from longer time periods (days to months) are averaged.

A post-study "as found" collocation of the fixed site and mobile Aethalometers was performed. The fixed site data were normalized to the response of the mobile Aethalometer

based on median values of time matched hourly collocation data; this minimizes between instrument bias. The correction to data from any one Aethalometer was 15% or less, within normal operating limits of the method.

Mobile Monitoring Route

The mobile monitoring route (shown in Fig. 1) included all the towns with fixed monitoring sites, as well as Saranac Lake. Location and elevation were recorded at 1-second intervals from a Delorme LT-40 USB GPS using Delorme Street Atlas USA 2009 software. Vehicle speed was recorded at 1-second intervals using an on-board diagnostics (OBD) data logger; at low speeds, these data are more precise than the GPS speed. The monitoring loop was driven both clockwise and counter clockwise to minimize temporal measurement biases by varying the time of night when a town was traversed. The typical mobile monitoring departure time from Saranac Lake was 8 p.m. local time, and the loop took six to seven hours to drive. Driving speeds were approximately 32 km/h (20 mph) or less in towns. At this speed we collected a nephelometer measurement roughly every meter and an Aethalometer measurement approximately every 50 meters; data for analysis were

aggregated up to the 3-minute level for both methods. Driving speeds were at posted speed limits outside of towns. The in-town route always included a drive-by of the fixed monitoring site. Typically 15 to 30 minutes were spent in each town, more in the larger towns and less in the smaller towns.

Mobile monitoring was performed for ten nights. Nights with low wind speed and radiational cooling with valley inversion potential were targeted for mobile monitoring, based on consultations with weather forecasters from the New York State Department of Environmental Conservation. Two of these runs were limited to the northern portion of the loop (the sub loop that included Lake Placid and Jay), with more intensive in-town circuits.

RESULTS AND DISCUSSION

Mobile Loop Measures

The DR4 PM data were considered to be strongly driven by woodsmoke PM concentrations for the mobile runs because other large sources of PM_{2.5} were unlikely at these rural locations in the winter and late at night. Data were screened to identify any periods when PM (DR4) concentrations were elevated with no evidence of

woodsmoke from the DC Aethalometer measurements; in fact, no such "other PM" events were evident. For example, Fig. 2 presents DR4 PM_{2.5} and Aethalometer estimated woodsmoke PM_{2.5} from the mobile loop runs of January 1–2 and 15–16, 2009. The data from both instruments were smoothed with a 3 minute running average; towns are labeled. It is clear from this time series plot that DR4 PM is only elevated when an elevated DC signal is observed. The highest woodsmoke concentrations were present mainly within towns, although there were examples of woodsmoke between towns at lower elevations (Elizabethtown to Port Henry). In high elevation areas between towns, DR4 PM_{2.5} went to essentially zero; this represented regional background concentrations. To provide spatial context for the mobile run data, Fig. 3 shows time series plots of estimated 1-hour mean woodsmoke (Aethalometer Delta C) from the six fixed sites for the three mobile run nights discussed in this paper.

The mobile loop data show that the quantitative relationship between the Aethalometer DC woodsmoke signal and the DR4 PM_{2.5} data varied substantially. Although woodsmoke (Aethalometer DC) was always present when DR4 PM_{2.5} is elevated, the relative relationship varied by a factor of six or more at the three-minute timescale of this mobile monitoring. This is

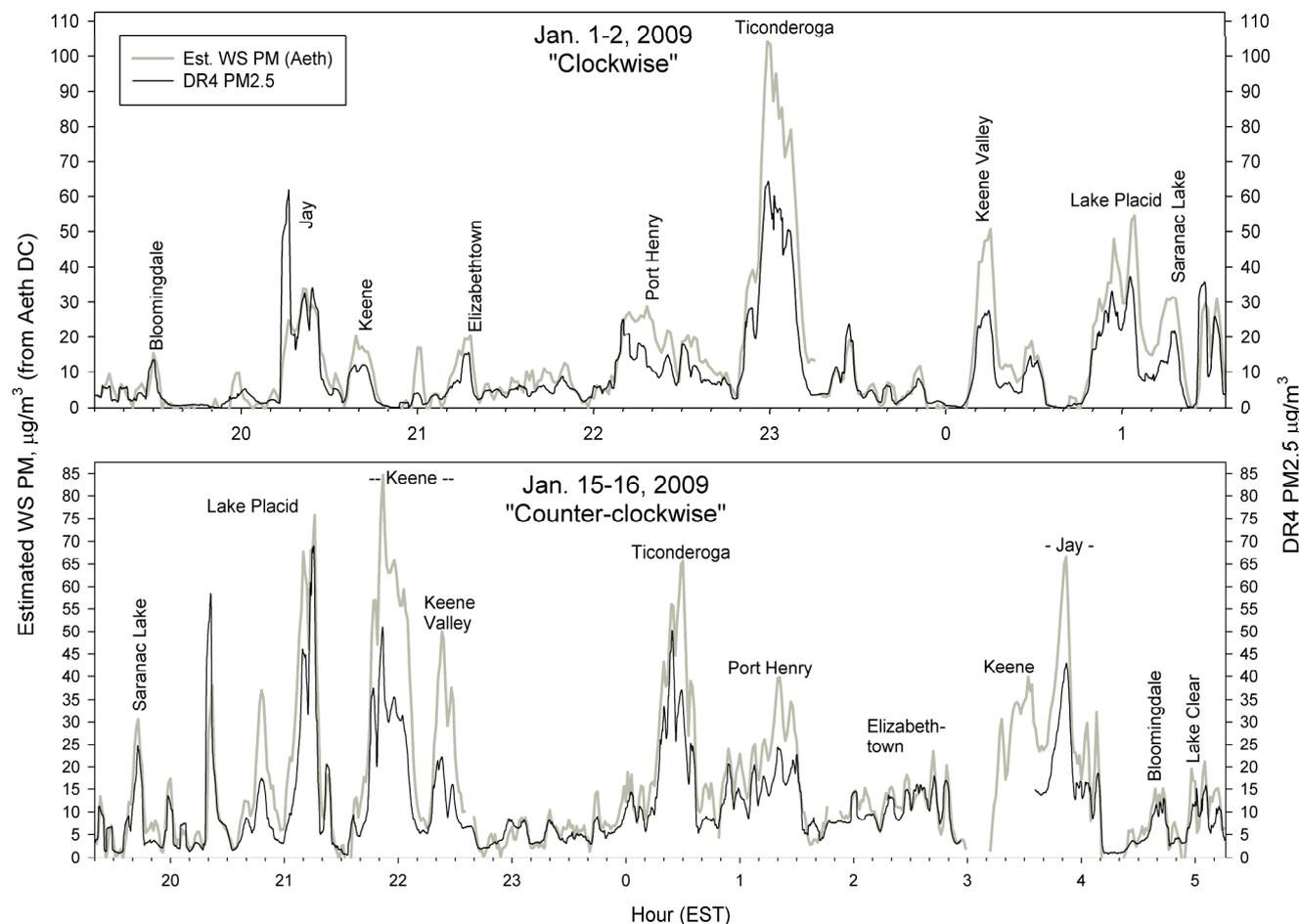


Fig. 2. DR4 PM_{2.5} and estimated woodsmoke from mobile measurements, 3 minute running averages, January 1–2 and 15–16, 2009.

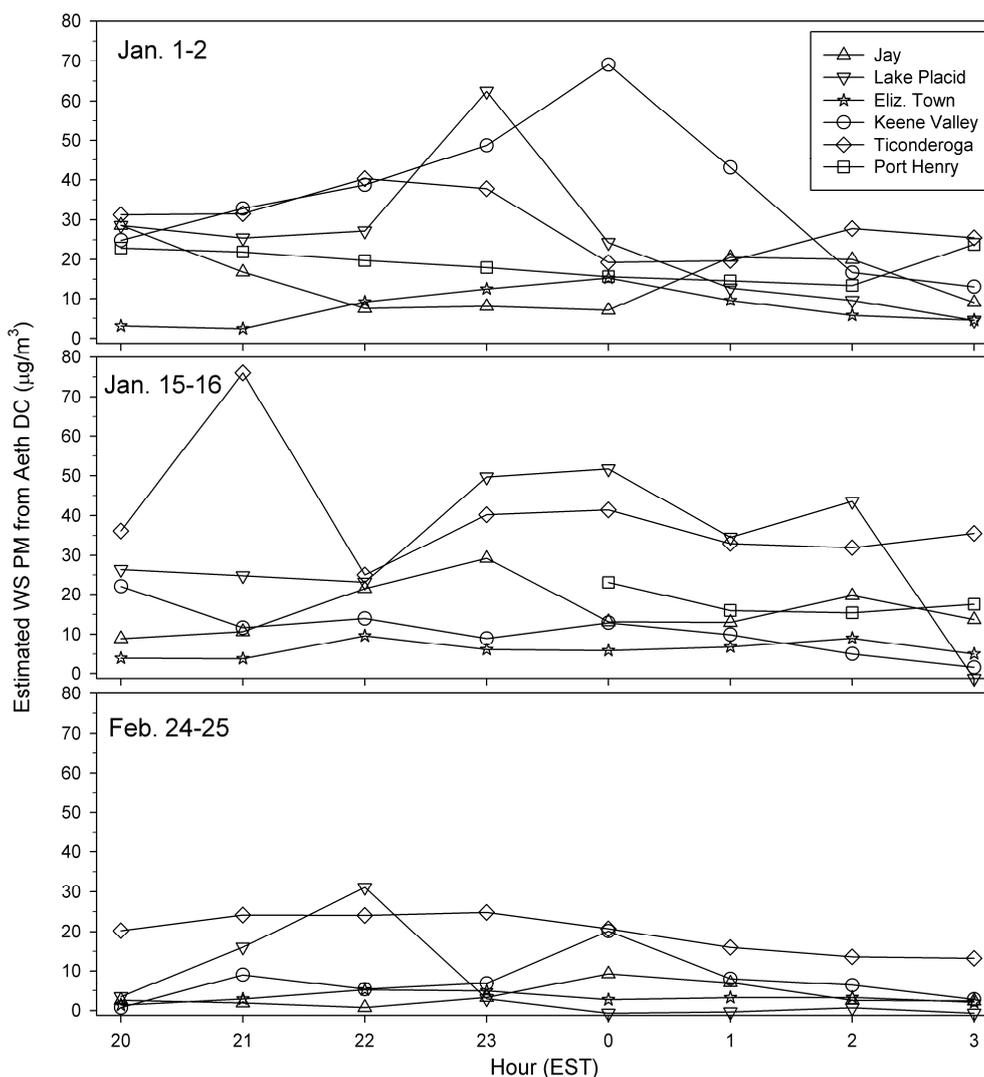


Fig. 3. Time series plots of estimated 1-hour mean woodsmoke (Aethalometer Delta C) from the six fixed sites for three mobile run nights.

likely due to highly heterogeneous woodsmoke composition and the short measurement time scale (a few minutes or less), compared to the long time scale (days to weeks) and neighborhood scale siting of earlier fixed-site modeling work. The DC to woodsmoke-PM ratio is influenced by particle composition and morphology, and is therefore a function of many factors, including type of wood burned, combustion conditions, and the age of the smoke. Thus, the mobile loop Aethalometer data were used primarily as a semi-quantitative indicator that woodsmoke was present.

Another example of the spatial patterns where woodsmoke was elevated primarily within towns from mobile monitoring on February 24–25, 2009 is shown in Figs. 4 and 5. Fig. 4 is a time-series of DR-4 and Aethalometer estimated WS PM with elevation. Fig. 5 is a bubble plot for the “return” west-bound portion of that night’s run. The diameter of a bubble is proportional to PM concentration. During this sampling session a quick run east was followed by a slow westbound return. Because substantial woodsmoke had been observed in the town of Saranac Lake (not one of the

fixed sites) on earlier runs, additional time was spent in that area during this run. This plot clearly shows that the highest concentrations were generally in towns, with very low levels between towns. The time-series plot (3-minute running averages of DR4 PM and estimated woodsmoke PM) is similar to those shown in Fig. 2.

The bubble plot in Fig. 5 shows 1-minute DR4 PM spatially during the entire return portion of this route, ending at 2:20 AM. Here, the axes are latitude and longitude, essentially a map of the domain driven. The diameter of the circle (one every minute) represents the concentration. “Home” represents the end point of the evening’s trip, a few miles southwest of Lake Clear (see the route map) near the northern tip of Saranac Lake. The highest 1-minute $PM_{2.5}$ concentrations (“plume hits”) were observed in Jay and on a small side road northeast of the top of Saranac Lake (at 2:10 AM).

Using the highly time resolved $PM_{2.5}$ data from the DR4, it is possible to identify very short term spikes due to what are likely individual smoke plumes. Fig. 6 shows 1 second $PM_{2.5}$ data (actual response time is ~5 seconds) from the Feb.

24–25 sub-loop return run. For this plot, it is important to also show vehicle speed because it has a strong influence on

the duration of a PM_{2.5} spike from a single fixed source. One second car speed data were from the OBD data logger.

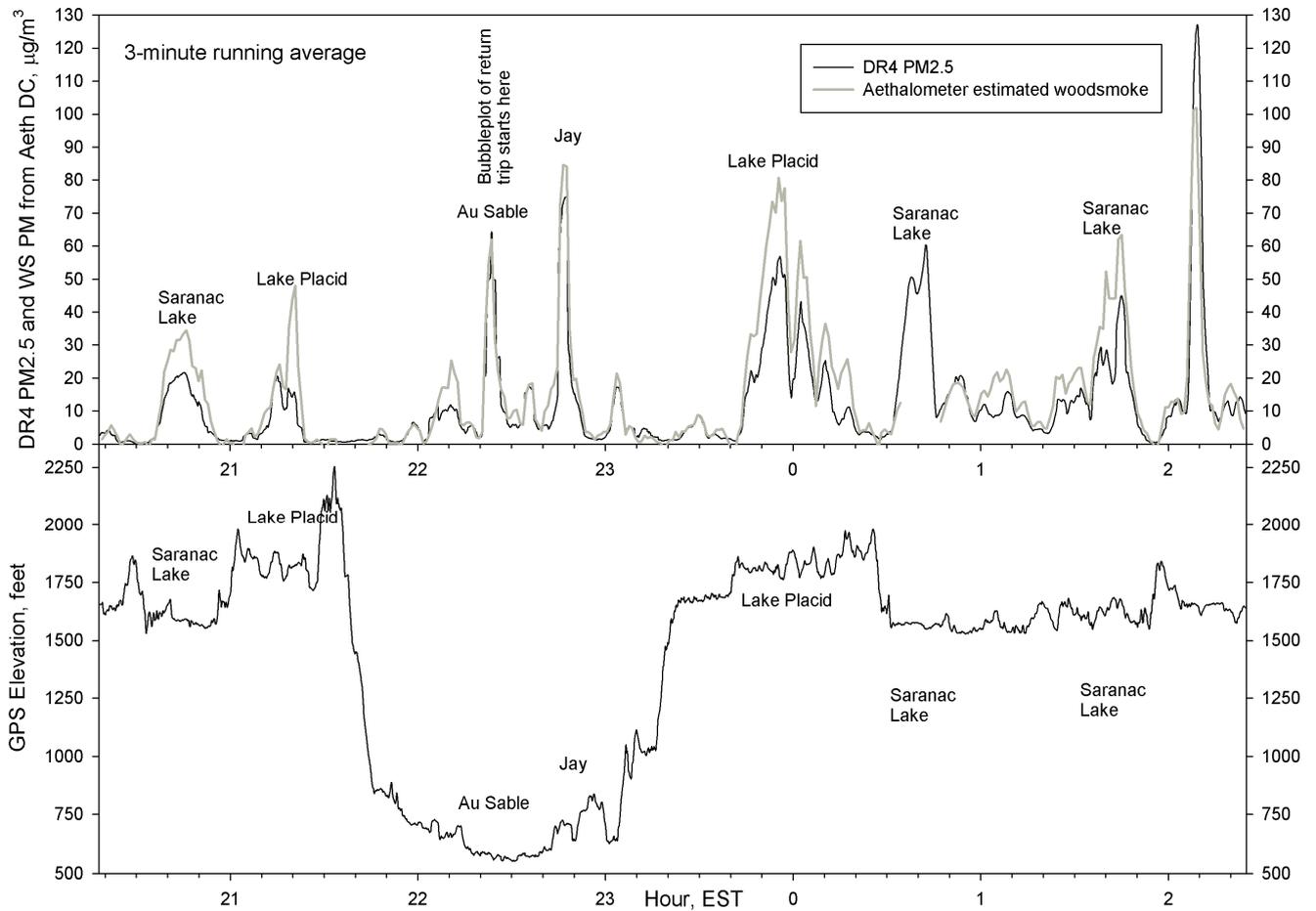


Fig. 4. Time-series of DR4 and Aethalometer estimated WS PM and elevation for full mobile loop drive, February 24–25, 2009.

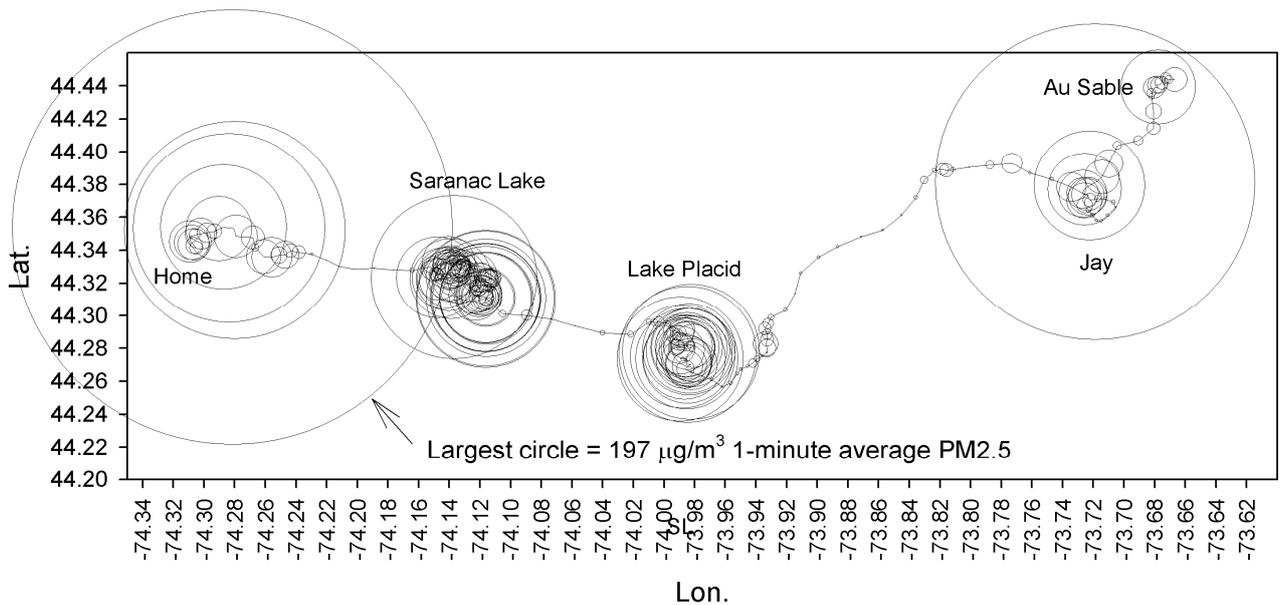


Fig. 5. Bubble plot of westbound portion of February 24–25, 2009 mobile monitoring ending at 2:23 AM.

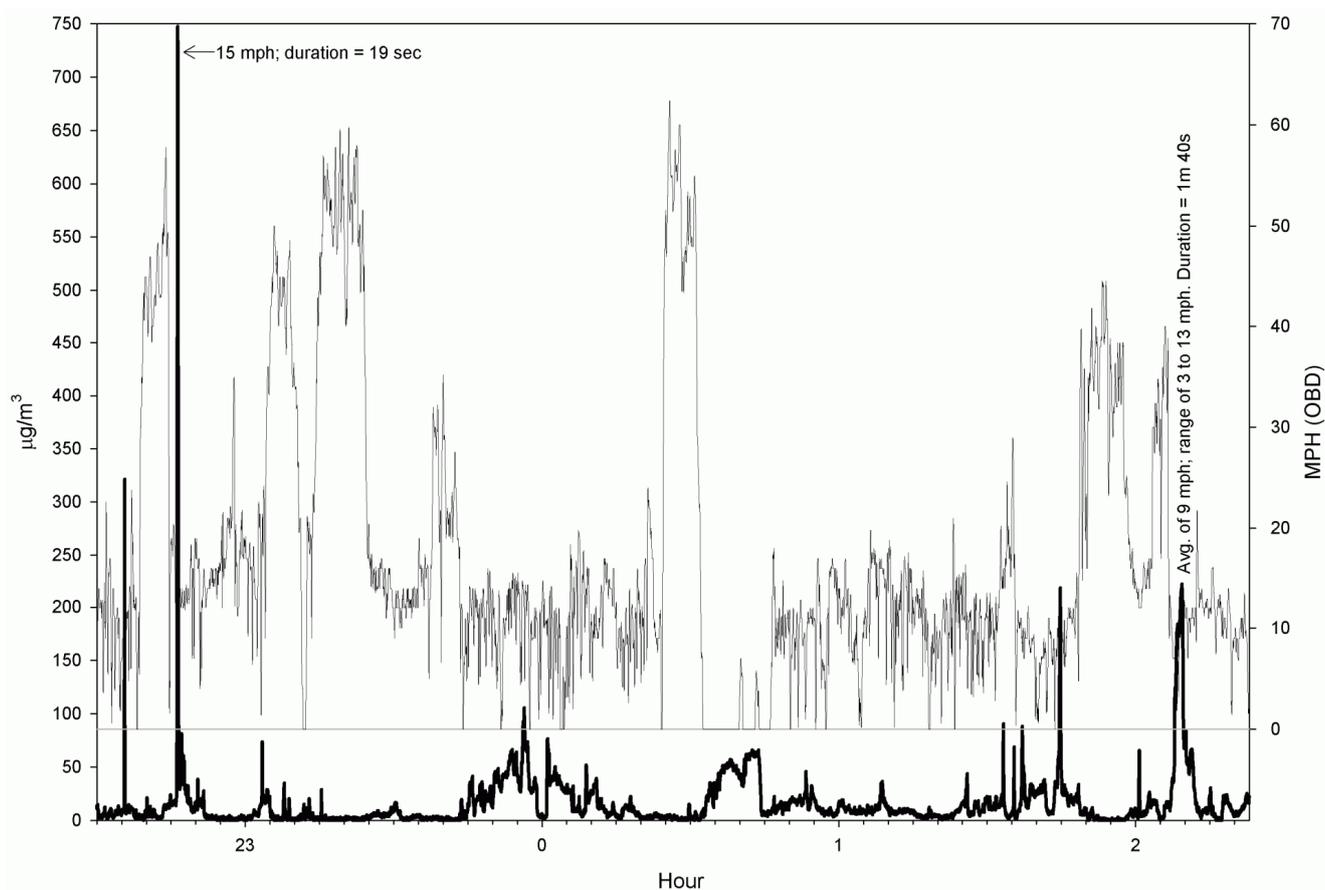


Fig. 6. Left axis: Plot of $PM_{2.5}$ 1 second data (lower dark line). Right axis: Plot of car speed (upper light line), February 24–25, 2009.

For this short averaging time, the highest $PM_{2.5}$ concentration of $750 \mu\text{g}/\text{m}^3$ occurred in Jay at 22:47 (10:47 p.m. local time), but was only present for ~ 20 seconds at a speed of 25 km/h. This represents a distance of about 135 meters. The largest circle in the bubble plot occurred near the end of the run, at 2:10 a.m. local time. That event was a broader peak of $225 \mu\text{g}/\text{m}^3$, lasting 1 minute 40 seconds. The car was traveling slower, at an average speed of 15 km/h; the distance covered during the peak of this event was 400 meters. It is not known if the larger spatial scale of this event was due to local topography or to the plume traveling more parallel to the road.

A detail of the last 2.5 hours of this trip to 2:00 AM is shown in Fig. 7, covering the intensive monitoring in Lake Placid and Saranac Lake. This plot shows 15 second averages of DR4 $PM_{2.5}$ and elevation. Of particular interest in this figure is the sharp drop in PM at midnight, corresponding to the few minutes when the mobile monitoring was out of the bottom of the Lake Placid valley, and near the Lake Placid fixed site location. This indicates that at least on this night, the fixed site was above the inversion layer later in the evening (as shown in Fig. 2), and PM measured there was not representative of the bottom of the valley.

Fixed Site Measures

Fig. 8 presents the distribution of hourly estimated woodsmoke PM from the six fixed site monitors during the ten evenings when mobile runs were performed. The median and mean levels of estimated woodsmoke PM during these ten evenings were $22 \mu\text{g}/\text{m}^3$ or less. The highest hourly peaks (95th percentile) were generally above $40 \mu\text{g}/\text{m}^3$ at four of the sites (Lake Placid, Ticonderoga, Port Henry, and Keene Valley), while two sites (Jay and Elizabethtown) were between 10 and $22 \mu\text{g}/\text{m}^3$. Of the six fixed monitoring sites, Keene Valley has the lowest population (approximately 1000), yet is among the sites with relatively high woodsmoke PM during these 10 evenings. This may be a reflection of its topography (a very narrow valley).

The domain of the mobile loop measurements did not always behave uniformly with regard to temporal patterns of elevated woodsmoke. A more detailed example of shorter term spatial variation over the study domain is shown in Fig. 9, a 3-hour smoothed time-series of estimated woodsmoke PM from the fixed sites over an eleven day period. There were periods where all sites had low woodsmoke concentrations (February 13), and where all sites were elevated (February 6 and 7). There were also several times where woodsmoke was elevated at some sites but not others (February 5 and 14). This spatial decoupling could be due to weather systems moving through the region. Again it should be noted that at these shorter timescales, the estimate of

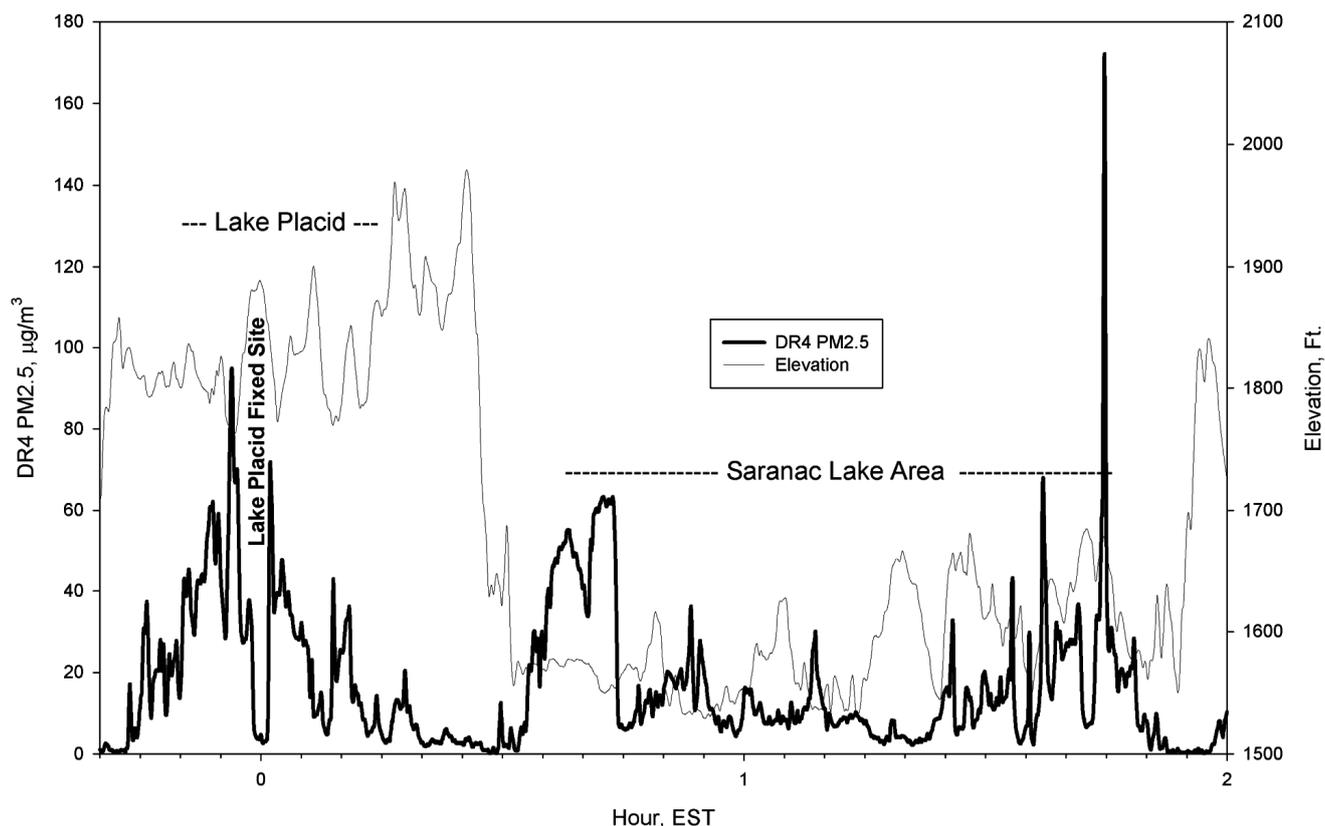


Fig. 7. Plot of elevation and DR4 PM_{2.5} (15 second running average), February 24–25, 2009 during second half of shortened loop mobile measurements.

woodsmoke PM_{2.5} from Aethalometer DC may be more variable compared to longer time periods (weeks or longer), because smoke from a single or few sources may dominate short duration concentrations and the DC to woodsmoke relationship can vary depending on the specific nature of the smoke (e.g., type of wood, combustion conditions, plume age).

The typical mid-day dispersion of valley pollution is clearly shown in Fig. 10, a diurnal plot of BC, DC, and the BC/DC ratio from Ticonderoga for the 4-month winter period of December through March. Vertical mixing (driven by solar radiation) and wind speed, and thus dispersion of local pollution sources, peaks at mid-day. This, along with reduced source emissions from both traffic and woodsmoke, results in a mid-day minimum in PM_{2.5}. Local sources of PM_{2.5} in Ticonderoga are from traffic and woodsmoke. Oil residential space heating does produce BC (“soot”) and could be a confounder for this analysis, but BC from oil heat is approximately two orders of magnitude lower than uncontrolled woodsmoke heating sources for an equivalent amount of heat generated (Schreiber and Chinery, 2008). This relationship assumes that BC is 5 to 10% of woodsmoke PM on a mass concentration basis (McDonald *et al.*, 2000; McDonald *et al.*, 2006). DC is only from woodsmoke, while BC is from both traffic and woodsmoke sources. Woodsmoke dominates the BC in this example, although the variation of the ratio of BC to DC (BC lower at night and higher morning and mid-day) suggests some influence by local traffic during

daytime hours. It is reasonable to assume that averaged over the winter, the BC to DC ratio in woodsmoke does not change much over the day. Thus, changes in the ratio of BC to DC are most likely traffic related.

CONCLUSIONS

The combination of nephelometer and 2-channel Aethalometer measurements provide a highly time resolved and specific measurement of woodsmoke PM that can be deployed on a mobile platform or in valley areas not monitored by existing networks. Mobile Aethalometer measurements clearly showed that elevated night-time PM was due primarily to woodsmoke. Woodsmoke was highly variable both temporally and spatially in a winter semi-rural valley setting, and night-time concentrations were elevated only within the valleys. Very short term (15 seconds) concentrations can exceed several hundred µg/m³, indicative of local plumes.

Fixed site wood smoke monitors demonstrated woodsmoke concentrations highest at midnight and again at 7 AM. Woodsmoke concentrations were lowest at mid-day. This illustrates the usefulness of Aethalometers to detect woodsmoke at all times of the day and may be an important tool when coupled with a nephelometer for identifying sources of woodsmoke in efforts to resolve woodsmoke exposure complaints.

Fixed-site estimated woodsmoke PM concentrations

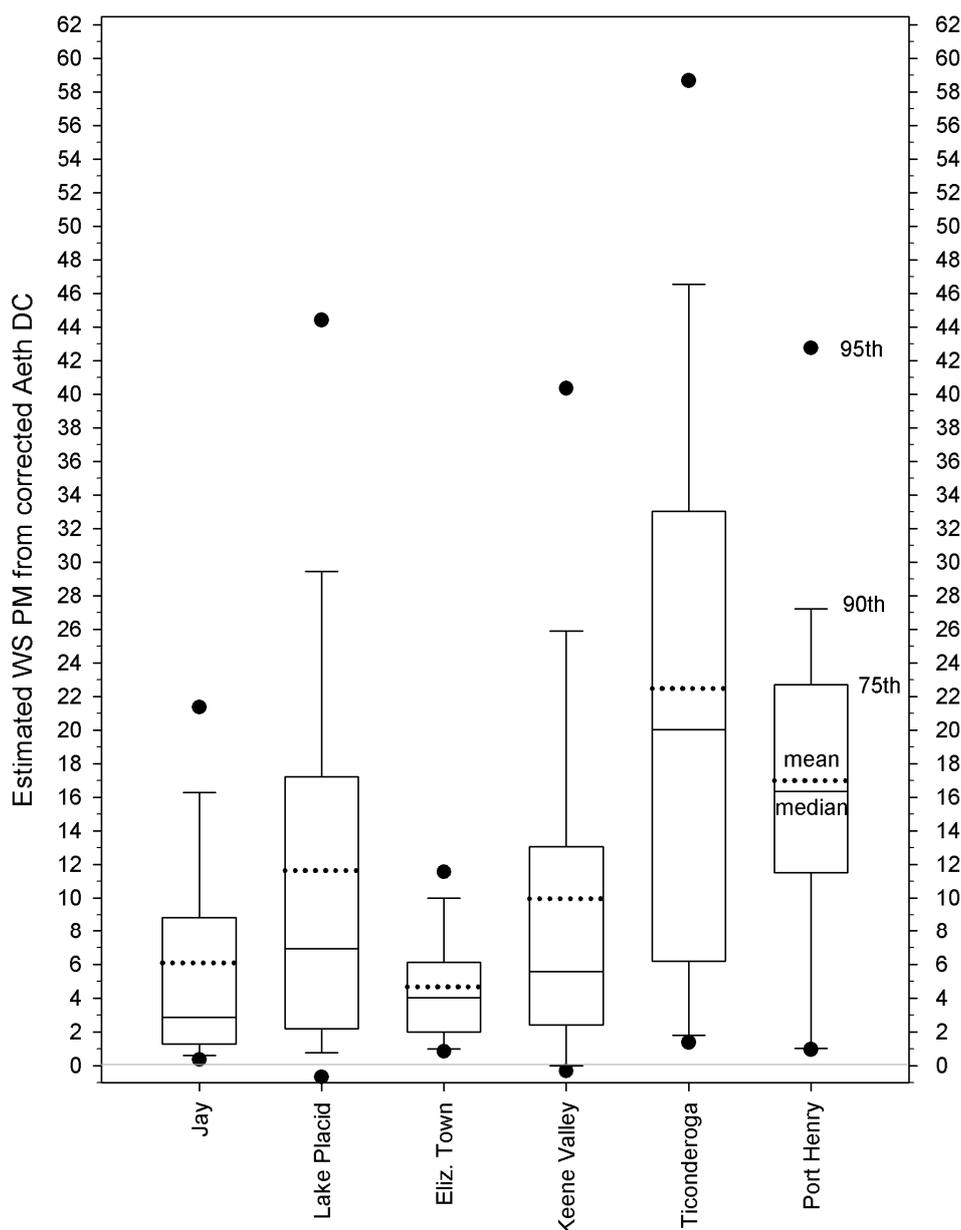


Fig. 8. Distribution of fixed site estimated hourly woodsmoke PM during evenings when mobile monitoring between fixed sites was conducted.

approached $100 \mu\text{g}/\text{m}^3$ for periods of three hours. These concentrations of $\text{PM}_{2.5}$ have been shown to be health-relevant (NYSERDA, 2006). Patterns of woodsmoke PM on the time-scale of a few hours are often not well coupled across the domain of the northern Adirondacks in upstate New York, suggesting variable meteorology. Diurnal analysis showed woodsmoke patterns were consistent with space heating use and improved mid-day dispersion. The BC to DC diurnal ratio can be used to distinguish between local traffic and woodsmoke contributions to PM at these sites.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

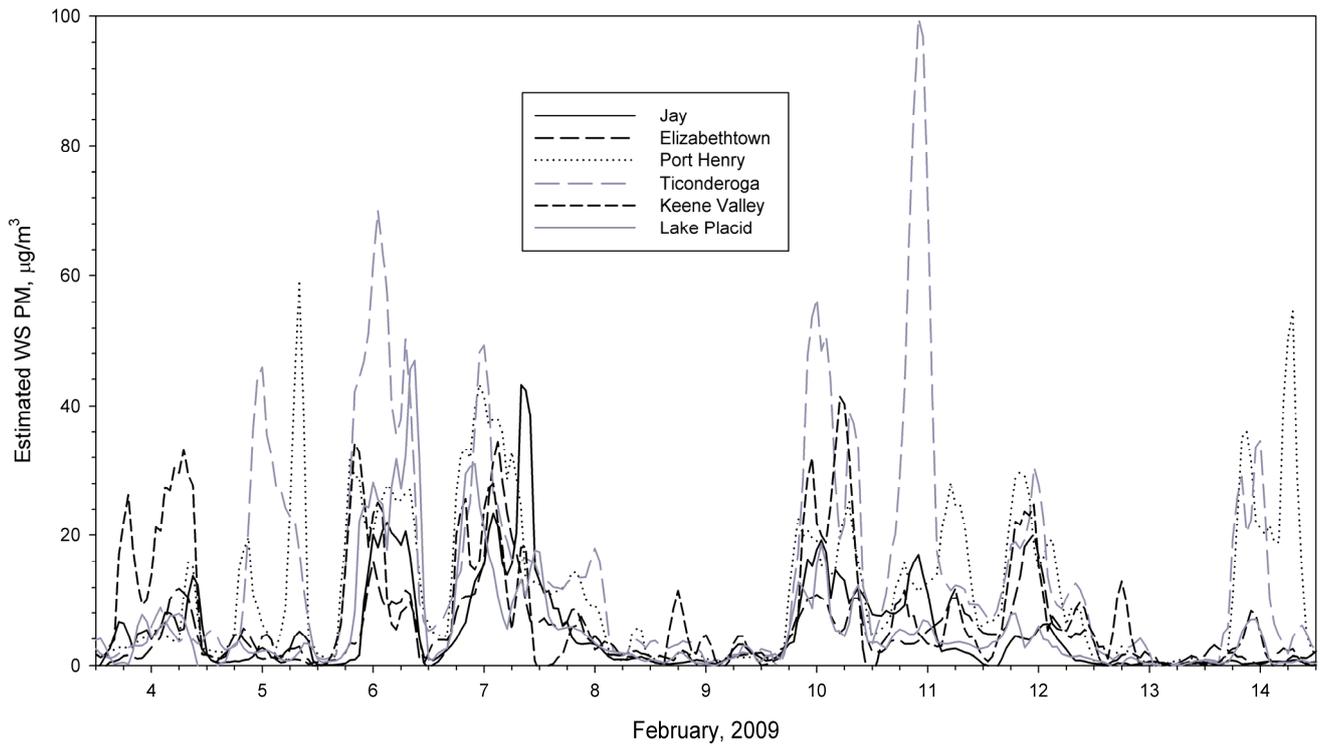


Fig. 9. Estimated woodsmoke PM, 3-hour running average, February 3–14, 2009.

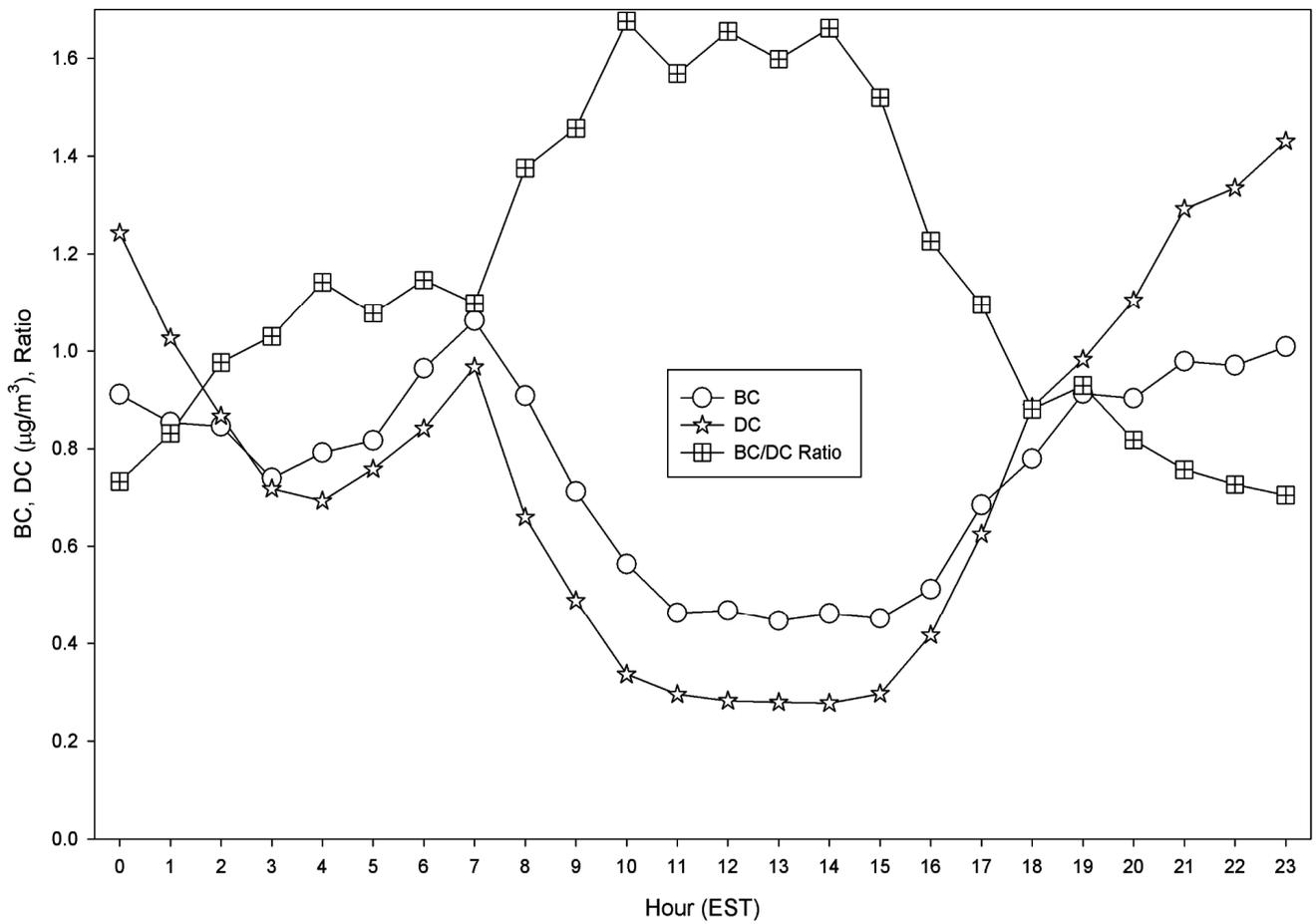


Fig. 10. Diurnal plot of PM_{2.5} and woodsmoke pollution at the Ticonderoga monitoring site, December though March.

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