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Collection of ambient particulate matter by porous vegetation barriers: Sampling and characterization methods

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Abstract

Sampling and characterization methods for assessing the effect of vegetative barriers on particulate matter (PM_{10}) concentrations and functional group composition were developed and applied in a case study. Ambient PM_{10} was concurrently sampled upwind and downwind of a hawthorn hedge at a rural location in the UK. Fourier transform infrared (FTIR) spectra of PM_{10} samples were collected to characterize the functional group composition. Absorbances associated with sulfate, nitrate, ammonium, aliphatic carbon-hydrogen, and carbonyl functional groups were observed in the FTIR spectra. Calculations with gravimetric measurements show that the hedge collects PM_{10} mass with a collection efficiency of 34% on average. FTIR results suggest that individual functional groups might exhibit different behavior in the hedge, but further method development and sampling is necessary to calculate functional group results with more confidence. Current results show the potential of using hedges to mitigate ambient concentrations of airborne PM_{10}, and applying these methods to a more statistically robust sample size is anticipated to aid in elucidating physico-chemical mechanisms driving collection of PM_{10} by hedge elements.
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1. Introduction

Widespread agricultural practices, specifically in the rural areas of several European countries, produce large emissions of ammonia (Theobald et al., 2001). Ammonia undergoes atmospheric photochemistry along with nitrogen oxides to form ammonium nitrate aerosols in anywhere from 1 to 3 h, providing a vigorous production mechanism for substantial concentrations of ambient particulate matter (PM) (Erisman, 1989; Seinfeld & Pandis, 1998). Transport and deposition of such nitrogen-rich aerosols result in ecosystem eutrophication and soil acidification (Fangmeier, Hadwiger-Fangmeier, Van der Eerden, & Jaeger, 1994; Theobald et al., 2001). Eutrophication can lead to out-competition of oligotrophic plants by nitrophilous plants, resulting in a loss of biodiversity at the species level (Sutton, Pitcairn, & Fowler, 1993). In addition, ambient PM with an aerodynamic diameter smaller than 10 μm (PM_{10}), comprised of

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both inorganic and organic components, is known to be associated with cardiovascular and respiratory morbidity and mortality (Klemm, Mason, Heilig, Neas, & Dockery, 2000; Pope, Dockery, & Schwartz, 1995), particularly among elderly individuals (Zelikoff et al., 2003). Numerous toxicological links between health effects and inorganic PM components such as sulfates and metals have been made (USEPA, 2004), and direct links to both the polar and non-polar fractions of organic aerosol have been observed recently (Reyes, Rosario, Rodriguez, & Jimenez, 2000). Investigations of cost-effective methods to reduce ambient PM concentrations are thus warranted in order to develop strategies for mitigating both human exposure and ecological damage resulting from ambient PM_{10} concentrations.

Recent investigations have explored the ability of porous vegetation barriers to mitigate exposures to high PM concentrations (Dierickx, 2003; Raupach, Woods, Dorr, Leys, & Cleugh, 2001; Tiwary, Morvan, & Colls, 2006). The collection of aerosol by hedge elements was attributed to both the surface roughness and to the creation of tortuous air flows. Tortuous air flows lead to higher turbulence and increased mixing of PM, and surface roughness increases the likelihood of particles impacting on their foliage surfaces. Vegetative barriers surrounding farmland, animal housing, and urban dwellings tend to reduce dispersal of PM through these near-ground particle deposition mechanisms. In addition, the Natural Resources Conservation Service of the United States recommends the use of filter strips, such as shelter belts, to reduce the movement of pathogens, particulate organic sediment and adsorbed nutrients, pesticides and other possibly toxic particulate materials into human microenvironments (NRCS, 2002). However, the physical and chemical dynamics involved in capturing ambient PM with foliage surfaces is complex and currently not well characterized (Tiwary et al., 2006). Flux measurements of individual particulate species through porous vegetation strips such as hedges will enhance their usefulness as tools to consider when designing effective PM exposure mitigation strategies.

In this work we have developed sampling and analytical methods for empirically characterizing the ability of a hawthorn hedge (the most abundant hedge species in the UK (NERC, 2000)) to filter PM_{10} from ambient air. Gravimetric analysis of PM_{10} samples was used to estimate the collection efficiency. Effects of the hedge on PM_{10} functional group composition was explored using Fourier Transform Infrared (FTIR) spectroscopy. Functional group analysis has been performed on ambient PM samples using FTIR in a number of earlier studies, and the technique has provided composition-dependent, average properties that influence the behavior, activity, and environmental fate of PM (Allen, Pallen, Haimov, Hering, & Young, 1994; Blando et al., 1998; Garnes & Allen, 2002; Maria, Russell, Turpin, & Porcja, 2002; Maria et al., 2003; Mylonas, Allen, Ehrman, & Pratsinis, 1991; Pickle, Allen, & Pratsinis, 1990; Reff et al., 2005, 2007; Russell, 2003; Shaka & Saliba, 2004). FTIR is thus potentially useful for exploring the effects of hedge barriers on bulk aerosol properties relevant to the aforementioned health and ecological issues (e.g. polarity and ammonium content) that depend on the bulk composition of ambient PM_{10}.

2. Experimental

2.1. Field sample collection

The sampling site in this study was a rural location in the UK near the Meteorological Office's automatic weather station on the University of Nottingham's Sutton Bonington campus (Location: Lat 52.82°N, Long 1.25°W and 48 m above mean sea level). Fig. 1 shows a schematic of the sampling site. Three pairs of 36 h PM_{10} samples were concurrently collected upwind and downwind of the hawthorn hedge on 47 mm Teflon® filters (Whatman®, Part # 7592-104) during autumn of 2005. Upwind and downwind PM samplers were identical (MiniVol™, AirMetrics, Oregon, USA) and used 10 μm cut point impactors. Filters were prepared for sampling by desiccating for 2 h at 55 °C, followed by equilibration and pre-sampling gravimetric analysis (see below). Afterwards, filters were wrapped in aluminium foil and stored in a freezer until removal for field sampling. Each sample was collected over three consecutive days during daylight hours (07:00–19:00 hrs) only. Upwind and downwind instruments were randomly assigned between samples to minimize potential bias in sampling by any particular sampler. Inlet nozzles were positioned at two-thirds of the hedge height since this region is found to be representative of filtration properties for the entire stand (Tiwary et al., 2006). Flow control circuits maintained a flow rate of 5 L min^{-1} by varying the speed of the pump as PM accumulated on the filters. Sample volumes were obtained by correcting the instrument flow rate for the temperature and pressure differences from factory calibration conditions (294 °K at 0.99 atm) as recommended by the instrument manufacturers. The moderate flow rate was chosen to minimize volatilization of organic PM (Turpin, Hering, & Huntzicker, 1994). An impaction plate was placed upstream of the Teflon filter to remove particles with an aerodynamic diameter greater than 10 μm,

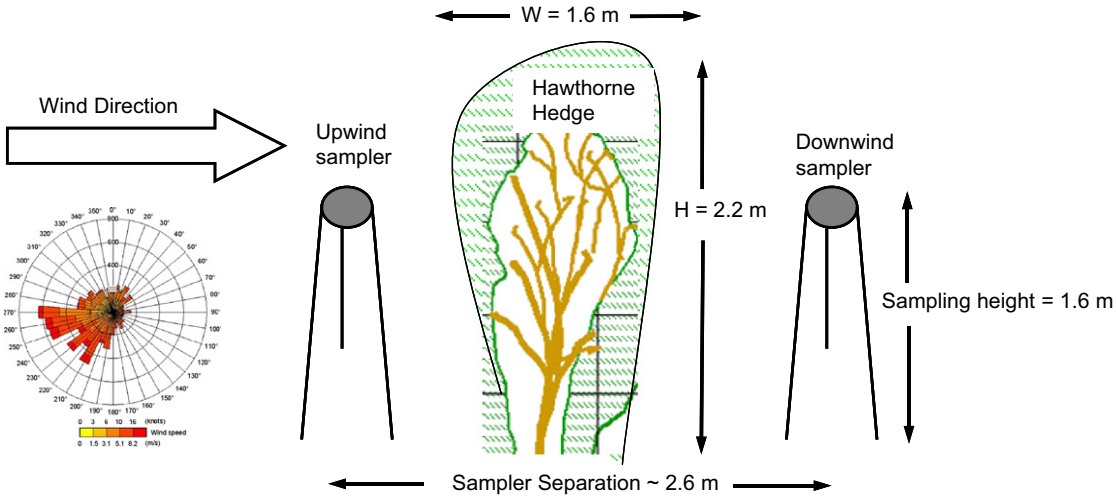

Fig. 1. Schematic diagram of the sampling equipment relative to the hawthorn hedge. The wind rose for the sampling period is displayed in the lower left-hand corner.

Table 1
Average meteorological conditions at the field site during sampling periods

Experiment	Total irradiance (kJ/m^2)	Air temperature (°C)	Wind speed ($m\,s^{-1}$)	Wind direction (deg)	% of wind measurements within 20% of normal	Max deviation of wind from normal (deg)
Exp. 1 (28 September–30 September)	847	15.6	4.8	230	79	60
Exp. 2 (04 October–06 October)	279	13.1	1.8	228	67	85
Exp. 3 (11 October–13 October)	561	17.6	4.4	268	83	55

which was coated with the hydrocarbon-based Apiezon® AP100 grease to minimize particle bounce. The possibility of sample contamination by either volatilization or splatter of the Apiezon grease was assumed to be very small due to the fact that the greases are designed to have very low vapor pressures ($< 10^{-9}$ Torr) and resistance to creep (Apiezon, 2005). After collection, samples were taken directly to the analytical laboratory, where gravimetric, XRF, and FTIR analyses were performed sequentially. A field blank was also generated for each sample collected during the study to monitor for sample contamination during transport and analysis. Field blanks were filters that were transported to and from the field in filter cases with the samples and underwent the same gravimetric and FTIR analyses, but were not used to sample ambient PM.

Average meteorological parameters for the site during the sampling period were calculated from data in an online database (http://www.metoffice.gov.uk/climate/uk/stationdata/) and are presented in Table 1. The prevailing wind direction during sampling days was SW (between 225° and 270°). The strip of hawthorn hedge under study was approximately perpendicular to this range of wind directions (Fig. 1), which ensured a uniform flow of air through the hedge that was perpendicular to the hedge faces.

2.2. Gravimetric analysis

Teflon filter samples were gravimetrically analyzed both before and after sampling using a microbalance (MC5, Sartorius AG, Goettingen, Germany) to determine the PM_{10} mass concentrations. The microbalance auto-calibrated either every 4 h or when the temperature changed by 1 °C. Filters were equilibrated between 21 and 23 °C, and between 40% and 45% relative humidity (RH) for 24 h in an air-conditioned weighing room before and after aerosol collection. Post-sampling weighing conditions were within 5% RH and 2 °C of the pre-sampling weighing conditions. A random subset of five blank filters was weighed over three consecutive days to calculate a gravimetric precision of 0.98. The

detection limit (DL) of the balance was determined by weighing five randomly selected laboratory blank filters 5 times each. Three times the σ_{pooled} of these filters yielded a DL value of 0.0001 mg (Carlton & Teitz, 2002). Effects of static electricity were minimized using an α-particle source and an ionizing blower (Staticmaster® Model BF2-1000).

2.3. FTIR analysis

2.3.1. Field sample preparation

PM was extracted from the Teflon filters prior to FTIR analysis to remove the strong influence of the Teflon filter from the FTIR spectrum of the PM samples (Reff et al., 2007 and references therein). A previously published extraction method (Harrison, Jones, & Lawrence, 2004) was followed to prepare samples for FTIR analysis. Each filter sample was oriented at 45° in a Petri dish and briefly rinsed with 0.3 mL of 2-propanol to wash off the non-polar deposits. Afterwards, polar PM constituents were extracted by consecutive rinsing with distilled de-ionized water (10 mL) followed by gentle shaking in the Petri dish for about 20–25 min. A few drops of the liquid extract were then placed between two ZnSe disks to create a liquid film. The ZnSe disks were placed in a mount and analyzed by the FTIR instrument.

2.3.2. FTIR instrumentation

Transmission sample spectra were collected using a Nicolet FTIR spectrometer (Protégé 460 Magna Infrared Spectrometer, Magna-IR® Tech.) equipped with a KBr beam splitter, a liquid nitrogen-cooled mercury cadmium telluride (MCT/A) detector, and a horizontal attenuated total reflectance (ATR) unit comprised of a ZnSe crystal with 12 internal reflections (refractive index = 2.4). Two hundred transmission scans of each filter were collected from 450 to 4000 cm^{-1} at a resolution of 4 cm^{-1} and averaged into a final sample spectrum by the instrument. The spectrometer was continually purged with dry air to keep the optical components free of water vapor. Every day, the instrument bench was reset to maintain an energy throughput (peak-to-peak ratio) of at least 4.2 V. Instrumental background spectra were collected by scanning the solvent used to prepare the liquid films (0.3 mL 2-propanol and 10 mL de-ionized water). The software provided with the instrument (Omnic 3.1, Nicolet Instrument Corp.) was used to convert the transmission spectra into the corresponding absorbance spectra. A baseline correction algorithm provided in the software was then applied to resulting absorbance spectra. Final sample spectra were obtained by subtracting the spectra of field blanks from the spectra of corresponding sample blanks to remove impurities introduced to field samples during transport and analysis.

3. Results and discussion

3.1. PM_{10} Concentrations and collection efficiencies

Gravimetrically determined PM_{10} concentrations of each sample collected upwind and downwind of the hawthorn hedge are presented in Table 2. All PM_{10} measurements were greater than DTs. The removal of particles by a hedge was parameterized by the filtration collection efficiency ($CE_{\text{Filtration}}$):

$$CE_{\text{Filtration}} = \frac{C_{\text{In}} - C_{\text{Out}}}{C_{\text{In}}}, \tag{1}$$

where C_{In} and C_{Out} are the particle concentrations (μg/m^3) at the upwind and the downwind faces of the hedge, respectively. $CE_{\text{Filtration}}$ values for PM_{10} calculated from measurements in this work are also shown in Table 2, which were calculated by applying Eq. (1) to the upwind and downwind concentrations in the same table. The average $CE_{\text{Filtration}}$

Table 2
PM_{10} concentrations measured upwind and downwind of the hawthorn hedge and corresponding collection efficiencies

Experiment	Upwind concentration (μg/m^3)	Downwind concentration (μg/m^3)	$CE_{\text{Filtration}}$ (%)
1	36.2 ± 12.4	22.3 ± 9.3	38.1
2	22.1 ± 10.5	15.4 ± 7.3	30.4
3	34.2 ± 9.6	22.8 ± 11.2	33.3

of PM_{10} across all three sampling periods is about 34%. To confirm that the observed reductions in PM_{10} concentrations are due to the presence of the hedge, $CE_{Filtration}$ values were calculated using particle number concentrations measured to evaluate the results of an earlier simulation study (Tiwary et al., 2006). In that experimental setup, optical counters at the same sampling site as the hawthorn hedge were arranged in the same manner as the samplers in Fig. 1 but without the hedge between them, and readings were taken every minute during a single afternoon. The $CE_{Filtration}$ of the PM_{10} concentrations from these samplers were found to be $-3\% \pm 10\%$ ($n = 150$). Although a somewhat smaller $CE_{Filtration}$ value is expected for particle number concentration vs. the same value from mass based concentrations (Hinds, 1982), the value from the control experiment is significantly lower than both the values in Table 2 and the value of 25.6% determined in the hawthorn hedge simulation study (Tiwary et al., 2006). This strongly suggests that the hedge is collecting PM_{10} and upwind–downwind differences are not due to ambient concentration gradients.

3.2. Functional group composition

Figs. 2a–c show FTIR spectra of upwind and downwind PM_{10} samples concurrently collected during the sampling experiments in this study. Functional groups associated with these infrared absorbances were identified by consulting previously published FTIR analyses of ambient PM (Allen et al., 1994; Blando et al., 1998; Carlton et al., 1999; Maria et al., 2002; Reff et al., 2005). Absorbances associated with particulate sulphate (near 618 and 1110 cm^{-1}), ammonium (2900–3200, 1430 cm^{-1}), inorganic nitrate (835 cm^{-1}), hydroxyl (3200–3500 cm^{-1}) aliphatic carbon (2920 and 2850 cm^{-1}) and carbonyl (1650–1800 cm^{-1}) functional groups are indicated in Fig. 2. The large downward peaks near 2300 cm^{-1} in Figs. 2a–c are due to the presence of CO_2 in the FTIR instrument, but did not interfere with any signal from the PM samples.

Some effects of the hawthorn hedge on the PM_{10} inorganic functional group composition may be seen in Figs. 2a–c by comparing the upwind and downwind spectra. In Experiments 1 and 3 (Figs. 2a and c) there is a clear diminishment in the strength of absorbances associated with sulphate (the 1110 cm^{-1} absorbance) and ammonium as the particles move through the hedge from the upwind to the downwind side. Absorbances for these functional groups have no clear presence in the FTIR spectra of Experiment 2 samples (Fig. 2b). The lack of these absorbances in Experiment 2 samples might also be due to less photochemical production of ammonium sulphate (Seinfeld & Pandis, 1998) resulting from the substantially lower sunlight irradiance (Table 1) for Experiment 2 than for Experiments 1 and 3. However, the relatively smaller absorbances near 835 and 618 cm^{-1} typically associated with inorganic nitrate and sulphate are more difficult to distinguish in all spectra than in FTIR spectra of PM samples from earlier studies (Allen et al., 1994; Blando et al., 1998; Carlton et al., 1999). The lack of the small sulphate and nitrate absorbances and the negative absorbances in Experiment 2 all illustrate the difficulties encountered by the authors in removing the influence of the solvent and background contamination to produce a meaningful sample spectrum.

The absorbances associated with carbonyl functional group also show a large decrease similar to those of ammonium and sulfate in Experiment 1, but upwind and downwind absorbances of carbonyl are similar in Experiment 3. Carbonyl absorbances have a much more substantial presence than ammonium and sulfate in Experiment 2. This appears to contradict the aforementioned hypothesis of decreased photochemistry for Experiment 2, and might be a further indication of spectral background removal issues. An alternative possibility is that lack of photochemistry at the sampling site did diminish the presence of ammonium and sulfate groups, and that the carbonyl absorbances are due to transport of aged, oxidized organic particles (Robinson, Donahue, & Rogge, 2006).

Aliphatic carbon has no obvious absorbances in Experiment 1, and the background subtraction difficulties makes interpretation of the aliphatic absorbances difficult for Experiment 2 due to the large spurious downward peaks on the left shoulder of those absorbances. In Experiment 3, absorbances that can be clearly distinguished as aliphatic carbon can only be seen in the downwind sample. One possible explanation for the increase in the downwind sample is the higher ambient temperatures present during sampling for Experiment 3 (Table 1), which might increase the release of hydrocarbon-rich vegetative material (Tarvainen et al., 2005) into the air stream followed by sorption onto particles as they move through the hedge to the downwind sampler. The weak presence of aliphatic carbon absorbances in the sample spectra does provide assurance that little to no contamination by the Apiezon grease on the impactor plates in the field samplers took place.

Strong absorbances in the hydroxyl region of the spectrum are present only in Experiment 2. The size of this peak relative to the rest of the spectrum is extremely large compared to what might be expected from other PM studies (Allen

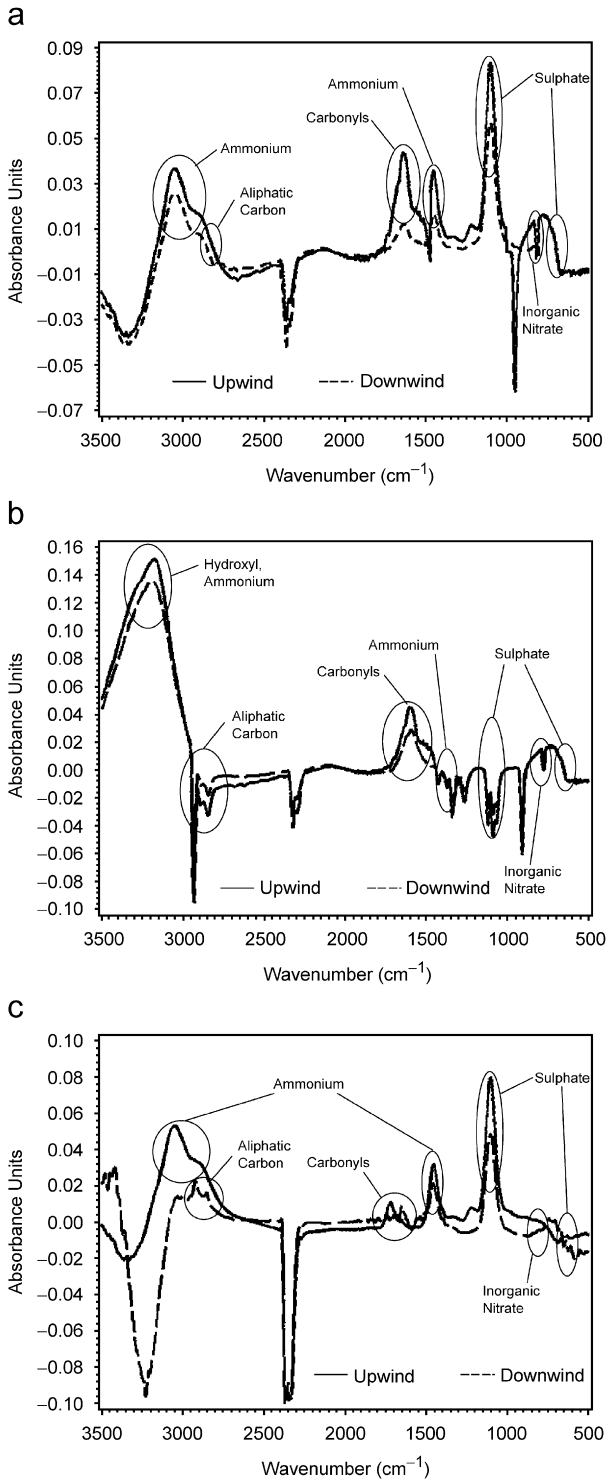

Fig. 2. FTIR spectra of PM_{10} samples collected upwind and downwind of the hawthorn hedge.

et al., 1994; Blando et al., 1998), which, when considered along with the other spectral background issues with the Experiment 2 spectrum mentioned above, suggests that the large peak between 3000 and 3500 cm^{-1} in Fig. 2b is at least in part a residual of background subtraction.

4. Conclusions

This work has presented methods and results of sampling and analyzing of PM_{10} and constituent functional groups for the purpose of characterizing the ability of hedges to filter PM_{10} from ambient air. The first known measurements of hedge $CE_{Filtration}$ values for PM_{10} have been presented, and effects of the hedge on organic and inorganic functional groups have also been explored. Current results suggest that hedges are potentially useful barriers for capturing ambient PM_{10}. The sample extraction and FTIR analysis method allowed us to explore the resulting spectra without the interference of the large absorbances that result from Teflon filters. The methods used to remove the influence of the extracting solvent and the field background impurities from the sample spectra appear to be introducing difficulties (sample peak distortion, creation of negative absorbances) in interpreting upwind–downwind differences and confounded attempts made to derive meaningful functional group concentrations from the spectra. To quantitatively characterize the effects of hedges on functional group composition, an FTIR analysis method more suitable for quantitation (Krost & McClenny, 1994; McClenny, Childers, Rohl, & Palmer, 1985; Reff et al., 2007) should be applied to a large number of paired upwind–downwind samples to robustly characterize how hedges affect concentrations of individual functional groups. The larger number of samples will also aid in accounting for instrumental anomalies in the spectroscopic analysis. Such an analysis between functional group composition and collection efficiency of the hedge is anticipated to help elucidate physico-chemical mechanisms by which hedges act as particle filters. This information will assist with considering and optimally employing hedges to mitigate concentrations in air quality management planning and implementation processes.

Disclaimer

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy.

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