Anonymous Referee #1

We greatly appreciate the valuable comments from Anonymous Referee #1. In addition to the replies below, the revised manuscript with marked changes are also enclosed.

1. *The authors are interested in quantification of the contribution of woodsmoke to PM2.5. Source apportionment always depends on the assumptions of the method employed to determine the sources. The authors make the statement that PM2.5 is dominated by woodsmoke on p. 5 (lines 12-13) and p. 10 (line 14). This should be the result of the work, not an initial assumption, which cannot be tested without additional chemical methods being applied to the same samples. The starting assumptions need to be checked and a "proof" that PM2.5 is exclusively due to woodburning needs to be produced. Alternatively, a reference to a monitoring agency report or previous publications would be advantageous.*

The authors would like to acknowledge the vague definition of "woodsmoke-dominated". In the original manuscript, we cited a report from the State of Vermont showing the wood is the dominant heating fuel in Rutland (Frederick and Jaramillo, 2016) and the 2014 National Emission Inventory (NEI) to show that woodsmoke is a dominating PM emission source. We have provided more quantitative information in the revised manuscript to make the point. The NEI only provide county-level annual emission inventories. The town of Saranac Lake spans two counties in New York, i.e., Essex and Franklin. The point we try to make is that woodsmoke emission is the predominant source of wintertime PM2.5 emissions in both Rutland and Saranac Lake. For the plume environments in Ithaca, NY, it is woodsmoke-dominant in nature as we purposefully sampled woodsmoke plumes.

In Section 2.1 of the revised manuscript, we added, "According to the 2014 National Emission Inventory, residential woood combustion (RWC) contributes to approximately 38.6% of the annual PM2.5 emissions in Rutland County. In comparison, on-road mobile sources only account for 1.4%. Considering the seasonal patterns of various emission sources, it is clear that RWC is the predominant primary PM2.5 source in Rutland during wintertime." and "The 2014 National Emission Inventory indicated that RWC accounts for approximately 22.4 to 25.4% of the annual PM2.5 emissions, while the contribution of on-road mobile sources is between 2.8 to 3.9%, which indicated that it is also a woodsmoke-dominated environment during wintertime."

In addition, the main goal of our study is not a source apportionment in the three reported towns/cities. As described above, even on annual basis, woodsmoke PM2.5 emissions is 10 to 40 times higher than mobile emissions in Rutland and Saranac Lake. During wintertime, we expect the woodsmoke emission would exceed mobile sources by over 100 times. Our primary goal is to address the concerns whether DC (aka Delta-C) is a useful woodsmoke marker for air quality management, in both qualitative and semi-quantitative sense. As presented in the last paragraph of the introduction section, "Our study can be regarded as a "necessary condition test" for DC serving as a woodsmoke PM marker. In other words, DC would be deemed an

inappropriate marker if it were unable to track woodsmoke PM patterns even under woodsmoke-dominated environments."

2. *BC and $PM_{2.5}$ have similar diurnal variation. We can see from Figures 3 and 4 that the BC/PM ratio is not constant during the day. This means that the composition of PM is changing. The authors correctly point out that unless UV-absorbing species feature a constant absorption cross section, DC cannot be considered quantitative. The change in composition implies the change in the absorption cross section. This part of section 3.3 needs to be expanded and arguments provided. Comparison to other source apportionment methods using similar methods would help (Sandradewi et al., 2008).*

We agree with the reviewer that the varying BC/PM ratio may indicate changing composition as well as changing absorption cross section. The plume data presented in Figure 6 also imply varying absorption cross section with combustion conditions. The main message in Figure 5 is that averaging stationary PM and BC data over a long period of time (e.g., over a winter month or longer in a fixed location) may lead to an average absorption cross section, i.e., a constant Δ(Ambient PM2.5)/ΔDC. We have revised the first paragraph in Section 3.3.1 by adding the following sentences, "Furthermore, Figure 5 suggests that averaging stationary PM and BC data over a long period of time (e.g., over a winter month or longer in a fixed location) may lead to an average absorption cross section, i.e., a constant Δ(Ambient PM2.5)/ΔDC, even though PM composition and the resulting absorption cross section may vary with time."

The research team initially planned to apply the method presented by Sandradewi et al. (2008ab) to the ambient data. However, as described in Section 1 of the original manuscript, that method often requires light absorption measurements at multiple wavelengths to have a reliable estimate on α. Since the ambient data to be presented in this paper were collected by a two-wavelength Aethalometer, we did not attempt to calculate α from the ambient data. Note that we employed a seven-wavelength Aethalometer in the plume measurement, which allowed us to apply the method of Sandradewi et al. (2008). We will report the related findings in a separate publication. We further revised the related paragraph in the Introduction to elaborate our rationale: "Another approach taking advantage of UV enhancement (or wavelength dependence of the aerosol absorption coefficient in general), as reported by Sandradewi et al. (2008a), derives light absorption Ångström exponents (α) from multi-wavelength Aethalometer readings. α is close to 1 for traffic sources, and varies for woodsmoke, but generally much larger than 1. Assuming certain value of α for woodsmoke, Sandradewi et al. (2008b) conducted quantitative analysis of source contributions to PM. This approach often requires light absorption measurements at multiple wavelengths to have a reliable estimate on α (Chen et al., 2015). Sandradewi et al. (2008b) showed that using different pairs of wavelengths led to different values of α for woodsmoke. Since the ambient data to be presented in this paper were collected by a two-wavelength Aethalometer, we did not attempt to calculate α. Given the uncertainties associated with values of α for woodsmoke for our study, we did not perform the source apportionment analysis similar to that presented by (Sandradewi et al., 2008a)."

Furthermore, as described in our reply to Comment 1, we purposefully selected woodsmoke-dominated environments to conduct a necessary condition test for DC as a woodsmoke marker. Our study does not directly address whether DC is a good woodsmoke maker in environments not dominated by woodsmoke, where source apportionment is probably necessary.

3. *Finally, the manuscript ignores the production of SOA. The change in composition of PM is already evident from the diurnal variation of the BC/PM ratio. SOA and the applicability of the method described in the manuscript need to be discussed in terms of primary and secondary PM. SOA can dominate PM in places where PM is heavily impacted by wood burning – the sites presented in the articles are such places.*

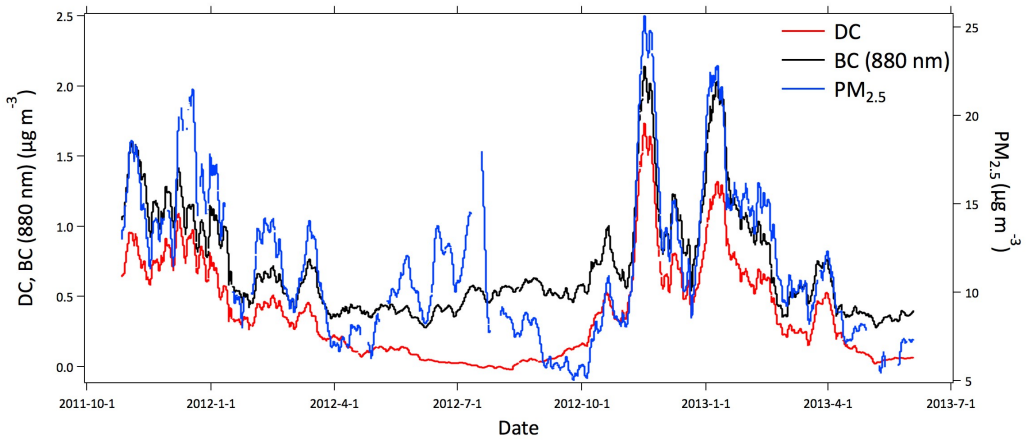
In the original manuscript, we described our thinking why SOA does not look like a main driver for DC. We revised the related discussions to make our point more clear. The revised discussions appear near the end of the second paragraph in Section 3.2: "As mentioned earlier, previous studies found that SOA products may result in DC signals. If SOA formation were significant, we would expect that PM2.5 and/or DC would peak around mid-day. The distinct diurnal patterns illustrated in Figure 4 is more consistent with strong influence of local emissions. Moreover, the seasonal trend shown in Figure 3 indicates that DC peaked during wintertime when SOA production is small and approached zero during summertime when SOA production is expected to be high. Therefore, both the diurnal and seasonal patterns indicate that SOA is not a main driver for DC in Rutland."

4. I agree with Reviewer 2 that the presentation of the “PAH” as measured in this campaign is weak. I would recommend to either remove this subsection or significantly redact it and expand the discussion on the PAH measurement method. If the authors expand the section, they should switch the PM2.5 and PAH axes. They argue that PAH is not as good an indicator as DC, hence the plot should be made into PAH as a constituent of PM2.5 by switching the axes.

We have selected the referred section in the revised manuscript.

5. *Dates need to be changed so that the European readers will find them unambiguous (Figure 3).*

The revised figure is attached below and included in the revised manuscript.



6. *Figure 7: Why just report this for Rutland?*

Among the sites included in our study, only Rutland had DC data over a year. We will search for more data in other locations, and likely report the findings in a separate publication. For long-term continuous light-absorption measurement in a woodsmoke-dominated environment, Rutland is probably one of very few in the U.S.

Anonymous Referee #2

1. *I found the analyses focused on the three fixed site data collection straightforward and the conclusions well-supported. I think Figure 7, DC/BC vs Heating Days is particularly compelling. Why is it only shown for Rutland site? It would be interesting to understand how this stable this relationship is.*

Among the sites included in our study, only Rutland had DC data over a year. We will search for more data in other locations, and likely report the findings in a separate publication. For long-term continuous light-absorption measurement in a woodsmoke-dominated environment, Rutland is probably one of very few in the U.S.

2. *How was the CO2 data used?*

We did not use the CO_2 data in the current manuscript. The data did not pass our quality assurance (QA) check. We are still trying to figure out the possible causes.

3. *There are two parts of this paper that are weak and I would recommend removal. The mobile monitoring in Ithaca is not well integrated and it does not add significant value to the paper. Likewise the PAH comparison is very lightly discussed and basically dismissed by the authors themselves. I recommend that these sections be removed so that the main point of the paper, the DC/BC analysis, is clear.*

We have removed the section discussing PAH measurement. We'd like to explain why we think the Ithaca plume analysis is inherent part of the paper. The main goal of our manuscript is to address the concerns whether DC (aka Delta-C) is a useful woodsmoke marker for air quality management, in both qualitative and semi-quantitative sense. By studying woodsmoke plume data, we showed linear relationships between PM2.5 and DC can be used to distinguish different combustion conditions. The fundemental principle is that different combustion conditions lead to different PM composition, which in turns lead to different absorption cross sections. The high time resolution AE-33 we deployed can capture the changes in absorption cross sections, which can potentially allows us to track combustion conditions. We added the following sentence near the end of the second paragraph of Section 3.3.2, "In other words, the different combustion conditions lead to different chemical compositions and absorption cross sections, which can be potentially captured by high time resolution light absorption measurements."

Joint Measurements of $PM_{2.5}$ and light-absorptive PM in woodsmoke-dominated ambient and plume environments

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Abstract. DC, also referred to as Delta-C, measures enhanced light absorption of particulate matter (PM) samples at the near-ultraviolet (UV) range relative to the near-infrared range, which has been proposed previously as a woodsmoke marker due to the presence of enhanced UV light absorbing materials from wood combustion. In this paper, we further evaluated the applications and limitations of using DC as both a qualitative and semi-quantitative woodsmoke marker via joint continuous measurements of $PM_{2.5}$ (by nephelometer pDR-1500) and light-absorptive PM (by 2-wavelength and 7-wavelength Aethalometer®) in three Northeastern U.S. cities/towns including Rutland, VT, Saranac Lake, NY and Ithaca, NY. Residential wood combustion has been shown to be the predominant source of wintertime primary $PM_{2.5}$ emissions in both Rutland and Saranac Lake, where we conducted ambient measurements. In Ithaca, we performed woodsmoke plume measurements. We compared the pDR-1500 against a FEM $PM_{2.5}$ sampler (BAM 1020), and identified a close agreement between the two instruments in a woodsmoke-dominated ambient environment. The analysis of seasonal and diurnal trends of DC, BC (880 nm) and $PM_{2.5}$ concentrations supports the use of DC as an adequate qualitative marker. The strong linear relationships between $PM_{2.5}$ and DC in both woodsmoke-dominated ambient and plume environments suggest that DC can reasonably serve as a semi-quantitative woodsmoke marker. We proposed a DC-based indicator for woodsmoke emission, which was then shown to exhibit relatively strong linear relationship with heating demand. While we observed reproducible $PM_{2.5}$-DC relationships in similar woodsmoke-dominated ambient environments, those relationships differ significantly with different environments, and among individual woodsmoke sources. ~~DC correlated much more closely with $PM_{2.5}$ than EcoChem PAS2000-reported PAH in woodsmoke-dominated ambient environments.~~ Our analysis also indicates the potential for $PM_{2.5}$-DC relationships to be utilized to distinguish different combustion and operating conditions of woodsmoke sources, and that DC-Heating demand relationships could be adopted to estimate woodsmoke emissions. However, future studies are needed to elucidate those relationships.

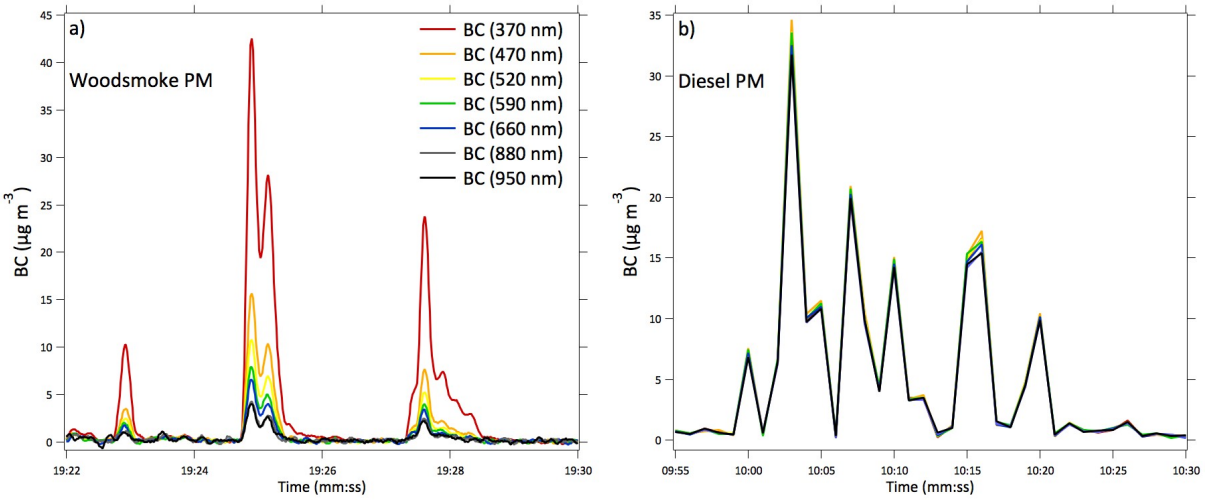
1 Introduction

Woodsmoke resulting from anthropogenic activities is a widespread air pollution problem in many parts of the world. For example, residential woodsmoke is estimated to account for 20% of total stationary and mobile polycyclic organic matter emissions, and 50% of all area source air toxic cancer risks according to the 2011 National Air Toxics Assessment in the U.S. (https://www.epa.gov/national-air-toxics-assessment). It is reported that around 35% of total $PM_{2.5}$ emissions in the United Kingdom came from domestic wood burning in 2015, while road transport only contributed around 13% of the total $PM_{2.5}$ emissions (DEFRA, 2016). In addition to its contribution to regional air quality, residential woodsmoke may cause significant near-source air quality impacts due to relatively low stack heights and low exhaust temperatures. While in some sense wood burning products may be considered natural substances, the health effects of wood smoke are found to be comparable to those of fossil-fuel combustion sources (Naeher et al., 2007).

Chemicals that are enriched in woodsmoke relative to other sources have been used to quantify woodsmoke impacts on ambient particulate matter (PM). Among them, levoglucosan, a sugar anhydride derived from the pyrolysis of the major wood polymer cellulose, has been used extensively as a molecular marker for woodsmoke because it is emitted at high concentrations and relatively stable in the atmosphere (Fine et al., 2001; Simoneit et al., 1999). However, detecting levoglucosan in PM samples at present requires detailed chemical analysis, and the related information is not widely available.

The widely deployed Aethalometer® has made possible continuous aerosol light absorption measurements, commonly referred to as Black Carbon (BC), at either two wavelengths (880 nm and 370 nm) or seven wavelengths (370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm, and 950 nm). Allen et al. (2004) first proposed using enhanced light absorption of ambient particulate matter (PM) at 370 nm relative to 880 nm, due to the presence of light absorbing materials from wood combustion near ultraviolet (UV) range, as a marker for woodsmoke PM. Figure 1 depicts the distinct responses of a seven-wavelength Aethalometer (Magee Scientific AE-33) to woodsmoke (Figure 1a) and diesel (Figure 1b) plumes, providing a context for our discussions in this paper. The source of the diesel plume was a backup diesel generator, and the measurement was conducted in 2015. The woodsmoke plume data was collected near a residential woodstove source in early 2016. Note that the purpose of Figure 1 was to reveal the qualitative differences between the two sources, rather than making a quantitative comparison.

The wavelength-dependent responses to woodsmoke were clearly shown in Figure 1a. At the longer wavelength end, there were virtually no differences in the signals from the 880 nm and 950 nm channels. At the shorter wavelength end, the 370 nm channel recorded the highest reading. We referred to the augmented responses at shorter wavelengths compared to the 880 nm and 950 nm as "UV enhancement". By contrast, virtually no wavelength-dependence (i.e., no UV enhancement) was observed for diesel exhaust (Figure 1b). There are some slight discrepancies among the different wavelength channels, likely due to the limitations of the real-time dynamic spot loading correction scheme used by the AE-33. The patterns of the wavelength-dependent responses shown in Figure 1 were consistent with the findings from several previous studies, which suggested that UV absorbing compounds are enriched in biomass-combustion PM but scarce in diesel PM (Chen et al., 2015; Olson et al., 2015) or traffic-related PM (Kirchstetter et al., 2004). Broadly, the light-absorbing organic compounds, referred to as "brown carbon" or BrC, have been shown to strongly absorb UV (Andreae and Gelencsér, 2006).



(a) (b)

Figure 1. Wavelength-dependent responses of AE-33 to (a) woodsmoke and (b) diesel plumes. Note that the purpose of this figure is to reveal the qualitative differences, rather than making a quantitative comparison, between the two types of plumes.

The concept of DC (also referred to as Delta-C) originated from using the level of the UV enhancement as a marker for woodsmoke PM (Allen et al., 2004). Traditionally, DC was calculated by the differences between 370 nm and 880 nm signals, i.e., DC = BC (370 nm) – BC (880nm), due to the availability of two-channel Aethalometer models. But the concept is not limited to those two particular wavelengths. Figure 1a indicates that woodsmoke UV enhancement started appearing at 660 nm, and more enhancement can be expected at even shorter wavelength (than 370 nm) not available in current Aethalometer models. Studies showed that woodsmoke enhancement peaks at ~300 nm (Kirchstetter et al., 2004; Kirchstetter and Thatcher, 2012). It is possible that including shorter wavelength in future instrumentation development would improve the sensitivity to woodsmoke PM (Olson et al., 2015). Another approach taking advantage of UV enhancement (or wavelength dependence of the aerosol absorption coefficient in general), as reported by ~~Sandradewi et al. (2008b, a)~~Sandradewi et al. (2008a), derives light absorption Ångström exponents (α) from multi-wavelength Aethalometer readings~~, and then use for quantitative analysis of source contributions to PM~~. α is close to 1 for traffic sources, and varies for woodsmoke, but generally much larger than 1. Assuming certain value of α for woodsmoke, Sandradewi et al. (2008b) conducted quantitative analysis of source contributions to PM. This approach often requires light absorption measurements at multiple wavelengths to have a reliable estimate on α (Chen et al., 2015). ~~Since most of the~~ Sandradewi et al. (2008b) showed that using different pairs of wavelengths led to different values of α for woodsmoke. Since the ambient data to be presented in this paper were collected by a two-wavelength Aethalometer, we did not attempt to calculate α~~.~~. Given the uncertainties associated with values of α for woodsmoke for our study, we did not perform the source apportionment analysis similar to that presented by (Sandradewi et al., 2008a).

Wang et al. (2011) reported a strong correlation between DC and woodsmoke markers ~~including levoglucosan and elemental potassium~~ such as levoglucosan during the heating season, and no statistically significant correlation between DC and vehicle

exhaust markers based on field data collected in Rochester, NY. A follow-up study from the same research group used DC as an input variable in source apportionment models, where it was found to play an important role in separating traffic (especially diesel) emissions from wood combustion emissions (Wang et al., 2012). Allen et al. (2011) adopted DC as woodsmoke marker for their fixed-site measurements in Northern New York State, and revealed temporally and spatially resolved patterns of woodsmoke PM (Fuller et al., 2014). However, Harrison et al. (2013) analyzed data for DC from an Aethalometer network in UK and suggested the presence of other UV absorbing contributors (such as coal burning) to the DC signal. Laboratory experiments conducted by Olson et al. (2015) ~~show~~ showed that besides biomass burning other sources such as uncontrolled coal (e.g., lignite) and kerosene combustion in lamps can also lead to high DC values. In addition, some secondary organic aerosol (SOA) products have also been found to result in UV enhancement (Zhang et al., 2011; Zhong and Jang, 2011), and increase DC responses.

Motivated by the findings from those previous investigations, we aim to further evaluate the applications and limitations of using DC as a qualitative and semi-quantitative woodsmoke marker. Our work is based on recent joint wintertime measurements of $PM_{2.5}$ and light-absorptive PM in woodsmoke-dominated ambient environments and woodsmoke plume environments in three cities/towns located in the Northeastern U.S. Woodsmoke is known to be the major PM source during wintertime, and predominant PM source during winter nighttime, in the three studied cities/towns. Neither heating by coal nor kerosene lamps are common in this region. Furthermore, SOA formation is typically slow during wintertime. Our study can be regarded as a "necessary condition test" for DC serving as a woodsmoke PM marker. In other words, DC would be deemed an inappropriate marker if it were unable to track woodsmoke PM patterns even under woodsmoke-dominated environments. The paper is organized in such a way that we distinguish the ambient and plume environments by discussing their field measurements and results separately, as the potential implications based on the two types of environments are inherently different. Data from multiple locations and different environments contribute to a more robust evaluation on DC.

2 Field Measurements

2.1 Woodsmoke-dominated environments: Ambient (Rutland, Clinton and Lakeview) and Plume (Ithaca)

In this paper, we reported the results from field measurements conducted in four sites in three Northeastern U.S. cities, i.e., Rutland, VT, Saranac Lake, NY and Ithaca, NY. Table 1 describes the general site characteristics.

Rutland is the third largest city in the state of Vermont with a population of 16,500, where residential wood combustion is a major source of winter space heating (Frederick and Jaramillo, 2016)~~and woodsmoke is the dominant PM source in the heating seasons according~~ . According to the 2014 National Emission Inventory~~.~~, residential woood combustion (RWC) contributes to approximately 38.6% of the annual $PM_{2.5}$ emissions in Rutland County. In comparison, on-road mobile sources only account for 1.4%. Considering the seasonal patterns of various emission sources, it is clear that RWC is the predominant primary $PM_{2.5}$ source in Rutland during wintertime. The ambient air quality monitoring site in Rutland (EPA AQS site number: 50-021-0002) is one of very few routine monitoring stations in the U.S. heavily influenced by woodsmoke (http://dec.vermont.gov/air-quality/monitoring/network/rutland). Even though Rutland is not a nonattainment area for annual or 24-hr $PM_{2.5}$ National

Table 1. Descriptions of field measurement sites

Site Name		Environment	Monitoring Method	Operation Period	Site Descriptions
Rutland, VT		Ambient	Fixed-site	October 2011 to June 2013	Co-located with FEM/FRM at AQS 50-021-0002, no nearby woodsmoke sources
Saranac Lake, NY	Clinton	Ambient		December 2014 to April 2015	Located in the backyard of a residential property on Clinton Street, minimal woodsmoke sources
	Lakeview	Ambient		January to April 2015	Located in the backyard of a residential property on Lakeview Street, no nearby woodsmoke sources
Ithaca, NY		Plume	Mobile	December 2015 to March 2016	Right outside the property lines of woodsmoke sources at downwind direction

Ambient Air Quality Standards, its $PM_{2.5}$ design value is among the highest in New England. The next two sites were located in Saranac Lake, a rural town of 5,400 people in Upstate New York~~, where residential wood combustion is the major source of air pollution in winters~~. The 2014 National Emission Inventory indicated that RWC accounts for approximately 22.4 to 25.4% of the annual $PM_{2.5}$ emissions, while the contribution of on-road mobile sources is between 2.8 to 3.9%, which indicated that it is also a woodsmoke-dominated environment during wintertime. Ambient PM concentrations are generally low in Ithaca, the final site and a city of 30,500 in Central New York. While residential wood combustion is not widespread in Ithaca, it has caused localized air pollution hotspots and complaints against woodsmoke were filed by affected residents living in the densely populated neighborhoods. A primary goal for the field measurements in Ithaca was to capture those hotspots. ~~In short, a common feature among those four sites, given the measurement periods and sampling methods (Table 1), was that they were situated in woodsmoke-dominated environments, and most of the PM could be attributed to woodsmoke sources~~It is woodsmoke-dominant in nature as we purposefully sampled woodsmoke plumes.

In short, a common feature for the three cities/towns is that woodsmoke is the predominant PM source during winter nighttime, and the only known major source of DC. Furthermore, the Rutland, Clinton and Lakeview sites represent ambient environments since they captured the mixture of multiple sources, not dominated by any one individual source. By contrast, the mobile monitoring technique employed in Ithaca was designed to capture individual sources, thus, representing plume environments.

Table 2 summarizes the major equipment deployed in the different sites. Detailed descriptions of the experimental methods are provided in Sections 2.2 and 2.3.

Table 2. Descriptions of air quality instruments deployed in various field measurements

Site Names		**$PM_{2.5}$**	**Light-absorptive PM**	**PAH**	**Others**
Rutland, VT		pDR-1500 at 5 min time resolution, 2.5 μm cyclone inlet	AE-21 at 5 min time resolution, 2.5 μm cyclone inlet	N/A	FEM and FRM $PM_{2.5}$ monitors
Saranac Lake, NY	Clinton	pDR-1500 at 1 min time resolution, 2.5 μm cyclone inlet	AE-42 at 1 min time resolution, 2.5 μm cyclone inlet	EcoChem PAS2000 at 30 s time resolution	2-D Sonic Anemometer for wind speed and direction
	Lakeview				
Ithaca, NY		pDR-1500 at 1 s time resolution, 2.5 μ m cyclone inlet	AE-33 1 s time resolution, 2.5 μm cyclone inlet	N/A	CO_2 probe

The Vermont State Department of Environmental Conservation maintains an air quality monitoring site in Rutland, VT (43.608056° N, 72.982778° W; Elevation: 179 m, EPA site number: 50-021-0002). This site is located in the downtown area of Rutland, not adjacent to any known woodsmoke sources. Routine measurements of $PM_{2.5}$, O_3, CO, SO_2, NO, NO_2, VOCs and meteorological variables are conducted.

We deployed a personal DataRAM™ Aerosol Monitor (Model pDR-1500, ThermoFisher Scientific, USA) and a two-wavelength AethalometerTM (370 and 880 nm, Model AE-21, Magee Scientific, USA) for continuous monitoring of $PM_{2.5}$ and Black Carbon (BC), respectively, at the Rutland monitoring site. Operating at 5 min time resolution, both pDR-1500 (1 L min-1, no RH and temperature correction) and AE-21 (2L min-1) were equipped with 2.5 μm sharp-cut cyclone inlets (BGI model SCC 0.732) placed 1.5 m above the roof of a trailer and ambient air was drawn to the instruments through an aluminum sample line. The pDR-1500 was running from December 2011 to April 2012, during which we were able to compare the $PM_{2.5}$ readings from both pDR-1500 and the collocated Federal Equivalent Method (FEM) instrument (BAM 1020, Met One, USA). The AE-21 was in operation from October 2011 to June 11, 2013.

All Aethalometer data were corrected for filter spot optical loading saturation effects (Drinovec et al., 2015; Park et al., 2010; Virkkula et al., 2007) using the "binned" approach, first described by Park et al. (2010), as implemented by version 7.1 of the Aethalometer "data masher" program (Allen et al., 2012). This correction provides a more robust measurement of the DC metric, since the optical attenuation for BC at 370 nm is 2.4 times larger than at 880 nm, resulting in a larger loading artifact at the shorter wavelength. If only BC is present, this results in a negative DC instrument response when the loading is not corrected for.

2.1.1 Saranac Lake, NY (Clinton and Lakeview)

Both sites in Saranac Lake, i.e., Clinton and Lakeview, were located in the backyards of residential properties that did not burn wood for either recreational or heating purposes. Both pDR-1500 (1L min-1, no RH and temperature correction) and AE-42 (2 L min-1) were deployed with the same 2.5 μm sharp-cut cyclone inlets as described in Section 2.2.1, mounted 1.83 m (or 6 feet) above the ground. Both sites were equipped with a 2-D Sonic Anemometer (Model Windsonic, Gill Instruments, UK) for wind speed and direction. In addition, the Lakeview site also included a Photoelectric Aerosol Sensor (Model PAS2000, EcoChem, USA) for continuous particle-bound PAH measurement. The operation periods for the three fixed sites are listed in Table 1.

2.2 Mobile Monitoring at Ithaca, NY

As mentioned earlier, we adopted mobile monitoring techniques in Ithaca, NY to identify air pollution hotspots caused by woodsmoke. The sampling inlet of both pDR-1500 and the seven-wavelength Aethalometer (370 nm, 470 nm, 520 nm, 590 nm, 660 nm, 880 nm, and 950 nm; Model AE-33, Magee Scientific, USA), equipped with 2.5 μm sharp-cut cyclones (BGI SCC 1.197 cyclone at 2.3 L min-1 for pDR-1500; BGI SCC 1.829 cyclone at 5 L min-1 for AE-33), were mounted one foot above the sunroof of a hybrid electric vehicle (HEV). Although the AE-33 employs automated real-time loading compensation (Drinovec et al., 2015), and thus no post data processing was attempted to account for filter loading effect. To account for the filter loading effect, that correction was not used here, since it is not appropriate for mobile monitoring where different combustion sources are sampled in rapid succession. Filter loading was kept relatively low to minimize any loading effects. A flow-through type CO_2 probe (Model CARBOCAP® GMP343, Vaisala, Finland) was connected to the outlet of AE-33 to record the CO_2 level. The pDR-1500 operated without RH correction. RH in the pDR-1500 sensing chamber was always less than 35% without additional sample heating as the instrument was inside a heated vehicle and the chamber temperature was well above ambient dew point. The pDR-1500 was zeroed prior to each mobile run. The pDR-1500 and AE-33 both operated at 1 s time resolution, and the GMP343 at 2 s time resolution to capture individual woodsmoke plumes.

The mobile monitoring occurred periodically from December 2015 to March 2016. Assisted by the weather forecast from New York State Department of Environmental Conservation (NYSDEC) staff, we chose to conduct mobile runs only during low temperature and low wind speed conditions, when the local air quality impacts from woodsmoke were expected to be significant. We made a total of 20 mobile runs (two in December 2015, seven in January, five in February and six in March 2016). The monitoring routes were recorded at 1 s intervals from a Delorme BU-353S4 GPS receiver using Delorme Street Atlas 2015 PLUS software.

At the beginning of the field campaign, we employed the mobile measurements as an efficient way to survey the air quality levels in the Ithaca area, which then enabled us to identify a few recurring hotspots. The rest of the field campaign focused on those recurring hotspots. Specifically, we parked the HEV right outside the property lines of residential woodsmoke sources in the downwind direction, and all instruments were powered primarily by the HEV battery without self-pollution. The internal combustion engine of the HEV occasionally turned on to recharge the battery, and caused brief periods of self-pollution. We

recorded those conditions, generally characterized by high CO_2 and low $PM_{2.5}$ levels, and removed them from subsequent data analysis.

3 Results and Discussions

3.1 Evaluation of pDR against BAM (and maybe FRM)

As mentioned in Section 2.2.1, we collocated a pDR-1500 with BAM 1020, which is a FEM $PM_{2.5}$ sampler, from December 2011 to April 2012 at the Rutland site. Figure 2 illustrates the comparisons of 24-hour average (Figure 2a), nighttime (10 pm to 6 am) average (Figure 2b), hourly (Figure 2c) and hourly nighttime-only (Figure 2d) $PM_{2.5}$ from the two instruments. The main reason to present the nighttime results was that PM during that period almost exclusively came from woodsmoke sources in Rutland. Therefore, Figure 1 not only presents the overall comparisons between the two instruments (Figures 2a and 2c), but also how their readings correlated for woodsmoke-dominated environments (Figures 2b and 2d). Note that the apparent horizontal lines in Figure 2c and Figure 2d result from the 1 μg m^{-3} resolution of the hourly BAM readings.

Table 3 lists the metrics for the regressions. Overall, we found a good agreement between the two instruments. The coefficients of determination, r^2, ranged from 0.895 to 0.960. As expected, the daily and nighttime multi-hour averages (0.956 and 0.960, respectively) showed better correlations than hourly and nighttime hourly averages (0.895 to 0.903, respectively). For the hourly data plots, we observed the BAM noise at the origin where pDR-1500 reads very low and the BAM PM is 2 ± 5 μg m^{-3}. In general, the comparison results gave us confidence in deploying pDR-1500 for other woodsmoke studies.

Table 3. Comparisons between BAM 1020 (y) and pDR-1500 (x) from December 2011 to April 2012 in Rutland, VT. The values inside the parentheses represent the corresponding one standard deviation.

	Regression	r^2
Daily Average	y = 1.082($\pm$ 0.023)·x+2.12($\pm$ 0.33)	0.956
Nighttime average	y = 1.095($\pm$ 0.022)·x+2.04($\pm$ 0.32)	0.960
Hourly average	y = 1.097($\pm$ 0.007)·x+2.63($\pm$ 0.10)	0.895
Nighttime hourly average	y = 1.040($\pm$ 0.011)·x+2.67($\pm$ 0.16)	0.903

The FRM sampler (Model 2025 $PM_{2.5}$ Sequential Air Sampler w/VSCC, R&P, USA) at the Rutland site operates every third day so that we did not include the FRM data in the comparisons. The $PM_{2.5}$ Continuous Monitor Comparability Assessment at the site reported $PM_{2.5}$, FEM = 0.97$PM_{2.5}$, FRM + 1.76 (R=0.97) for Year 2011 and $PM_{2.5}$, FEM = 1.07$PM_{2.5}$, FRM + 0.74 (R=0.92) for Year 2012 (https://www.epa.gov/outdoor-air-quality-data/pm25-continuous-monitor-comparability-assessments).

3.2 DC as a qualitative marker for woodsmoke PM

Figure 3 shows the two-week moving average for DC, BC (880 nm), and $PM_{2.5}$ values measured at the Rutland site from October 2011 to June 2013. DC is strongly linked to the season, with highest values in the winter months and much lower

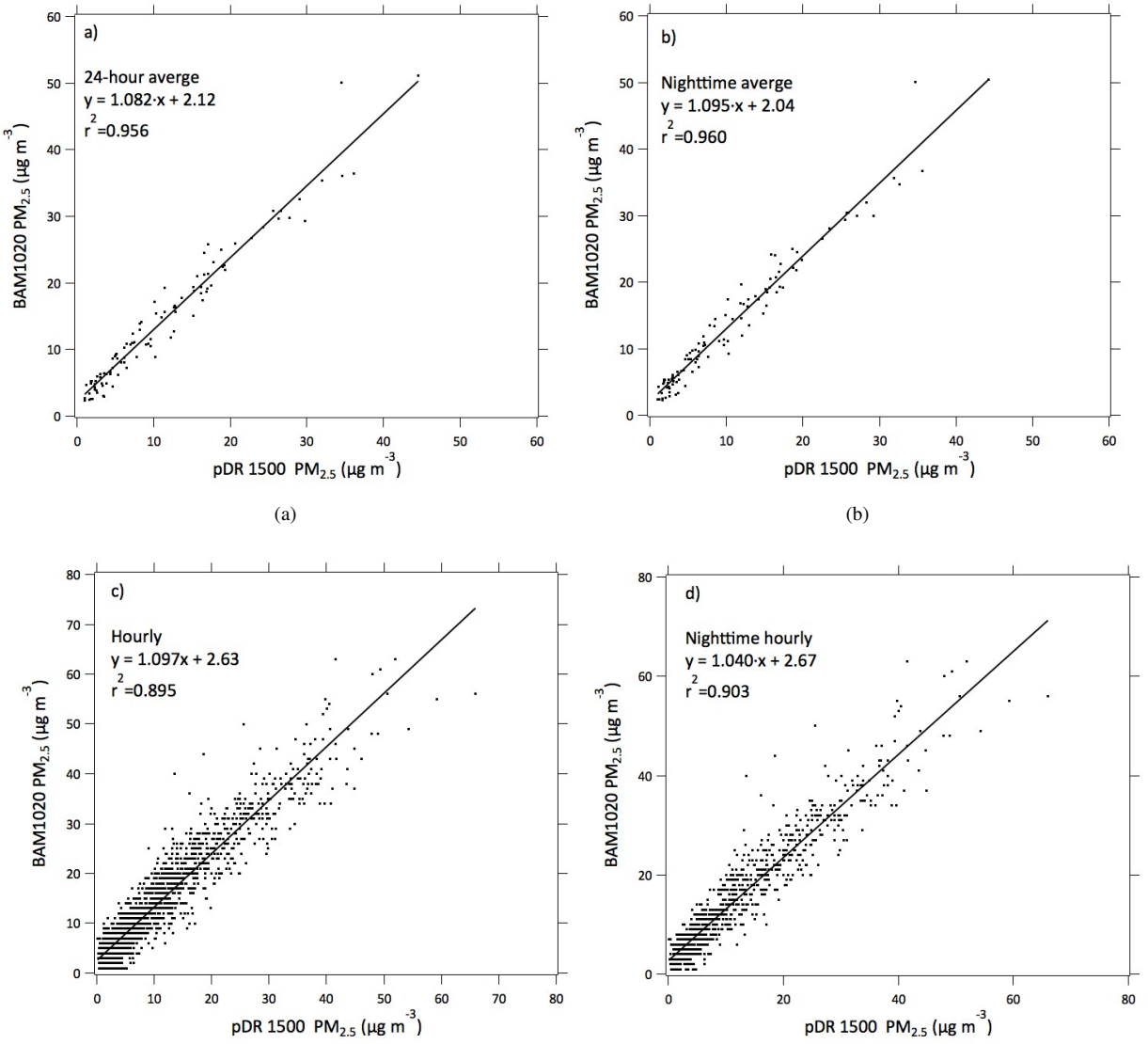


Figure 2. Comparisons between $PM_{2.5}$ values from BAM 1020 (FEM) and pDR-1500 in terms of (a) 24-hour average, (b) Nighttime (10 pm to 6 am) average, (c) hourly average and (d) nighttime hourly average. The apparent horizontal lines in (c) and (d) result from the 1 μg m^{-3} resolution of the hourly BAM readings.

values during the summer months. The summertime DC was close to zero, and the non-zero values could be attributed to Canadian forest fires events typically taking place during summer months (Dreessen et al., 2016; Dutkiewicz et al., 2011) and other recreational biomass burning activities. DC, BC (880nm) and $PM_{2.5}$ all peaked at winter months, when they showed very similar temporal trends. This is as expected since a fraction of woodsmoke PM is BC and woodsmoke sources led to high

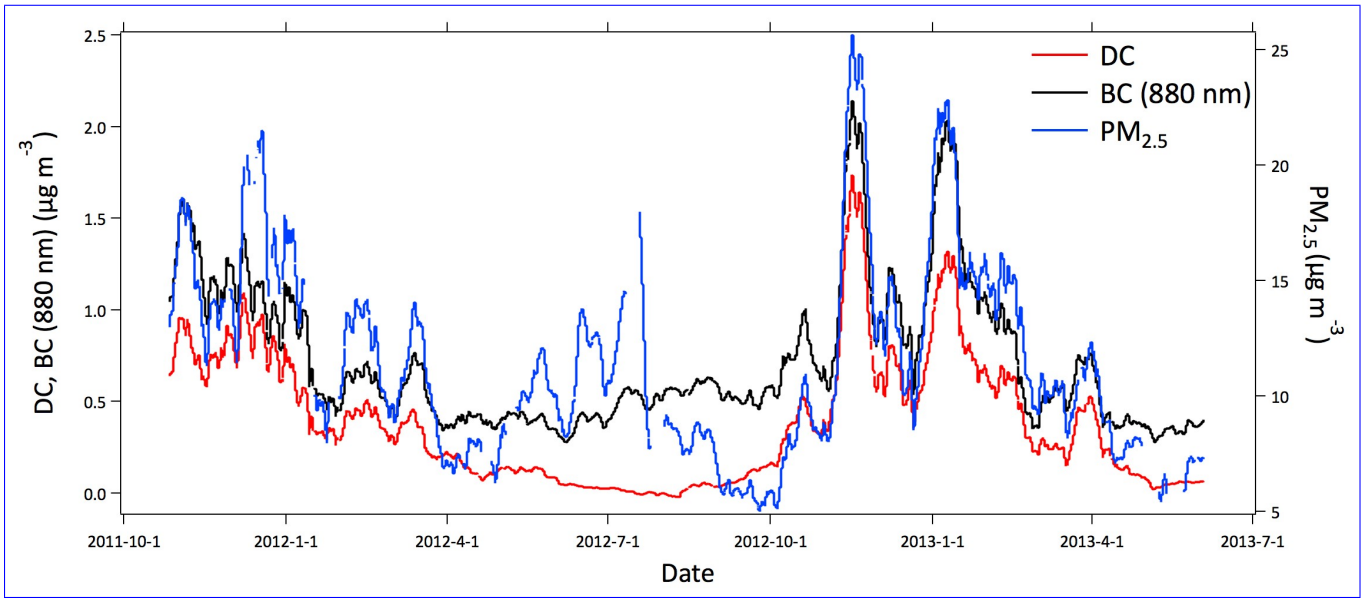
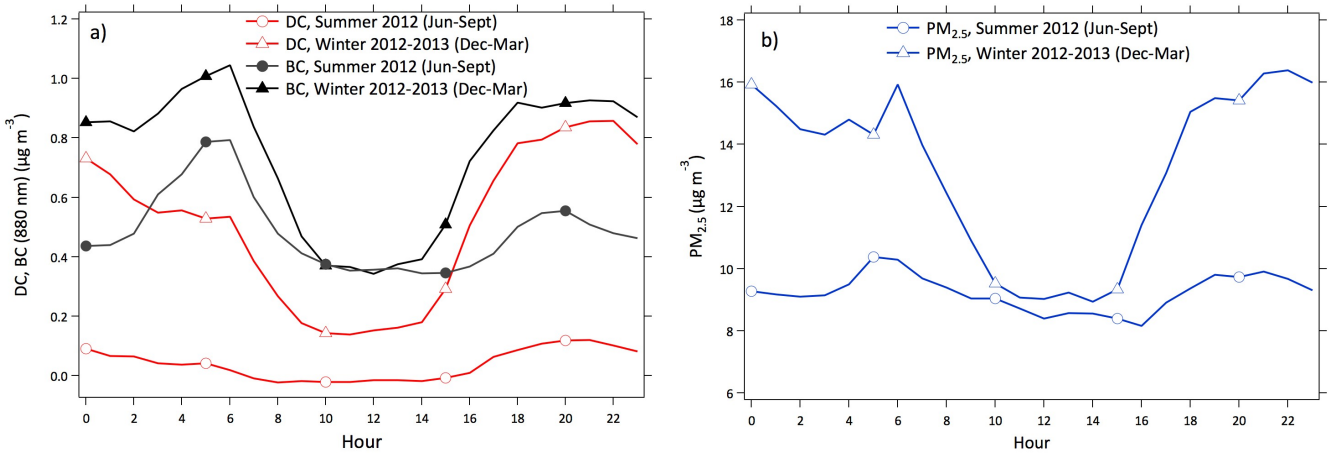


Figure 3. Two-week moving average DC (i.e., BC (370 nm)-BC (880 nm)), BC (880 nm), and $PM_{2.5}$ values measured at the Rutland site from October 2011 to June 2013

$PM_{2.5}$ concentrations in heating seasons. Nevertheless, unlike DC, the concentrations of BC (880nm) and $PM_{2.5}$ were also significant on occasion in the summertime, likely driven by traffic and other emission sources. This comparison supports DC as a qualitative woodsmoke marker.

Figure 4 illustrates the diurnal variations of DC, BC (880 nm), and $PM_{2.5}$ concentrations, for both summer months (July to September 2012) and winter months (December 2012 to March 2013) at Rutland. As expected, DC showed a strong diurnal pattern in the winter months, elevated during nighttime and peaking around 10 pm, and little variation during the summer months. The diurnal patterns of BC (880 nm) persisted over seasons, but driven by woodsmoke sources in the winter months and likely by traffic sources in the summer months. The wintertime $PM_{2.5}$ exhibited a strong diurnal pattern, driven by woodsmoke sources, and less significant but still noticeable diurnal pattern in the summertime, driven by traffic sources, which were not as dominant as woodsmoke sources in Rutland, VT. The nighttime enhancement in pollutant concentrations due to changes in the atmospheric boundary layer also contributed to the diurnal patterns both in summertime and wintertime. This comparison further supports DC as a qualitative woodsmoke marker. As mentioned earlier, previous studies found that SOA products may result in DC signals. ~~However, Figure 4 indicates that SOA is not a main driver for either $PM_{2.5}$ or DC.~~ If SOA formation were significant, we would expect that $PM_{2.5}$ and/or DC would peak around mid-day. The distinct diurnal patterns illustrated in Figure 4 is more consistent with strong influence of local emissions. Moreover, the seasonal trend shown in Figure 3 indicates that DC peaked during wintertime when SOA production is small and approached zero during summertime when SOA production is expected to be high. Therefore, both the diurnal and seasonal patterns indicate that SOA is not a main driver for DC in Rutland.



(a) (b)

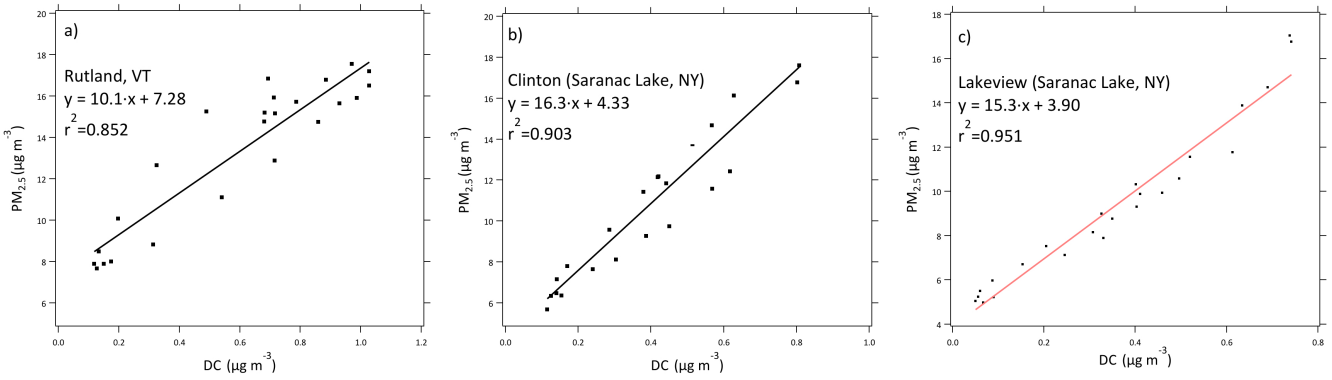
Figure 4. Diurnal plots (i.e., averaged into 24 hours) of a) DC (i.e., BC (370 nm)-BC (880 nm)) and BC (880 nm), and b) $PM_{2.5}$ values measured at the Rutland site from October 2011 to June 2013.

3.3 DC as a semi-quantitative marker for woodsmoke PM

Under woodsmoke-dominated environments we were studying, woodsmoke is the leading source of $PM_{2.5}$. Thus, we explored in this section the relationships between measured $PM_{2.5}$ and DC to assess whether DC can be used as semi-quantitative predictor of woodsmoke $PM_{2.5}$, for both ambient and plume environments. We used the terms "semi-quantitative" for two reasons. One is that both highly time-resolved $PM_{2.5}$ and BC measurements contains significant uncertainties. The other reason is that the DC cannot be quantitatively interpreted as an exact amount of a specific compound unless the mixture of UV-absorbing species remains constant enough and an average absorption cross-section can be assumed.

3.3.1 Ambient environments (Rutland, Clinton and Lakeview)

Figure 5 depicts $PM_{2.5}$ vs DC for the three ambient sites, where we averaged all the hourly data, binned by hours of the day (i.e., 24 data points), over the wintertime operation periods for Rutland and over the entire operation periods for Clinton and Lakeview, respectively. The slopes derived from the linear regressions represent Δ(Ambient $PM_{2.5}$)/ΔDC. Table 4 presents the linear regression results with all correlation coefficients of determination exceeding 0.85, which indicates strong positive correlations between changes in DC and changes in ambient $PM_{2.5}$ changes at the three sites. The most plausible explanation is that DC ~~represents the~~ is an indicator of woodsmoke PM, which typically have a strongly diurnal pattern, considering that wood burning and traffic are the only two major local PM emissions sources, and that wood burning is ~~the only~~ typically the dominant source of DC ~~.~~ in ambient atmosphere. DC signals only occur in the presence of wood burning. Furthermore, Figure 5 suggests that averaging stationary PM and BC data over a long period of time (e.g., over a winter month or longer in



(a) (b) (c)

Figure 5. Diurnal $PM_{2.5}$ vs DC (i.e., BC (370 nm)-BC (880 nm)) averaged over the wintertime operation periods for a) the Rutland site, and over the entire operation periods for b) the Clinton site and c) the Lakeview site, respectively, into 24 hours.

a fixed location) may lead to an average absorption cross section, i.e., a constant Δ(Ambient $PM_{2.5}$)/ΔDC, even though PM composition and the resulting absorption cross section may vary with time.

Furthermore, the regression coefficients for Clinton and Lakeview, the two ambient sites in Saranac Lake, NY, were very similar, suggesting that the Δ(Ambient $PM_{2.5}$)/ΔDC is reproducible for similar ambient environments. However, the same relationship did not hold for the different environment of Rutland. The inclusion of two heating seasons for the Rutland site, compared to one season in Clinton and Lakeview, may have also contributed to the discrepancy.

Table 4. Semi-quantitative relationship between ~~Delta-C~~ DC (μg m^{-3}) and $PM_{2.5}$ (μg m^{-3}) in woodsmoke dominated ambient environments. The values inside the parentheses represent the corresponding one standard deviation.

Site		**Regression**	r^2
Rutland, VT		$PM_{2.5}$ = 10.1($\pm$0.90)·DC+7.28($\pm$0.60)	0.852
Saranac Lake, NY	Clinton	$PM_{2.5}$ = 16.3($\pm$1.14)·DC+4.33($\pm$0.52)	0.903
	Lakeview	$PM_{2.5}$ = 15.3($\pm$0.74)·DC+3.85($\pm$0.31)	0.951

3.3.2 Plume environments (Ithaca)

Figure 6 presents the $PM_{2.5}$-DC relationships from two reoccurring woodsmoke sources based on the plume measurements, reported as 5-second moving averages, that were conducted in Ithaca, NY. Figure 6a-6d, and Figure 6e-6f characterized two sources different days, respectively. Both sources were woodstoves as the configurations of the exterior stacks were consistent with this type of heating equipment. We estimated the background $PM_{2.5}$ concentrations for each day, and the values were $\sim$ 3 μg m^{-3}. Thus, we only included data points with $PM_{2.5}$ concentrations larger than 5 μg m^{-3} in Figure 6 in order to capture

the plume signals. The slopes derived from the linear regressions represent Δ(Woodsmoke $PM_{2.5}$)/ΔDC, as we conducted sampling in woodsmoke plume environments.

Overall, we observed a dominant set of correlated measurements, likely representing the average woodstove combustion conditions, on each day. On both Figure 6c and 6f, "Condition 2" marked data points that define a different correlation are plotted with different symbols and a separate regression line. Each "Condition 2" line consisted of plume data recorded continuously. Possibly, during those conditions the woodstove combustion had been disturbed for some reasons (such as reloading the stove) for both Sources 1 and 2, thus significant deviation from the average conditions (denoted as "Condition 1" on both Figure 6c and 6f). For both Conditions 1 and Conditions 2, the correlations are generally strong. PM vs. DC slopes vary significantly for individual sources (from 3 to 9.6 for Source 1, and from 7.4 to 28.6 for Source 2). Even for the same source, the slopes can change considerably during different operating conditions. Our analysis also suggests that the $PM_{2.5}$-DC relationships can be potentially utilized to distinguish different combustion and operating conditions of woodsmoke sources. It is expected that cleaner burns would have a larger slope—, i.e., less organic aerosol per unit woodsmoke PM (Chandrasekaran et al., 2011, 2013). In other words, the different combustion conditions lead to different chemical compositions and absorption cross sections, which can be potentially captured by high time resolution light absorption measurements. However, further studies are needed to link the $PM_{2.5}$-DC relationships to specific conditions.

3.3.3 DC and Heating degree days

Heating degree days (HDD), counted as the number of degrees that the daily average ambient temperature (F) is below 65°F, have been shown to be a better way to estimate energy use for space heating than actual temperature, as most homes or facilities are maintained at a temperature above 65°F. In a woodsmoke-dominated environment, we expected more woodsmoke with higher HDD.

We calculated the monthly average HDD for Rutland using the temperature data recorded at the weather station located in the Rutland-Southern Vermont Regional Airport (KRUT). In our analysis, DC/BC was adopted as a semi-quantitative woodsmoke emission indicator.

The rationale to use DC/BC, rather than DC directly, was to take BC as a dilution indicator to normalize DC. Even though the absolute values of DC change with meteorological conditions, DC/BC should be driven by the amount of woodsmoke PM emissions generated, not woodsmoke PM concentrations.

Figure 7 illustrates the relationship between DC/BC and HDD, both presented as monthly averaged values. We observed a relatively strong linear relationship between DC/BC, which is an indicator for woodsmoke PM emissions, and HDD, which is a surrogate for space heating energy use. In other words, Figure 7 reveals not only a qualitative relationship (i.e., colder the weather, the more woodsmoke PM), but also a potentially semi-quantitative relationship linking space heating energy and woodsmoke PM emissions. Note that the proportionality between DC/BC and HDD will vary from place to place, depending on various factors such as fraction of heating obtained from biomass, and types of biomass fuels burned.

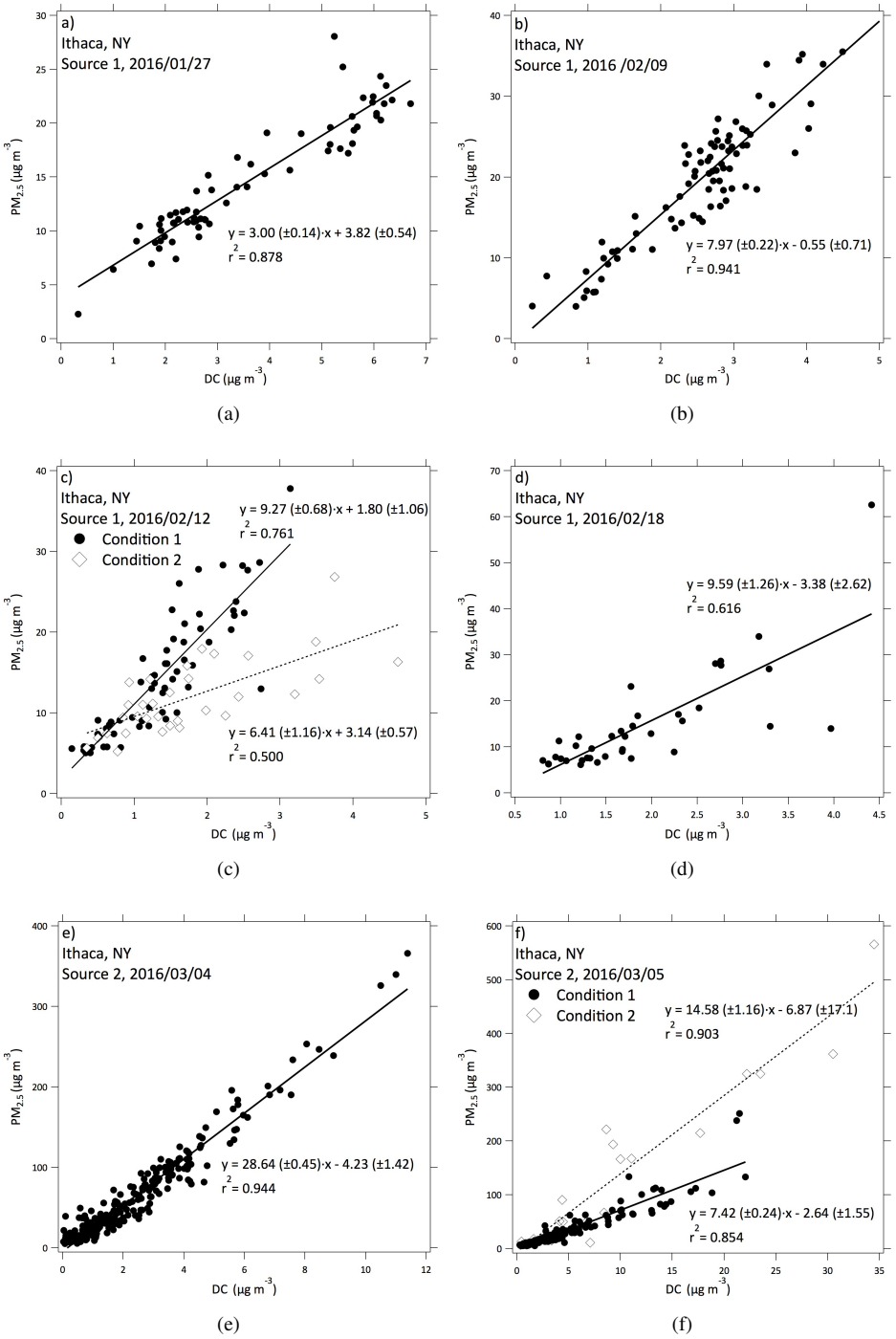


Figure 6. PM2.5 vs. DC relationships from two reoccurring woodsmoke sources based on the plume measurements conducted in Ithaca, NY. Data are reported as 5-second averages. The dates are expressed in YYYY/MM/DD. The values inside the parentheses represent the corresponding one standard deviation.

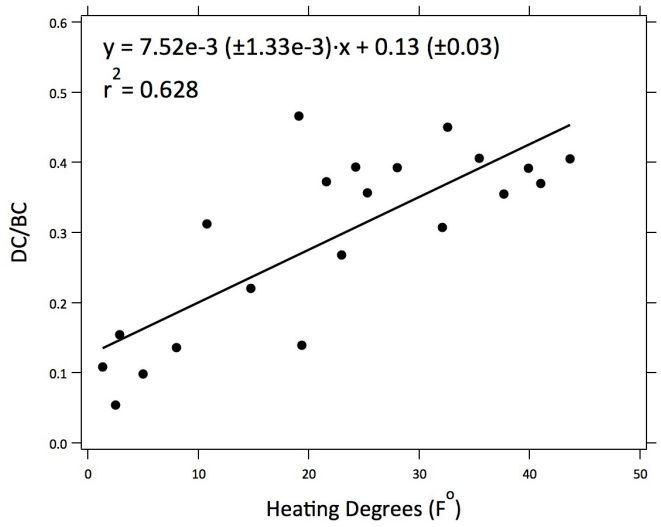


Figure 7. The relationship between DC/BC and HDD, both presented as monthly averaged values based on Rutland data. DC/BC is proposed as a woodsmoke PM emission indicator. The values inside the parentheses represent the corresponding one standard deviation.

3.3.4 ~~Comparison between PAH and DC as a potential woodsmoke marker~~

~~Woodsmoke is known to contain polycyclic aromatic hydrocarbons (PAH)(Weimer et al., 2008), and large molecular weight PAHs have been suggested to contribute to light absorption by organic carbon in wood combustion (Chen and Bond, 2010). The simultaneous measurements of DC, $PM_{2.5}$ and PAH allowed us to conduct a preliminary comparison between particle-bound PAH as measured by the Ecochem PAS2000 and DC as a potential woodsmoke marker.~~

~~]]Diurnal average PM2.5 vs. PAH measured by EcoCHEM PAS2000 for a) the Clinton and b) Lakeview sites. We averaged the hourly $PM_{2.5}$ and PAH data over the entire operation periods at the two sites into 24 hours, respectively. The values inside the parentheses represent the corresponding one standard deviation.~~

~~Figure ?? presents the correlations between diurnal average $PM_{2.5}$ and PAH for the Clinton (Figure ??) and Lakeview (Figure ??), compared to the similar plots for $PM_{2.5}$ vs. DC at the same sites (Figure 5b for Clinton and Figure 5c for Lakeview, respectively). Using the same method applied to Figure 5, we averaged the hourly $PM_{2.5}$ and PAH data over the entire operation periods into 24 hours. Overall DC was highly correlated with $PM_{2.5}$ (r^2 0.9 for Clinton and 0.95 for Lakeview, respectively), whereas the correlation was much lower between $PM_{2.5}$ and particle-bound PAH much closer than PAH (r^2 0.57 for Clinton and 0.27 for Lakeview, respectively). The preliminary comparison suggests that DC is a better woodsmoke indicator than PAH. However, this finding was inconclusive since PAHs contain a large number of individual components, and what we have shown here were those measured by the particular instrument we deployed (i.e., EcoCHEM PAS2000), which may not capture the large molecular weight PAHs.~~

4 Conclusions

We presented the results from the joint wintertime measurements of $PM_{2.5}$ and light-absorptive PM in woodsmoke-dominated ambient and plume environments in three Northeastern U.S. cities/towns, where other types of sources contributing to DC such as uncontrolled coal and kerosene burnings are usually rare. Our main conclusion is that DC can be a useful woodsmoke PM marker, both qualitatively and semi-quantitatively.

As a qualitative marker, DC can track the diurnal and seasonal woodsmoke PM patterns~~.~~, approaching zero in the summertime, reaching highest values in the wintertime, and peaking at winter nights.

As a semi-quantitative marker, ~~DC can be used to estimate the amounts of woodsmoke PM~~we showed strong linear relationships between $PM_{2.5}$ and DC in the ambient environments, and the resulting nearly constant Δ(Ambient PM2.5)/ΔDC values can be potentially estimate woodsmoke contributions to $PM_{2.5}$. $PM_{2.5}$ vs DC relationship has been shown to be reproducible for similar ambient environments (like the Clinton and Lakeview sites in Saranac Lake, NY)~~, but~~. Nevertheless, the same relationship did not hold for the different environment (like Rutland, VT). In other words, the relationship depends on the environment and combustion conditions.

This paper also presented several other potentially interesting findings: the $PM_{2.5}$-DC relationships can be utilized to distinguish different combustion and operating conditions of woodsmoke sources; the semi-quantitative relationship between DC vs. HDD could link space heating energy and woodsmoke PM emissions~~; DC tracks woodsmoke PM better than PAH measured by EcoCHEM PAS2000~~. Those findings could have important implications and applications in air quality management. However, as elaborated in the paper, further studies are needed to elucidate those findings.

Competing interests. There are no competing interests.

Acknowledgements. The authors acknowledge funding support from the New York State Energy Research and Development Authority (NYSERDA) contracts #32974, 63035 and 63036, and appreciate the assistance of Aleshka Carrion-Matta, Neng Ji and Ye Lin Kim at Cornell University with conducting the field measurements. The New York State Department of Environmental Conservation provided forecasting support for mobile measurements, and the authors thank Robert Gaza, John Kent and Julia Stuart for their kind assistance. The authors also thank Magee Scientific for loaning the Aethalometer Model AE-33 employed in the field measurements.

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