Contribution of wood burning to PM$_{10}$ in London

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HIGHLIGHTS

- Aethalometer and levoglucosan methods used to estimate the contribution of wood smoke to PM$_{10}$ in London.
- Annual mean PM$_{10}$ from wood burning in London was 1.1 μg m^{-3}.
- PM was most likely from a mixture of wood types burnt as decorative or secondary heating.

GRAPHICAL ABSTRACT

ABSTRACT

Ahead of measures to incentivise wood heating, the current level of wood burning in London was assessed by two tracer methods; i) a six week campaign of daily measurements of levoglucosan along a 38 km transect across the city during winter 2010, ii) a three year (2009–2011) measurement programme of black carbon and particulate matter from wood burning using differential IR and UV absorption by Aethalometer. Mean winter levoglucosan concentrations were 160 ± 17 ng m^{-3} in central London and 30 ± 26 ng m^{-3} greater in the suburbs, with good temporal correlation ($r^2 = 0.68$–0.98) between sampling sites. Sensitivity testing found that the aethalometer wood burning tracer method was more sensitive to the assumed value of the Ångström coefficient for fossil fuel black carbon than it was to the Ångström coefficient for wood burning PM, and that the model was optimised with Ångström coefficient for fossil fuel black carbon of 0.96. The aethalometer and levoglucosan estimates of mean PM from wood burning were in good agreement during the winter campaign; 1.8 μg m^{-3} (levoglucosan) and 2.0 μg m^{-3} (aethalometer); i.e. between 7% and 9% of mean PM$_{10}$ across the London transect. Analysis of wood burning tracers with respect to wind speed suggested that wood burning PM was dominated by sources within the city. Concentrations of aethalometer and levoglucosan wood burning tracers were a greatest at weekends suggesting discretionary or secondary domestic wood burning rather than wood being used as a main heating source. Aethalometer wood burning tracers suggests that the annual mean concentration of PM$_{10}$ from wood burning was 1.1 μg m^{-3}. To put this in a policy context, this PM$_{10}$ from wood burning is considerably greater than the city-wide mean PM$_{10}$ reduction of 0.17 μg m^{-3} predicted from the first two phases of the London Low Emission Zone which was introduced to reduce PM from traffic sources.

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1. Introduction

Air pollution from domestic wood burning has long been recognised as an important contributor to poor ambient air quality in Scandinavian and alpine regions of Europe where wood burning is routinely used for residential space heating (Puxbaum et al., 2007; Yttri et al., 2005). However, recent evidence is suggesting that biomass burning might be more widespread. Fuller et al. (2013) highlighted PM_{10} from wood burning in three major European cities: London, Paris and Berlin, and Caseiro et al. (2009) estimated that wood burning was responsible for around 10% of wintertime PM_{10} in Vienna. In Flanders, Belgium wood burning has been estimated to contribute between 5 and 6% of annual mean PM_{10} in six cities and up to 13% in rural areas (Maenhaut et al., 2012). In southern Europe, wood combustion is estimated to comprise 60% of residential energy use in Portugal but accounts for almost 99% of domestic PM_{10} emissions (Borrego et al., 2010).

Across the European Union, the use of biomass (including wood) in heating looks set to rise by 57%—111% between 2010 and 2020, as the 27 member states are committed to obtain 20% of their energy requirements from renewable sources, including biomass, as part of a raft of proposals to reduce CO_2 emissions (Wagner et al., 2010). In response to these targets, the UK Department for Energy and Climate Change has announced the world's first renewable heat incentive, which will provide a financial incentive to switch from fossil fuel to renewables as part of a strategy to 'de-carbonise' the generation of heat in domestic, business and industrial premises in the UK (DECC, 2011). Additionally, the so-called Merton Rule requiring 10% onsite renewable energy in large new developments is now part of the UK planning policy (Merton, 2009). Further, there is a risk that increased fossil fuel prices may contribute to an increase in biomass burning; in Denmark Glasius et al. (2006) found that increasing fossil fuel costs contributed to doubling of wood stoves and boilers over a ten year period.

There is considerable uncertainty concerning the likely magnitude of the change in air pollution concentration arising from an increase in wood burning. The impact will depend not only on the uptake of wood burning but also on the type of boilers installed, how much they will be used in practice and the type of wood used. Any estimation of changes to PM_{10} concentration from installed capacity is likely to be further confounded by burning of wood in existing fire places; a source where knowledge is scarce.

The current study sought to determine the existing contribution of wood burning to PM_{10} in London so that the impacts of increased biomass burning can be quantified in the future. The measurement strategy employed 1) a six week campaign of daily filter based wintertime measurements of levoglucosan, an acknowledged wood burning tracer and 2) three years of highly time resolved (15 min mean) measurements of wood burning tracers using aethalometers. Measurements of PM_{10}, particulate sulphate and ethane were also made to inform source attribution.

2. Methods

2.1. Filter sampling and measurement of levoglucosan, mannosan and galactosan

Sampling for levoglucosan, mannosan and galactosan was carried out during the middle of the 2010 winter heating period. Sampling took place between 18th January and 28th February at five urban background monitoring sites situated along a 38 km transect across the city, as well as at a site (*Islington*) located within 20 m of a modern wood pellet boiler used for heating an education centre (for map details see Fig. 1). Daily samples were collected
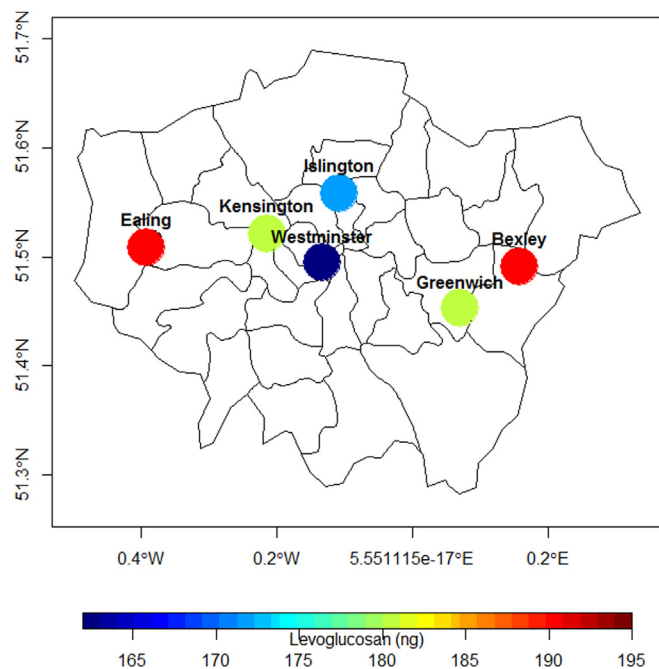

Fig. 1. Sampling locations for levoglucosan. Locations are colour coded according to mean concentrations (ng m^{-3}). Aethalometer measurements were undertaken at *Kensington* and at a site 2 km north of the *Greenwich* sampling location. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

onto quartz fibre filters (47 mm, QM-A, Whatman) using Partisol (Thermo, 2025) samplers. The sample flow was checked using a traceable flow meter (Bios model DC-HC-1 Rev E). Sampling uncertainties due to flow calibration have been assumed to be 1.7% ($k = 1$, $\sim 1\sigma$), as in EN14904.

Quantification of the levoglucosan, mannosan and galactosan was performed according to the method described in Dye and Yttri (2005). Briefly, a 2 cm^2 punch from each quartz fibre filter was soaked in tetrahydrofuran (2 ml) and subjected to ultrasonic agitation (30 min). The filter extract was then filtered through a syringe filter (0.45 μm) to remove PM and filter parts. Each filter was extracted twice. The extracted volumes were pooled and evaporated to a total volume of 1 ml in an N_2 atmosphere. Before analysis the sample solvent elution strength was adapted to the mobile phase by adding Milli-Q water (0.8 ml). The concentrations of levoglucosan, mannosan and galactosan were determined using High Performance Liquid Chromatography (HPLC) in combination with HRMS-TOF (High Resolution Mass Spectrometry Time-of-Flight) operated in the negative electrospray mode. The compound separation was performed with two series-connected reversed-phase C18 columns (Atlantis dC18, Waters). Levoglucosan, mannosan and galactosan were identified on the basis of retention time and mass spectra of authentic standards and quantification was performed using isotope labelled standards. The limit of quantification at a signal to noise ratio of ten was approximately 30 ng of injected levoglucosan. Uncertainty due to the laboratory determination of levoglucosan was estimated at 5% ($k = 1$, $\sim 1\sigma$) which was assumed to be non-random. If random uncertainty from the punch is considered, the uncertainty in individual samples was estimated at 11% ($k = 1$, $\sim 1\sigma$). Combined with the smaller sampling uncertainty an overall uncertainty of 11% ($k = 1$, $\sim 1\sigma$) was expected for daily measurements of levoglucosan concentrations. Following the methods in ISO 11222, for the 42 day mean concentrations an uncertainty of 5.2% ($k = 1$, $\sim 1\sigma$) is

expected. Identical uncertainty was assumed for mannosan and galactosan.

2.2. Estimates of PM_{10} from wood smoke using IR/UV absorption

When compared with black carbon from road transport sources, brown carbon from wood smoke has a greater absorption in the UV wavelength compared to IR. If the wavelength dependent absorption (the Ångström coefficient) of black and brown carbon is known, then the wavelength dependent absorption of unknown mixtures of carbon particles in ambient air can be disentangled to apportion black carbon (BC) between fossil fuel and wood burning and to provide a tracer for PM from wood burning (Wang et al., 2011; Kirchstetter et al., 2004; Sandradewi et al., 2008a). Magee AE-21 dual wavelength aethalometers (Hansen, 2005) measured the IR (880 nm) and UV (370 nm) absorption of sampled PM during 2009 and 2010 at two urban sites in London: in North Kensington (Kensington) where levoglucosan sampling also took place and in Greenwich, (2 km north of the levoglucosan sampling site). Aethalometer measurements were subject to spot darkening corrections using the Virkkula et al. (2007) method. This adjusts the measured absorption so that the measurement immediately before a tape advance is equal to that immediately after the tape advance.

Briefly, it is assumed that the total absorption (B_{abs}) at a wavelength λ is a combination of absorption due to fossil fuels; B_{abs} (ff) and wood burning; B_{abs} (wb) (1)

$$B_{abs}\,\lambda = B_{abs}(ff) + B_{abs}(wb) \qquad (1)$$

The wavelength dependent absorption of light (B_{abs}) by aerosols is proportional to $\lambda^{-\alpha}$ where α is the Ångström exponent such that:

$$B_{abs}(ff)\,(370)/B_{abs}(ff)\,(880) = (370/880)^{-\alpha ff} \qquad (2)$$

$$B_{abs}(wb)\,(370)/B_{abs}(wb)\,(880) = (370/880)^{-\alpha wb} \qquad (3)$$

Values of around 1 have been found for the Ångström exponent of fossil fuel black carbon from road transport (α_{ff}). Values of around 2 have been reported for the Ångström exponent of PM from wood smoke (α_{wb}) (Kirchstetter et al., 2004; Sandradewi et al., 2008a). By solving (1)–(3) unique values for B_{abs} (wb) (880) and B_{abs} (wb) (370) can be found for any aethalometer measurement of black carbon. B_{abs} (wb) (880) allows equivalent black carbon concentrations to be apportioned between the fossil fuel and wood burning sources i.e.

$$B_{abs}(880) = B_{abs}(ff)\,(880) + B_{abs}(wb)\,(880) \qquad (4)$$

Additionally, B_{abs} (wb) (370) has also been used as a tracer for total PM from wood burning (Sandradewi et al., 2008b; Favez et al., 2009; Sciare et al., 2011). B_{abs} (wb) (370), B_{abs}(ff) (880), B_{abs}(wb) (880) and PM from wood burning were calculated for each 15 min mean aethalometer measurement.

2.3. Other air pollution measurements

The Tapered Element Oscillating Microbalance - Filter Dynamics Measurement System (TEOM-FDMS, Thermo) was used to measure PM_{10}. PM_{10} instruments were independently calibrated and audited twice yearly by the National Physical Laboratory or Ricardo-AEAT plc. Sulphate was measured on daily PM_{10} filter samples by the UK Particle Numbers and Concentrations Network using ion chromatography (Beccaceci et al., 2011). Ethane was measured using gas chromatography (Perkin Elmer) by the UK Hydrocarbon Network (Yardley et al., 2011).

2.4. Meteorological measurements and analysis

Wind speed measurements (Gill, Windsonic) were made on a 5 m mast at the Bexley site at the east end of the levoglucosan transect. The mast was situated in an open grass area free from obstructions to a distance of approximately 20 m. Ambient temperature was also measured at the site at a height of around 2 m. Meteorological measurements at Bexley were assumed to represent conditions along the whole levoglucosan sampling transect which largely followed the line of the tidal Thames estuary.

3. Results and discussion

3.1. Levoglucosan

Levoglucosan was employed as the most recognized marker for tracing emissions of particulate matter from biomass burning for several reasons: it is emitted in high concentrations, it is not present in the vapour phase, it is associated with fine aerosols exclusively, and, until recently it was not thought to be selectively removed from the atmosphere (Simoneit et al., 1999). More recently, chamber studies by Hoffmann et al., (2010) and Hennigan et al. (2010) found evidence of levoglucosan being depleted following from reaction with the OH radical. However, this is not likely to be a major sink when tracing local sources during wintertime. Concentrations of levoglucosan (Fig. 2 and Table 1) showed substantial daily variation, between 43 ± 9 ng m^{-3} and 729 ± 160 ng m^{-3} ($k = 2$, $\sim 2\sigma$). Mean concentrations of levoglucosan ranged from 160 ± 17 ng m^{-3} in central London to 190 ± 20 ng m^{-3} ($k = 2$, $\sim 2\sigma$) in the east and west London suburbs as shown in Fig. 1; a difference of 30 ± 26 ng m^{-3} ($k = 2$, $\sim 2\sigma$) or $19 \pm 16\%$ of the inner London concentration. Correlations (r^2) between daily levoglucosan measurements at pairs of sites were lowest between inner London sites and those in the suburbs (0.68) and highest between the two inner London sites (0.98). A maximum (r^2) of 0.89 was found between suburban sites. Correlations (r) between the levoglucosan concentrations and temperature were low, ranging between -0.15 and -0.22 suggesting that where biomass is being used, it is not being used as a main or sole heating source. Levoglucosan measured at the Islington site was within the range of the other sites in the study despite its proximity to a wood chip burner, however wind only blew from the direction
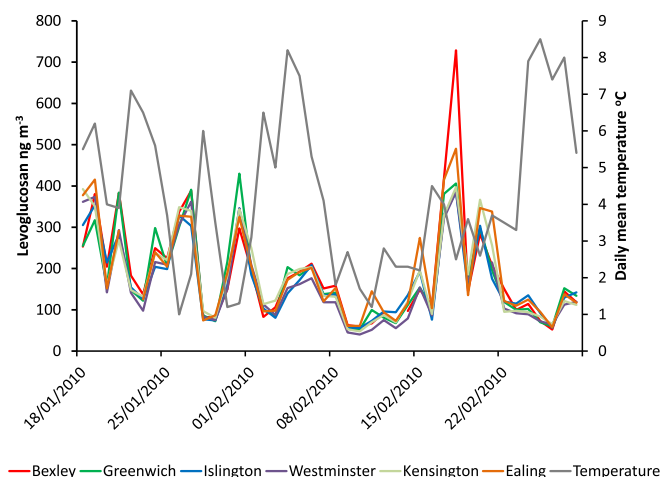

Fig. 2. Daily mean concentrations of levoglucosan and mean temperature.

Table 1
Minimum, mean, and maximum levoglucosan along with mean concentrations of mannosan and glactosan at each sampling site. Sites are listed from west to east along the transect.

Site	ng m^{-3}				
	Levoglucosan			Mannosan	Galactosan
	Min	Mean	Max	Mean	Mean
Ealing	57 ± 6	190 ± 21	490 ± 55	24 ± 6	11 ± 3
Kensington	48 ± 5	180 ± 20	397 ± 44	24 ± 5	11 ± 2
Westminster	40 ± 4	162 ± 18	385 ± 43	22 ± 5	10 ± 2
Islington	56 ± 6	171 ± 19	385 ± 43	24 ± 5	10 ± 2
Greenwich	53 ± 5	180 ± 20	430 ± 48	23 ± 5	10 ± 2
Bexley	52 ± 5	190 ± 21	729 ± 82	24 ± 5	10 ± 2

of the wood chip burner ($\pm 20°$) for 6% of the sampling period. Additionally it is possible that the sampling site was too close to the wood chip burner for the buoyant burner plume to be detected.

With considerable variation expected in atmospheric dispersion it is difficult to draw conclusions about levoglucosan sources from a short measurement campaign. Derwent et al. (1995) suggested the use of ambient ethane, which arises from leakages in the natural gas distribution network, as a tracer for dispersion of diffuse urban sources. Measurements of ethane were made at the kerbside in central London as part of a measurement programme that began in 1998. To disentangle the effects of dispersion from variations in emissions, simple regression models (using standard major axis regression) were constructed using ethane as a predictor of the daily mean levoglucosan concentration. A separate model was constructed for each levoglucosan sampling location using data from all of the other sites. The variation in the mean relative residuals [(measured − prediction)/prediction] for each weekday are shown in Fig. 3. It is clear that the levoglucosan concentration is greater than predicted at weekends; by up to 30% on Saturdays and up to 54% on Sundays, suggesting greater emissions when compared with weekdays.

In the absence of data on wood consumption, ratios of anhydrous sugars can point to the type of wood being burnt (Schmidl et al., 2008). Mean levoglucosan to mannosan ratios were 7.1–7.8 across the six London sites suggesting a mixture of hard and soft wood burning.

3.2. Sensitivity testing of the aethalometer wood smoke model

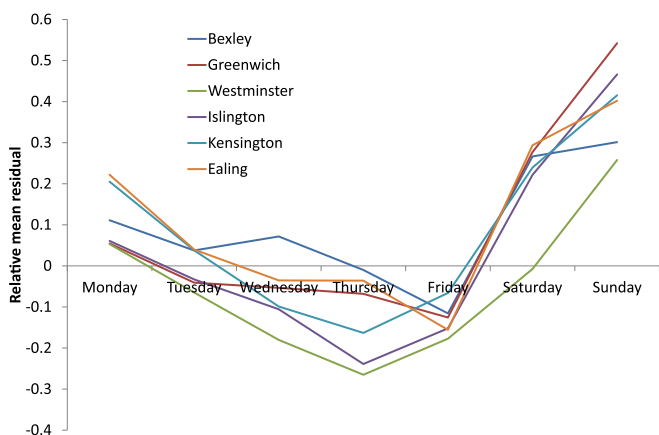The aethalometer method for tracing of wood burning PM depends on differences between the values of the wavelength dependent absorption (the Ångström coefficient) α for fossil fuel black carbon and for brown carbon from wood burning. It is however unclear how the assumptions about the values of α for fossil-fuel black carbon and wood burning brown carbon affect the estimates of wood burning PM using this approach. A series of sensitivity tests were therefore undertaken using the measurements from the *Kensington* aethalometer. Additionally, comparison of the aethalometer and levogluocosan methods at *Kensington*, provided further opportunities to conduct sensitivity testing of the aethalometer model. Clearly measurements from both methods should be correlated but the magnitude of the levoglucosan and B_{abs} (wb) (370) cannot be usefully used to test variations in α_{ff} and α_{wb} since both require the applications of further factors before an estimate of PM mass concentration can be obtained. However, it is expected that zero (or null) concentrations of both tracers should occur at the same time. Although zero measurements were not present in the dataset, the regression intercept of levoglucosan on B_{abs} (wb) (370) should be zero if the methods are consistent and both tracers experience the same rate of atmospheric removal.

By varying α_{ff} and α_{wb} by 10% from the commonly used values of $\alpha_{ff} = 1.0$ and $\alpha_{wb} = 2.0$ it was found that mean values of the wood burning tracer B_{abs} (wb) (370) was more sensitive to changes in α_{ff} (+20% at $\alpha_{ff} = 0.9$ and −26% at $\alpha_{ff} = 1.1$) than it was to changes in α_{wb} (−10% at $\alpha_{wb} = 2.2$ and +16% at $\alpha_{wb} = 1.8$). Increasing α_{wb} by 50% to 3.0 led to a decrease in B_{abs} (wb) (370) by 30%.

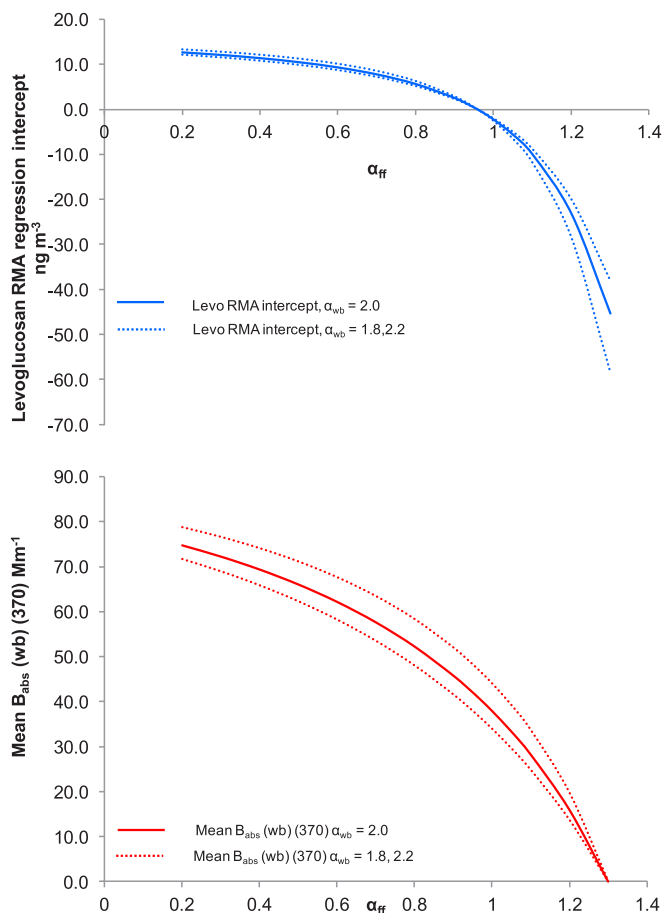**Fig. 3.** Mean relative residual levoglucosan from ethane regression model, averaged by day of week.

Fig. 4. Upper panel shows the standard major axis regression intercept of levoglucosan tracer B_{abs} (wb) (370) as a function of α_{ff}. Lower panel shows the aethalometer wood burning tracer B_{abs} (wb) (370) as a function of α_{ff}. In both cases measurements were made at *Kensington*. Bold lines show results with $\alpha_{wb} = 2$ and dotted lines indicate results when α_{wb} was varied between 1.8 and 2.2.

Using $\alpha_{ff} = 1.0$ and $\alpha_{wb} = 2.0$, correlations (r^2) between levoglucosan and B_{abs} (wb) (370) were good (0.79) and, as expected from solution to Equations (1)–(3), this did not vary when α_{wb} was varied between 1.8 and 2.2. Fig. 4 (upper) shows how the reduced major axis (RMA) regression intercept varied as α_{ff} was varied between 0.2 and 1.3. It was found that a levoglucosan (RMA) regression intercept of zero was only possible at an α_{ff} value of 0.96, consistent with the literature estimates of α_{ff}. At this value the intercept became insensitive to variations in α_{wb}. Looking at this test another way, this use of aethalometer and levoglucosan measurements to derive an α_{ff}; a physical property of ambient black carbon that was consistent with literature estimates, contributes to the validity of the aethalometer method to trace wood burning PM. Fig. 4 (lower) shows the effect of a varying α_{ff} on the magnitude of B_{abs} (wb) (370), with α_{wb} additionally varied between 1.8 and 2.2 as suggested by Kirchstetter et al. (2004). It can be seen that the sensitivity of B_{abs} (wb) (370) to changes in α_{wb} were fairly constant for values $\alpha_{ff} < 1$ but it became insensitive to changes in α_{wb} as the aethalometer model broke down and produced large negative intercepts of levoglucosan on B_{abs} (wb) (370) and as B_{abs} (wb) (370) approached zero with values of α_{ff} above 1.2.

3.3. Wood smoke tracers and wind speed

Relationships between wind speed and pollutants have been explored extensively by Jones et al. (2010) who found differing wind speed dependences for local and distant sources. To investigate the origins of wood smoke PM in London, daily mean levoglucosan and mean B_{abs} (wb) (370) across the study sites were compared to daily mean wind speed according to 0.25 m s^{-1} wind speed intervals. The top 5% of wind speed measurements were excluded from the analysis due to the relative scarcity of observations, which could induce large uncertainty in the averaging process. To aid interpretation, daily mean concentrations of sulphate at *Kensington* were included in the analysis as a tracer for long range PM transport, whereas daily mean ethane concentrations and absorption due to fossil fuel black carbon, B_{abs} (ff) (880), were used as tracers of diffuse urban sources.

As shown in Fig. 5, the mean concentration of sulphate was greatest for wind speeds between 1 and 2 m s^{-1}. A similar relationship was found by Smith et al. (2001) who concluded that the long range transport of PM$_{10}$ into London is not maximal when wind speeds are lowest, but instead wind speed is required for advection
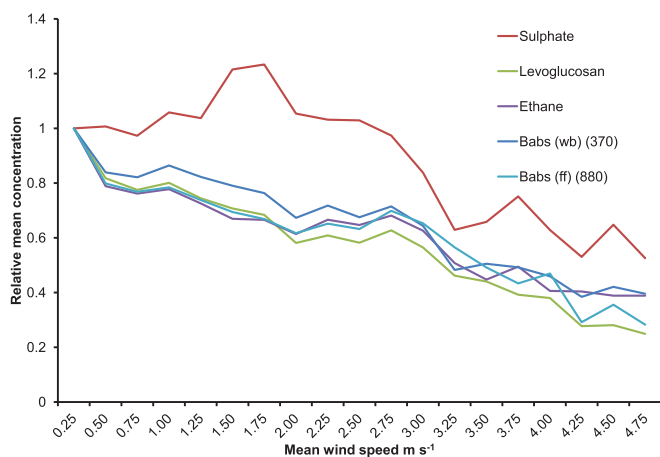
to take place. By contrast, the mean concentration of levoglucosan and B_{abs} (wb) (370) showed greatest concentrations at lowest wind speeds and decreased with increasing daily mean wind speed, consistent with that of ethane and B_{abs} (ff) (880), suggesting that wood smoke PM in London was mainly from diffuse urban sources and not advected into the city from more distant source regions.

3.4. Quantifying PM from wood burning

Both levoglucosan and aethalometer methods provide measurements of wood burning tracers, which require multiplication by factors to provide estimates of PM from wood burning.

For levoglucosan, a range of factors are reported in the literature. Further, emissions ratios of PM are known to vary according to the wood burnt. Having reviewed a wide range of laboratory tests, Puxbaum et al. (2007) suggested a factor of 7.35 to convert levoglucosan to organic carbon with an additional factor of 1.4 to estimate the organic mass for fresh wood burning. This is similar to the factor of 10 (range 5.5–14) estimated by Szidat et al. (2009). However, it is clear from the studies reviewed in Puxbaum et al. (2007) that wood burning also gives rise to elemental carbon (EC) emissions. These studies show a mean elemental carbon to levoglucosan emissions ratio of 0.9 (range 0.1–2.4, from five studies). An additional EC to levoglucosan factor of 1 was therefore included to provide a levoglucosan to PM emissions ratio of 11.

Other studies have derived factors to estimate PM from wood burning for the aethalometer model using linear regression with ambient elemental and organic carbon concentrations. A factor of 675,000 µg Mm^{-2} was chosen as a mean based on the studies by Favez et al. (2009, 2010), Sandrewi et al. (2008a,b) and Sciare et al. (2011) (range 541,000–810,000 µg Mm^{-2}, at 470 nm). This is similar to the value of 632,420 µg Mm^{-2} derived for the UK by Harrison et al. (2012). This factor was converted to the 370 nm channel response using equation (3). Fig. 6 shows good agreement between the time series of estimated PM at *Kensington* using both methods, although, the aethalometer model notably underestimates the levoglucosan method on 22nd and 29th January and 18th February. RMA regression between the two variables at *Kensington* gives:

$$[\text{PM wood Aethalometer}] = (0.95 \pm 0.0)\,[\text{PM wood levo}]$$
$$+ (0.06 \pm 0.14),\ r = 0.92\ n = 42.$$

Fig. 5. Relative, mean concentration of levoglucosan, sulphate and ethane along with the black carbon wood burning tracer B_{abs} (wb) (370) and the black carbon fossil fuel tracer B_{abs} (ff) (880) averaged by wind speed.
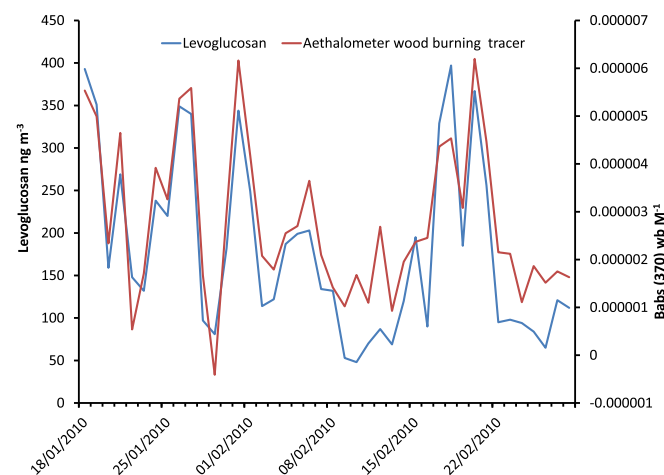

Fig. 6. Daily mean wood burning PM at *Kensington* using levoglucosan and aethalometer methods.
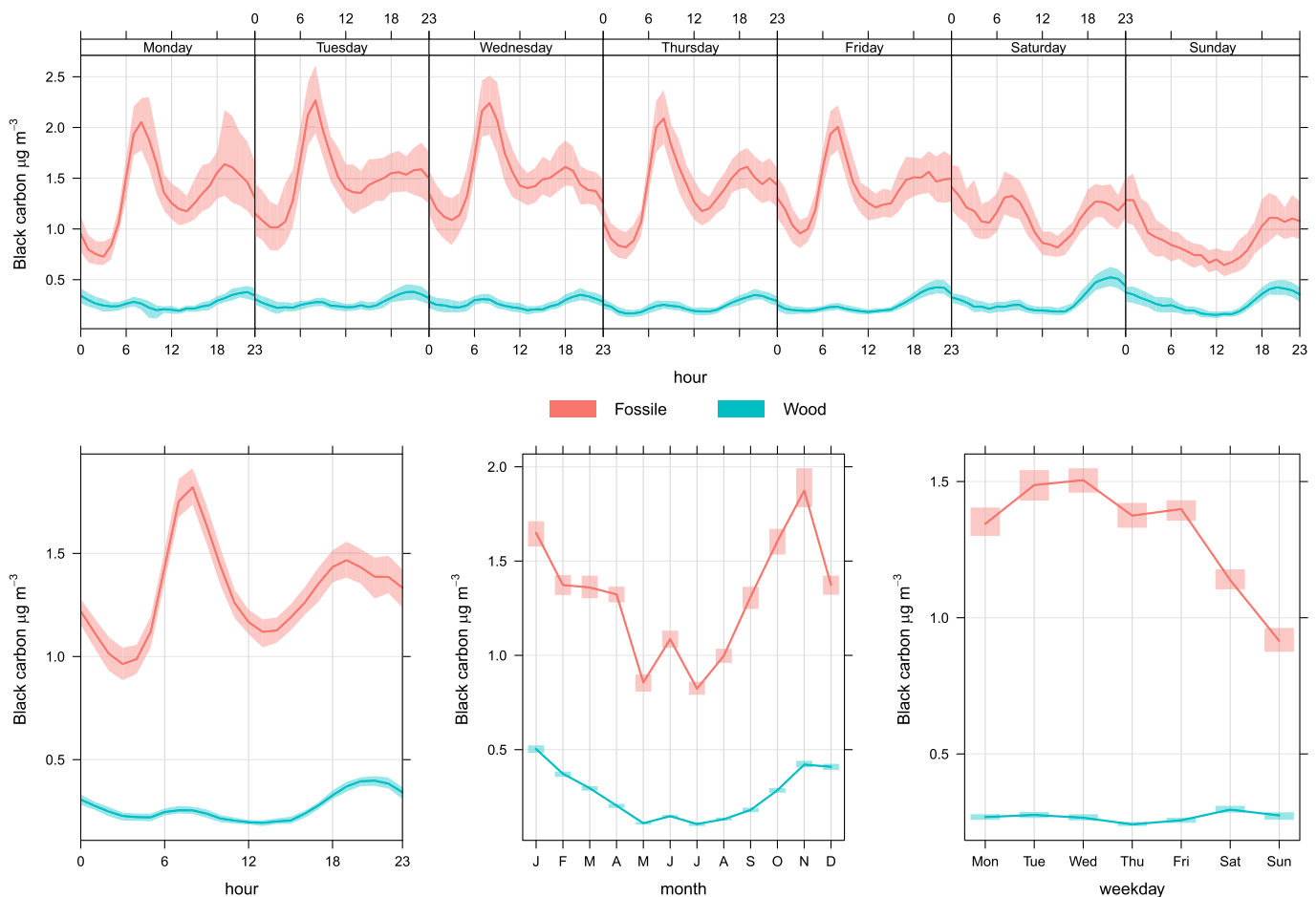
Fig. 7. Equivalent black carbon concentrations apportioned between fossil fuel and wood burning at *Kensington*. Concentrations were averaged by hour, day of week, and month using Openair (Carslaw and Ropkins, 2012).

A slightly worse agreement was obtained between the *Greenwich* aethalometer and levoglucosan estimates of PM from wood burning, which may be due to their 2 km separation distance:

$$[\text{PM wood Aethalometer}] = (0.78 \pm 0.05) \,[\text{PM wood levo}] \\ + (0.06 \pm 0.13), \ r = 0.89 \ n = 42.$$

During the wintertime levoglucosan sampling campaign, the mean estimated concentration of PM from wood burning at *Kensington* was 1.8 μg m^{-3} from the levoglucosan method and 2.0 μg m^{-3} from the aethalometer model; corresponding to approximately 8% of the total PM$_{10}$ concentration (22.7 μg m^{-3}) and approximately 12% of PM$_{2.5}$ (16.4 μg m^{-3}). Based on the levoglucosan sampling, PM from wood burning comprised between 8% and 10% of the mean PM$_{10}$ across the London transect during the wintertime sampling period.

3.5. Aethalometer estimates of equivalent black carbon and PM from wood burning 2009 to 2011

Fig. 7 shows equivalent black carbon concentrations from fossil fuel and wood burning, calculated using Equation (4) and assuming same mass absorption coefficient applies to equivalent black carbon from both origins. The default aethalometer value of 16.6 m^{-2} g^{-1} was applied following analysis by Butterfield et al. (2011) which showed good agreement (linear regression gradients between 0.86 and 1.25, r^2 range 0.55–0.94) between aethalometer black carbon using this default value and elemental

carbon measured using a NIOSH protocol at UK three locations, including *Kensington* (Butterfield et al., 2011). Each panel shows mean concentrations averaged over different time periods. The upper panel shows a clear weekday traffic profile in the equivalent black carbon from traffic, with a pronounced morning peak which is not present at weekends. By contrast equivalent black carbon from wood burning shows elevated concentrations during the evenings with greatest concentrations at weekends. The monthly contributions of wood burning to equivalent black carbon varied seasonally; from 23% during January and December to 11% during May and July.

Looking at the aethalometer estimates of total PM from wood burning during the three years 2009–2011, it was found that the mean PM from wood burning at Greenwich was 1.1 μg m^{-3}; 5% of the mean PM$_{10}$. At Kensington, the mean PM from wood burning was 1.2 μg m^{-3}; 6% of the mean PM$_{10}$. The monthly contributions of wood burning to PM$_{10}$ varied seasonally; from 10% during January to 2% during August.

4. Conclusions

The combination of levoglucosan and aethalometer measurements as wood burning tracers provided new insights into sources of ambient airborne particles in London. Good agreement was found between the two methods in terms of their temporal correlation and the magnitude of the estimated PM from wood burning. By using levoglucosan measurements to constrain sensitivity testing it was shown that the aethalometer

model was optimised at an Ångström exponent (α_{ff}) of 0.96 for black carbon from fossil fuel; very close to the value of 1.0 expected from other experimental literature, further confirming the validity of the aethalometer approach. At a value of $\alpha_{ff} = 0.96$, a $\pm 10\%$ change in α_{wb} varied the estimates of wood burning PM by -10% and $+16\%$.

Although the 2010 wintertime concentration of levoglucosan in London (184 ng m^{-3}) was at the low end of wintertime measurements in other European urban areas (60–900 ng m^{-3} across 15 studies reported in Szidat et al., 2009), both levoglucosan and aethalometer measurements suggest that PM from wood burning already makes an important contribution to wintertime PM$_{10}$ concentrations in London. New government financial incentives and requirements for new building are highly likely to lead to an increase in PM$_{10}$ from this source. Analysis of the relationship between wood smoke tracers and wind speed indicated that wood burning PM was not related to long range transport but instead arose from sources within London. Both methods suggested that wood smoke PM$_{10}$ was greatest during evenings and also at weekends, which also reflected local domestic burning. Poor correlations between wood burning tracers and daily mean temperature suggest that Londoners are burning wood as a decorative or secondary heating source rather than as a primary form of heating. This secondary heating is unlikely to merit investment in specifically designed wood stoves but likely reflects the UK practice of burning wood in existing fire places and grates originally designed for coal burning (a practice noted in Lee et al., 2005). Smoke Control Areas, introduced to almost all of Greater London under the 1956 Clean Air Act, should prohibit the burning of wood unless it is carried out in a specifically designed and authorised boiler or stove (Defra, 2010). The evidence of widespread wood burning in London may suggest that smoke control legislation is no longer effective. Increases in PM$_{10}$ from wood burning risks undermining other polices aimed at achieving compliance with EU Limit Values. For example the first two phases of London's Low Emission Zone, the largest such scheme in the world, were estimated to achieve a London-wide mean PM$_{10}$ reduction of 0.17 μg m^{-3} (TfL, 2008); considerably less than the annual mean concentration of 1.1 μg m^{-3} estimated to arise from wood burning.

The aethalometer measurements also suggested some summertime sources of wood smoke, as have been previously observed (Puxbaum et al., 2007; Saarikoski et al., 2008). The presence of a summer-time wood burning raises the possibility that garden waste burning, patio wood burners and other summertime sources also make measurable contributions to London's PM$_{10}$. Another possibility is a small contribution from long-range transport that was not apparent during our winter campaign.

Levoglucosan measurements suggested slightly greater wood burning in suburban London when compared to the central area, similar to the spatial gradients found in Berlin by Wagener et al. (2012) but importantly wood burning was not confined to the outer suburbs.

Toxicological evidence also gives rise to concerns about health effects from increased wood burning (Bølling et al., 2009). Additionally, domestic wood burning takes place where people live and at times when they are at home. Coupled with poor winter nighttime dispersion, Reis et al. (2009) suggested that even modest wood burning in densely populated residential areas may lead to PM exposures comparable to those from traffic sources.

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