**Atmospheric
Chemistry
and Physics
Discussions**

Comparison of methods for evaluation of wood smoke and estimation of UK ambient concentrations

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Evaluation of wood smoke and estimation of concentrations

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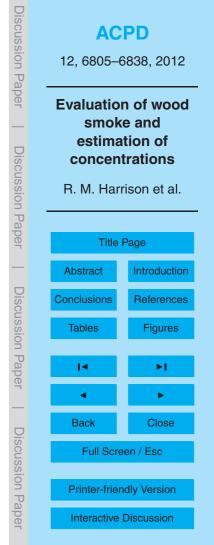Printer-friendly Version

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Abstract

Airborne concentrations of the wood smoke tracers, levoglucosan and fine potassium have been measured at urban and rural sites in the United Kingdom alongside measurements with a multi-wavelength aethalometer. The UK sites, and especially those in cities, show low ratios of levoglucosan to potassium in comparison to the majority of published data. It is concluded that there may be two distinct source types, one from wood stoves and fireplaces with a high organic carbon content, best represented by levoglucosan, the other from larger, modern appliances with a very high burn-out efficiency, best represented by potassium. Based upon levoglucosan concentrations and a conversion factor of 11.2 from levoglucosan to wood smoke mass, average concentrations of wood smoke including winter and summer sampling periods are $0.23\,\mu g\,m^{-3}$ in Birmingham and $0.33\,\mu g\,m^{-3}$ in London, well below concentrations typical of other northern European urban areas. There may be a further contribution from sources of potassium-rich emissions amounting to an estimated $0.08\,\mu g\,m^{-3}$ in Birmingham and $0.30\,\mu g\,m^{-3}$ in London. Concentrations were highly correlated between two London sites separated by 4 km suggesting that an advected regional source is responsible. Data from the aethalometer are either supportive of these conclusions or suggest higher concentrations, depending upon the way in which the data are analysed.

1 Introduction

The enforcement of legislation internationally is acting to reduce airborne concentrations of particulate matter, measured as $PM_{2.5}$ and PM_{10}. In the European Union, member states have to meet an air quality objective of $25\,\mu g\,m^{-3}$ for $PM_{2.5}$ as an annual mean by 2015, as well as an exposure reduction target to be met by 2020 whose magnitude depends upon the annual average concentration across qualifying sites in the years 2009–2011 (Harrison et al., 2012). While the contribution of components such as sulphate and nitrate to concentrations of $PM_{2.5}$ is readily determined, it is

more difficult to quantify the contributions of primary sources such as road traffic to PM concentrations, and estimating the secondary organic aerosol contribution is especially problematic (Pio et al., 2011).
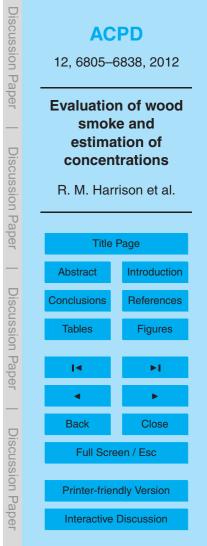
Amongst the sources most difficult to quantify by receptor modelling, and for which emissions inventory data are least reliable, wood smoke stands out as posing particular problems. Collecting activity data for emissions inventories is challenging and emission factors are highly variable depending upon the type of combustion appliance, or open combustion practice adopted (Kelz et al., 2010; Pio et al., 2008).

Since the estimation of emissions poses such large difficulties, most estimates of the source contribution are based upon receptor modelling methods. These may be based upon a CMB modelling approach using a range of compositional components (e.g. Chow et al, 2007), or more often upon individual source tracers. The majority of studies have measured anhydrosugars, of which levoglucosan is the predominant component. This method appears to be relatively specific to biomass combustion, but the ratio of levoglucosan to wood smoke mass is highly variable, depending upon the specific wood type being burnt and the combustion conditions (Puxbaum et al., 2007). Nonetheless, levoglucosan has been widely used to estimate contributions of wood smoke to organic carbon and to wood smoke mass (e.g. Puxbaum et al., 2007; Caseiro et al., 2009).

Fine particle soluble potassium has also been recommended as a wood smoke tracer (Chow et al., 2007). If it is to be used as a specific tracer, as opposed to part of a source profile in a CMB model, then it must be corrected for the contributions of sea salt and soil to airborne potassium (K). While the former can be achieved rather reliably in most situations based upon measurements of sodium, correction for the soil contribution is more difficult due to the inhomogeneity of soils and hence the difficulty of taking soil samples which give a relevant ratio of K/Ca to permit use of Ca as a soil tracer. Soil in the fine particle fraction may have travelled for significant distances and may hence differ substantially in composition dependent upon the source area. A third method for estimating wood smoke concentrations is based upon use of a dual,

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or multi-wavelength aethalometer (Sandradewi et al., 2008a,b). This method depends upon the assumption that elemental carbon arises from only two sources, road traffic and wood smoke, which may be differentiated from their Angstrom coefficients (i.e. the wavelength dependence of the optical absorption coefficient). This assumption may be correct in Swiss valleys under the atmospheric inversion conditions prevailing when the method was developed (Sandradewi et al., 2008a,b), but the applicability of the method under other circumstances has not been systematically explored.

There have been few studies of the source apportionment of particulate matter in the UK atmosphere. Yin et al. (2010) applied a CMB model to chemical data including a range of organic source tracers including levoglucosan. Their estimates of wood smoke contributions to annual mean $PM_{2.5}$ mass were only 0.07 and 0.06 $\mu g\,m^{-3}$ respectively for urban and rural sites, with rather little difference between seasons. These estimates, measured in the UK West Midlands area, are in sharp contrast to data reported by Fuller et al. (2011) based upon levoglucosan concentrations measured in the London area showing estimated wood smoke concentrations of approximately 3 $\mu g\,m^{-3}$ during winter. Even, allowing for a divergence of a factor of approximately two revealed by an inter-laboratory comparison, there remains a very large discrepancy which seems unlikely to be explained by the different geographic areas in which measurements were made.

In this study, we report measurements of the wood smoke tracers levoglucosan and fine K at a number of UK locations. These are used to estimate concentrations of wood smoke, which are compared with estimates derived from simultaneous deployment of a multi-wavelength aethalometer.

2 Experimental

Four air sampling sites were used as follows:

Elms Road Observatory Site (EROS), Birmingham

EROS (52.45° N; 1.93° W) is a urban background site located in an open field within the Birmingham University campus. The site is about 3.5 km southwest of the centre of Birmingham which has a population of over one million and is part of a conurbation of 2.5 million population. The nearest anthropogenic sources are a nearby railway, and some moderately trafficked roads. There is little residential accommodation within 300 m. Data from this site have previously been used in a Chemical Mass Balance receptor model of $PM_{2.5}$ (Yin et al., 2010).
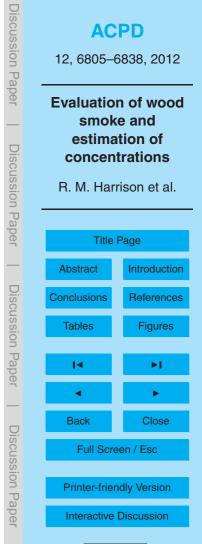
North Kensington, London

This is an urban background site in West London (51.52° N; 0.21° W). It is in the grounds of a school in a residential area, 7 km to the west of central London and hosts a station of the National Automatic Urban and Rural Network. The site is widely accepted as representative of air quality across a large part of London and the air pollution climate at this site has been analysed in detail by Bigi and Harrison (2010).

Marylebone Road, London

This is on a heavily trafficked (ca. 80 000 vpd) six lane highway running through a street canyon in central London (51.52° N; 0.16° W). It shows a large increment in particle mass above neighbouring central urban background sites (Charron et al., 2007) and the particle sources influencing the site have been identified tentatively by Harrison et al. (2011).

Budbrooke, Warwick Sampling Site

This is a rural location (52.17° N; 1.38° W), 55 km to the southeast of Birmingham and 4 km to the west of Warwick). The sampler was located in a clearing on the edge of an area of hardwood forest and was subject to local wood smoke emissions both from domestic wood stoves and from the open burning of waste wood about 50 m north of the sampling location.
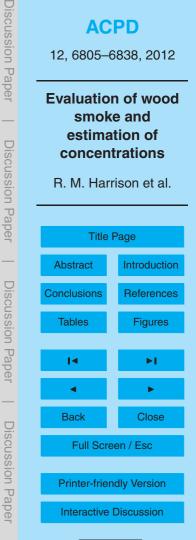
2.1 Sampling procedures

Samples for levoglucosan analysis were collected with a Graseby-Andersen high volume sampler operated at $0.9\,\text{m}^{-3}\,\text{min}^{-1}$ for 24 hours with a 20.3×25.4 cm QM-A Whatman quartz fibre filter substrate after selection of the $PM_{2.5}$ size fraction with a pre-impactor. At the London North Kensington sampling site, samples were collected with the Digitel DH-80 automated sampler with a $PM_{2.5}$ inlet operating at $0.5\,\text{m}^3\,\text{min}^{-1}$ over 24 h onto a Whatman QM-A 150 mm diameter quartz fibre filter. Samples for analysis of potassium and calcium were collected using a Rupprecht and Patashnik dichotomous Partisol Plus model 2025 sequential air sampler which collects separate coarse and fine particle fractions, and only data for the fine fraction sampled at $15\,\text{l}\,\text{min}^{-1}$ onto 47 mm polypropylene-backed PTFE filters were used in the study.

For some of the sampling periods, a Magee Scientific multi-wavelength aethalometer providing absorption data at seven wavelengths ($\lambda = 370, 470, 520, 590, 660, 880$ and 950 nm) was deployed for the measurement of black and brown carbon. The instrument was fitted with a $PM_{2.5}$ inlet and operates at $4\,\text{l}\,\text{min}^{-1}$. Data were corrected for loading effects according to the commonly used algorithm of Weingartner et al. (2003).

2.2 Chemical analysis

Potassium

PTFE filters were extracted into water and analysed by ion chromatography for potassium, sodium and calcium according to the methods described by Yin and Harrison (2008).
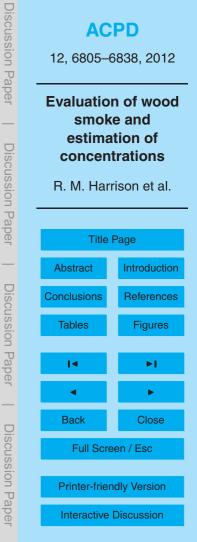
   The concentrations of potassium needed to be corrected for the minor contributions of sea salt and soil to the fine fraction. The corrections were based on the method of Pio et al. (2008) which use sodium as a tracer for sea salt and calcium for a tracer of soil and assumes a mass ratio of 10 for K/Ca in wood smoke. In order to generate the corrections, the standard ratio of K/Na of 0.036 in sea salt was adopted and the soil calcium concentration was estimated by the sampling of local soils followed by sieving to collect a < 20 µm size fraction which was extracted with water and analysed according to the same methods as the air filters. Therefore, site specific corrections were adopted. The equation used to estimate the potassium arising from wood smoke is as follows:

$$K_{ws} = (K_{measured} - 0.036 \times Na_{measured} - [K/Ca]_{soil} \times Ca_{nss})/(1 - 0.1 \times [K/Ca]_{soil})$$

where the subscript nss = non-sea salt.

Levoglucosan

Two procedures were used for the analysis of levoglucosan. Samples from the EROS and Budbrooke sampling sites were analysed according to the method of Zdrahal et al. (2002). Before extraction, an internal standard (methyl-beta-D-xylopyranoside, $C_6H_{12}O_5$) was spiked evenly across the aerosol filter segment (1/4) to account for losses during sample extraction and concentration.

The spiked filters were extracted three times, each time for 20 min with 30 ml of dichloromethane under ultrasonic agitation. The first extraction was performed under acidic conditions by addition of acetic acid (200 µl). The combined dichloromethane extracts were reduced with a rotary evaporator (400 hPa, 25 °C) to approximately 5–10 ml. Then the concentrated extracts were filtered through a PTFE filter and completely evaporated under a stream of nitrogen. Finally, the evaporated samples were re-dissolved in pyridine (200 µl) then kept in the 4 °C refrigerator until derivatization.
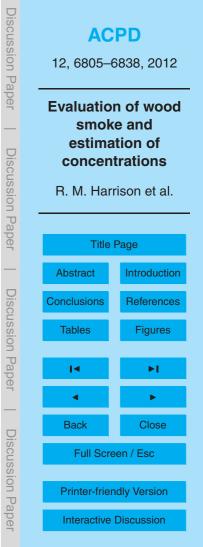
Aliquots of 50 µl of the sample solutions were taken out and derivatied by trimethylsilylation mixture (40 µl). After derivatization, a 10 µl recovery standard (1 ppm) was added in to the sample and total of 100 µl final sample was analysed by GC/MS. In this study, 1-phenyl dodecane ($C_{18}H_{30}$) was applied as recovery standard. The addition of recovery standard was to compensate for injection volume effects and variations in the GC/MS detector response.

The derivatization process was carried out on batches of 10 samples using the trimethylsilylation mixture which comprised 99 % of N-methyl-N-trimethylsilyltrifluoroacetamide (MSTFA) and 1 % trimethylchlorosilane (TMCS). The trimethylsilylation mixture was freshly made before the derivatization process which was performed in a sealed vial for 60 min at 80 °C in a dry heater block. After derivatization, the sealed vials were opened to add the recovery standard, and then analysed immediately by GC/MS.

Quantification was carried out by GC/MS and was based on an internal standard calibration procedure with appropriate recovery and blank corrections. A sample volume of 1.0 µl was injected into a split/splitless injector, operated in the splitless mode at a temperature of 250 °C. The carrier gas was helium at a pressure of 120 kPa. The temperature program was started at 45 °C for 4 min , a gradient of 20 °C min^{-1} was used up to 100 °C, followed by 10 min at this temperature, then the temperature increased to 315 °C at 5 °C min^{-1} and was held for 20 min. The *m/z* ratios for the ions used in the GC/MS analysis are 204, 217 and 333.

Samples from London North Kensington and Marylebone Road were analysed according to the method of Yue and Fraser (2004) as described by Yin et al. (2010). This method uses derivatization with N, O-Bis (trimethylsilyl), trifluroacetamide plus 10 % trimethylchlorosilane (BSTFA-TCMS) with subsequent GCMS analysis on an Agilent Technologies GCMS (GC-6890N plus MSD-5973N) fitted with a HP-5MS column (30 m, 0.25 mm diameter, 0.25 μm thickness). A levoglucosan-$U^{13}C6$ isotopically used internal standard was utilised.

The two procedures for levoglucosan analysis were intercompared both using a set of environmental samples and by analysis of levoglucosan in NIST SRM 1649B which contains $81.1 \pm 6.1 \, \mu g \, g^{-1}$ levoglucosan.
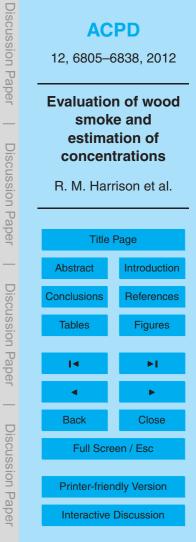
Aethalometer

Using the methodology of Sandradewi et al. (2008a,b) the contributions from traffic and wood burning to the total measured carbonaceous material, CM (organic matter + black carbon) can be apportioned using the different absorption coefficients measured for both sources at 370 nm and 950 nm by the aethalometer. By assuming that the Ångström exponent for both traffic and wood burning has a fixed value (1.0 and 2.0 were used, respectively), simultaneous equations can be derived – and solved – to calculate an absorption coefficient for both wood smoke and traffic. Linear regression then correlates these values to a $PM_{traffic}$ and PM_{wb} value which sum to give the total CM values.

The absorption coefficients b_{abs} (370) and b_{abs} (950) were derived from attenuation values ATN of the two different wavelengths ($\lambda = 370$ nm and 950 nm) measured across a quartz tape filter.

$$ATN = 100 \ln \left(\frac{I_o}{I} \right)$$

in which I_o and I are both beam intensity before and after attenuation by the particle-laden quartz tape filter.

Given the measured values of ATN for each of the wavelengths, the flow rate Q, the sampling time t and the filter spot size $A = 1.67\,\text{cm}^2$, values of aerosol attenuation coefficients b_{abs} were calculated using

$$b_{ATN} \equiv \frac{A}{Q} \frac{\Delta ATN}{\Delta t} \tag{1}$$

For both wavelength (370 and 950 nm) channels, the b_{ATN} values were "despiked" and smoothed using an in-house algorithm, before a correction due to Weingartner (2003) was applied to account for the "shadowing" effect of the particles as the filter becomes loaded.

The absorption coefficient b_{abs} was wavelength (λ) dependent

$$b_{abs} \propto \lambda^{-a} \tag{2}$$

and from this relation, the Ångström exponent α, can be derived. When comparing the ratio of the absorption by the wood smoke (which has a higher content of organic material) to the absorption by traffic particles (which have a higher of content of black carbon), both traffic and wood smoke have a stronger absorption at the shorter wavelengths, but wood smoke absorbs significantly more radiation than traffic at 370 nm compared to 950 nm. This is reflected by the typically higher absorption exponents for wood smoke compared to traffic. It follows that

$$\frac{b_{abs}(370\,\text{nm})_{traffic}}{b_{abs}(950\,\text{nm})_{traffic}} = \left(\frac{370}{950}\right)^{-\alpha_{traffic}}$$

$$\frac{b_{abs}(370\,\text{nm})_{wb}}{b_{abs}(950\,\text{nm})_{wb}} = \left(\frac{370}{950}\right)^{-\alpha_{wb}}$$

$$b_{abs}(\lambda) = b_{abs}(\lambda)_{traffic} + b_{abs}(\lambda)_{wb}$$

where the wood burning and traffic absorption components are $b_{abs}(370)_{wb}$ and $b_{abs}(950)_{traf}$, respectively.

Combining the above equations gives the following matrix:

$$\begin{bmatrix} b_{abs}(370) \\ b_{abs}(950) \end{bmatrix} = \begin{bmatrix} \left(\frac{370}{950}\right)^{-\alpha_{traf}} & 1 \\ 1 & \left(\frac{370}{950}\right)^{+\alpha_{wb}} \end{bmatrix} \begin{bmatrix} b_{abs}(950)_{traf} \\ b_{abs}(370)_{wb} \end{bmatrix}$$

$$\begin{bmatrix} b_{abs}(370) \\ b_{abs}(950) \end{bmatrix} = \begin{bmatrix} 1 & \left(\frac{370}{950}\right)^{-\alpha_{wb}} \\ \left(\frac{370}{950}\right)^{+\alpha_{traf}} & 1 \end{bmatrix} \begin{bmatrix} b_{abs}(370)_{traf} \\ b_{abs}(950)_{wb} \end{bmatrix}$$
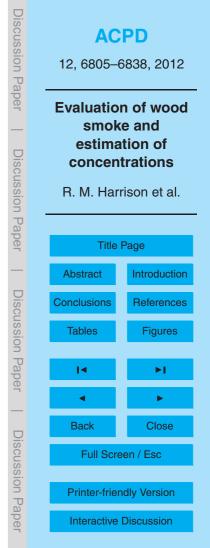
Having solved for the wood burning and traffic absorption components, $b_{abs}(370)_{wb}$ and $b_{abs}(950)_{traf}$ they are regressed against the total daily carbonaceous matter concentrations, CM

$$CM(PM_{2.5}) = c1 \times b_{abs}(950\,nm)_{traffic} + c2 \times b_{abs}(370\,nm)_{wb}$$

where each term corresponds to $PM_{traffic}$ and PM_{wb}. In this work, the coefficients took the following values: c1 = 196 438 and c2 = 351 591 $\mu g\,m^{-2}$.

During the course of the work, the Delta-C method was proposed by Wang et al. (2011) and data have also been analysed according to this method. Delta-C was simply taken as the difference between the BC values measured at 370 nm and 950 nm using the aethalometer and the in-built absorption coefficient values.

$$Delta\text{-}C = BC_{370\,nm} - BC_{950\,nm}$$
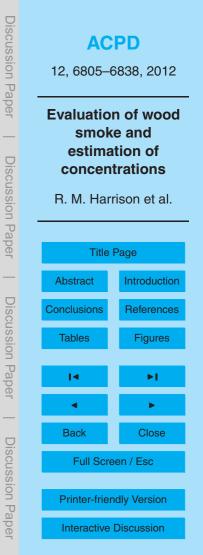
3   Results

3.1   Elms Road Observatory Site (EROS), Birmingham

This was the longest campaign, lasting in all for 11 months. The measured concentrations of levoglucosan are summarised in Fig. 1. This shows the expected seasonal variation with elevated concentrations from October to February when home heating is required and consistently very low concentrations of around $0.01\,\mu g\,m^{-3}$ from March to September. Basic statistics and ratios of levoglucosan/K are summarised in Table 1 and the relationship between the variables appears in Fig. 2. No clear seasonal variation in ratio was evident. Calculation of wood smoke concentration from the aethalometer data (Fig. 3) using the default coefficients gave a diurnal profile of $PM_{traffic}$ as is normally observed for CO or NO_x from traffic. However, the weekday diurnal pattern for estimated wood burning smoke (PM_{wb}) is rather surprising, especially the concentration reductions during traffic rush-hour periods which look as if they may be an artefact of the method. The different diurnal profile and higher concentrations seen on weekends are more in line with expectations.

3.2   North Kensington

Data from this site were limited to two campaigns in summer and winter respectively, rather than sampling through the entire year. Basic statistics and ratios of levoglucosan/K appear in Table 1, and the relationship between the variables in Fig. 2. The summer and winter data are differentiated in Fig. 2 showing the clear seasonal dependence of concentrations, but no obvious systematic difference in levoglucosan/K ratios between seasons. The middle panels in Fig. 3 show diurnal variations in PM from traffic and wood smoke estimated from the aethalometer data using the default coefficients. In this case, these conform to expectations in terms of the shape of both profiles, with a typical rush hour-related pattern to the traffic-related concentrations and a nocturnal elevation in the wood smoke, with a lesser diurnal variation at weekends when houses

may be occupied throughout the entire day.

In the case of the N. Kensington data, a mass of traffic particles was estimated and regressed upon the mass of black carbon (BC). This showed a very close relationship.

$$PM_{2.5}traffic = 1.57(\pm0.03)BC - 0.03(\pm0.05); r^2 = 0.97$$

The gradient is consistent with a primary OC:EC ratio in traffic emissions of 0.3–0.4 estimated by Harrison and Yin (2008) and Pio et al. (2011), together with an OM:OC ratio of around 1.5, which is high suggesting a modest over-estimation of the traffic particle mass.
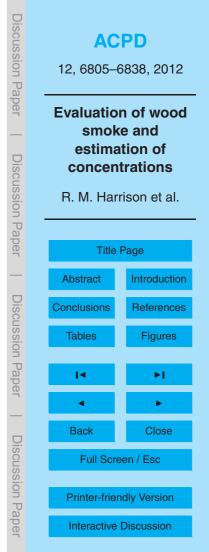
3.3 Marylebone Road, London

Air samples were collected over the same one month period as the winter campaign at North Kensington. The sites are separated by about 4 km and the difference between the two has often been used to infer a traffic contribution. A summary of the measured data appears in Table 1, showing concentrations of levoglucosan and K at Marylebone Road to be very similar in magnitude to those at North Kensington. Regression plots between the sites gave the following relationships:

for K_{ws} $\qquad$ MR = 0.87NK − 0.015 µg m^{-3} $\qquad$ $r^2 = 0.85$

for levoglucosan $\quad$ MR = 1.141NK − 0.002 µg m^{-3} $\qquad$ $r^2 = 0.57$

(Note: The subscript "ws" has been used to distinguish the results of chemical tracer studies from those derived from the aethalometer, designated "wb".)

These gradients of close to one and moderate to high r^2 coefficients strongly suggest a rather uniform distribution of wood smoke markers across central London. The lower r^2 coefficient for levoglucosan than potassium is not readily explained, but may reflect greater localised emissions of the former than the latter marker, but this is hard to explain as the Marylebone Road site is surrounded largely by institutional and commercial buildings, rather than domestic, and this has the slightly higher levoglucosan concentrations.

3.4 Budbrooke, Warwick

This campaign ran from November 2009 to April 2010 and therefore included the season most impacted by wood smoke. As outlined above, the site was subject to the influences both of local and regional sources of wood smoke from home heating and from forestry activities. The latter involved burning of wood in the open air, and hence a wide ratio of levoglucosan/K ratios was expected, and was observed (see Table 1). Another consequence of the mixed sources was that the correlation between levoglucosan and K seen in Fig. 2 was much weaker than for the other sites. At this site the levoglucosan/K ratio showed a strong seasonality (Fig. 4) not seen at the other sites. We interpret this as reflecting a greater contribution to wood smoke from open burning in the warmer months, with the low ratio in January dominated by the home heating emissions.
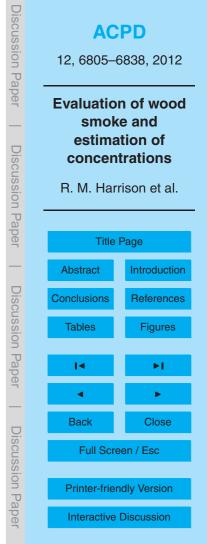
The analysis of aethalometer data for this site using the default coefficients (see Fig. 3) showed profiles rather similar to North Kensington, but with much higher PM_{wb} than $PM_{traffic}$ mass concentration.

4 Discussion

4.1 Levoglucosan: Potassium (levoglucosan/K_{ws}) ratios

The range of levoglucosan/K ratios (10–90th percentile) shown for all sites in Table 1 is considerable. Negative values arise from occasionally negative values of K_{ws}, probably associated with advection of soils with a low K/Ca ratio, therefore causing an excessive correction. The seasonal difference for N. Kensington in the median, but not the mean is in the expected direction, with summer garden bonfires expected to have a higher levoglucosan/K_{ws} ratio than winter emissions from wood burning stoves. However, this is an area of London with high density housing where garden bonfires are expected to be infrequent. Greater degradation of levoglucosan in summer than winter (Hennigan
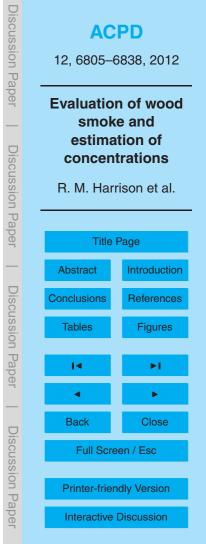
et al., 2010) would be expected to reduce levoglucosan/K$_{ws}$ ratios in summer. Taking the gradients of Fig. 2 as the best representation of levoglucosan/K$_{ws}$ ratios, these were respectively 0.74, 0.34, 0.18 and 0.15 for Budbrooke, EROS, Marylebone Road and N. Kensington respectively and fall well outside the range cited by Puxbaum et al. (2007) from published studies. The reported data from Puxbaum et al. (2007) range from ratios of 1.1 for *quercus* in a US fireplace (Schauer et al., 2001) to 50 for spruce in an Austrian wood stove (Schmidl, 2005), both for PM$_{10}$. Ranges reported for PM$_{2.5}$ are narrower, from 33.3 for softwood in a US woodstove (Fine et al., 2004) to 6.25, for softwood in a US fireplace (Fine et al., 2001). Caseiro et al. (2009) also provided a review of ratios from the recent literature, ranging from 0.11 for PM$_{2.5}$ in a forest fire (Pio et al., 2008) to 200 from softwood burned in a woodstove (Schmidl et al., 2008). Chow et al. (2007) report chemical profiles of hardwood and softwood combustion in which levoglucosan/K ratios are 0.77 and 0.15 for hardwood and softwood, respectively, nicely spanning the values measured in this study.

There appears to be a huge variation in levoglucosan/K ratio according to the wood type. According to data reported by Goncalves et al. (2010) derived from combustion studies in an Austrian-design wood stove, levoglucosan/K ratios can vary from as low as 0.19 for *acacia* to 4.9 for *eucalyptus* with *pinus* and *quercus* in between, with no systematic variation between hardwoods and softwoods.

Schmidl et al. (2008) measured a range of organic compounds and metals in emissions from an old type of domestic tiled stove (from 1994). The reported levoglucosan/K ratios ranged from 19.5 to 216 for the various wood types. However, these authors commented that the abundance of ionic species (including K) in their work was lower than in many previous studies due to samples being taken at the start and middle of the burn. This is likely to have not only biased the K data downwards, it will also have biased the levoglucosan data upwards leading to unreliable levoglucosan/K ratios if considering the entire combustion.

The ranking of levoglucosan/K ratios of Budbrooke > EROS > N. Kensington ~ Marylebone Road is in the order anticipated if the contribution from smouldering

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combustion in open air sources leads to high levoglucosan/K ratios, given the nature of the activities local to the three sites. However, the absolute magnitude of the levoglucosan/K_{ws} ratios is clearly out of line with earlier studies, and requires explanation. The form of the levoglucosan/K_{ws} relationships revealed by Fig. 2 is not suggestive of any systematic over-estimation of K_{ws} or under-estimation of levoglucosan as might be reflected in a large intercept. The concentrations range from low in summer to high in winter as might be anticipated (Fig. 1), and relate inversely to temperature (Fig. 5).

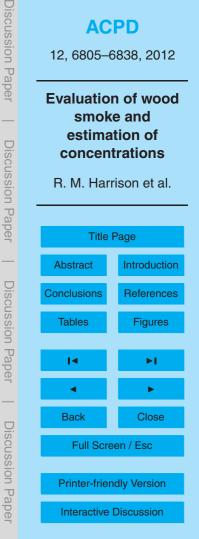Three explanations for low levoglucosan/K_{ws} relationships appear plausible:

1. the levoglucosan/K_{ws} ratio is highly sensitive to combustion conditions with high temperature flaming combustion leading to a more complete burn-out of the organic carbon content and hence a low levoglucosan/K_{ws} ratio. Levoglucosan as a proportion of wood smoke particles is reported to range from 43 to 309 mg g^{-1} (Puxbaum et al., 2007) in published studies, while according to Khalil and Rasmussen (2003), the abundance of K in wood smoke can range from 1.6–102 mg g^{-1}. While these are not independent of one another, there still appears to be much scope for levoglucosan/K ratios well outside the range of those reported in the literature. Kelz et al. (2010) compared particulate matter emissions from modern state-of-the-art and older combustion appliances. The modern devices included a pellet boiler, wood chip boiler, logwood boiler, logwood stove and tiled stove, and the older devices comprised a logwood boiler and logwood stove. The particulate matter from the modern pellet boiler was almost 100 % inorganic, while the modern woodchip boiler and logwood boiler emitted particles with < 20 % carbonaceous content. Particulate emissions from the other four devices were predominantly carbonaceous with ca. 60–90 % elemental carbon and organic compounds. The emissions from the three modern devices with lowest carbon content contained 70–90 % alkali metal sulphates, chlorides or carbonates (Kelz et al., 2010) in which potassium dominated with only minor amounts of sodium present (T. Brunner, personal communication, 2011). Boman et al. (2004) reported that alkali metals dominate the mass of inorganic particulate matter from

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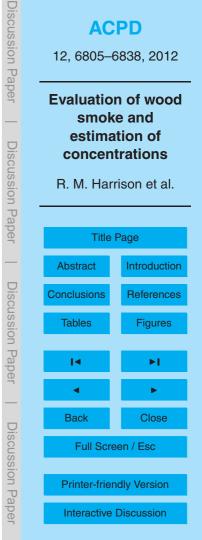Printer-friendly Version

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the combustion of pelletised biomass fuels. Potassium and chlorine were reported to be the dominant elements. There is a lack of information on the relative use of open fireplaces and wood burning stoves in UK homes, or on the types of wood burning stoves most in use. However, there has been a considerable increase in the use of wood as a domestic fuel in recent years, accompanying the rapid rise in the prices of natural gas and electricity. There may, therefore, be a preponderance of modern designs offering a more efficient combustion and lower levoglucosan/K ratios. There has also been a significant uptake of larger wood-burning heating systems for schools and office buildings, etc.

2. The atmospheric lifetimes of levoglucosan and K_{ws} may be different. Levoglucosan has a potential for atmospheric degradation (Hennigan et al., 2010; Hoffman et al., 2010), whereas K does not. The lifetime of levoglucosan with respect to reaction with a typical summer concentration of hydroxyl radical is estimated as 0.7-2.2 days (Hennigan et al., 2010). As winter concentrations of hydroxyl are lower than those in summer, such degradation will be appreciably slower in winter. This alone therefore seems unlikely to influence the levoglucosan/K ratio greatly, even if the majority of wood smoke is advected from distant sources. Appreciable decomposition of levoglucosan would lead to higher levoglucosan/K ratios in winter than summer, for which Table 1 gives no clear support, with little systematic difference between the seasons, except at Budbrooke, where summer ratios are higher for reasons explained earlier in this paper. Another possible influence is from differential lifetimes relative to deposition. It is likely that levoglucosan and K co-exist in the same particles, but with a range of ratios which may depend upon particle size. No information is available on the respective size-associations, but as the sampling was of the $PM_{2.5}$ fraction, it seems likely that the atmospheric lifetime will be several days and any differences between levoglucosan and K will be a minor influence upon the airborne ratios.
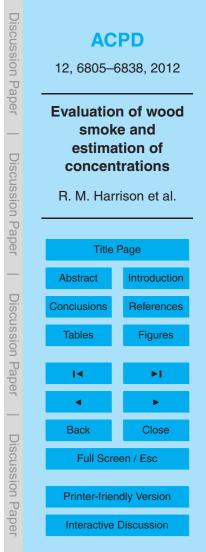
3. There is an unrecognised source of K. To be significant, this would need to be larger in winter than summer and to correlate with levoglucosan. While this possibility cannot be entirely excluded, the probability is thought to be low.

4.2 Comparison with Delta-C method

Wang et al. (2011) have recently shown that the enhanced absorption between 370 nm and 880 nm in the two wavelength aethalometer (referred to as Delta-C) correlates with wood smoke markers, although they do not propose a factor with which to calculate wood smoke masses. Delta-C is calculated using the conventional absorption coefficient for black carbon at both wavelengths and taking the difference in estimated black carbon concentration. The gradient observed by Wang et al. (2011) in their winter data (giving the greatest range of concentrations) was Delta-C/levoglucosan = 5.9. In our data, the ratio Delta-C/levoglucosan at the Budbrooke site was 4.5 ($r^2 = 0.25$) and at the North Kensington site (winter data) was 6.5 ($r^2 = 0.25$). Wang et al. (2011) also showed a relationship to potassium although this was not corrected for sea salt or soil. Our data for both sites showed a very poor relationship to K_{ws}. Figure 6 shows the black carbon measurements at the two wavelengths together with the calculated values of Delta-C in the Budbrooke and North Kensington winter datasets. These diurnal profiles in Delta-C with the strong evening maximum look extremely plausible but it needs to be borne in mind that Delta-C is a surrogate for wood smoke mass and is not a direct measurement of wood smoke mass. At Budbrooke, the gradient of the Delta-C/K_{ws} plot was 4.0 ($r^2 = 0.2$) and at North Kensington (winter) the two variables were uncorrelated. Wang et al. (2011) found a Delta-C/K slope of 6.2 ($r^2 = 0.63$) in their winter dataset, and lower values in spring and fall, broadly consistent with the ratio measured at Budbrooke. The generally low correlations between Delta-C and K_{ws} seen in our data, and the fact that these fall below the values of the levoglucosan-K_{ws} correlations is suggestive of an interference (possibly by SOA) in the Delta-C method.
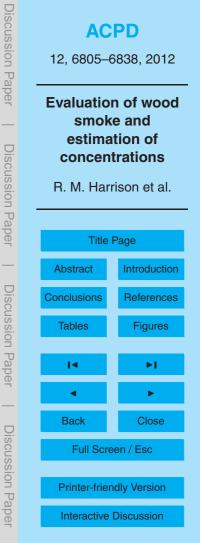
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4.3   Estimation of wood smoke concentrations

From single marker data, it is very difficult to estimate the airborne wood smoke concentrations. The factor of 10.7 between levoglucosan and wood smoke mass relating to Austrian conditions reported by Schmidl et al. (2008) has been widely adopted, but as indicated above, this may be an over-estimate for the entire combustion process. Our levoglucosan/K ratios are entirely out of line with Schmidl et al. (2008) and hence we think it inappropriate to use these data directly. The relationship between levoglucosan and K_{ws} is considerably less close at our sites than was reported by Caseiro et al. (2009) from measurements in Vienna, Graz and Salzburg. It is also lower than that reported by Saarikoski et al. (2008) between monosaccharide anhydrides and potassium in four central and northern European cities. Additionally, the relationship between Delta-C and levoglucosan is weaker than that observed by Wang et al. (2011) despite the fact that their data were not corrected for potassium from other sources. This gives rise to the question as to whether more than one source type is influencing the measured data. At Budbrooke, this is clearly the case with influences both of wood stoves and open burning of wood which would be expected to give different K_{ws}/levoglucosan relationships. However, the situation is less clear for the other sites.

A survey of consumption of solid fuel in the UK and Ireland (Ecosolidfuel, 2008) has suggested the following split: open fireplace 11.5 %; closed fireplace 22.1 %; pallet stove 0.1 %; domestic boiler 66.3 %. However, the energy consumption in this form of around 75 PJ yr^{-1} is far smaller than the 322 PJ yr^{-1} used in non-domestic boilers with outputs of up to around 500 KW (R. Stewart, personal communication, 2011). The latter are more likely to be used in cities, where in the UK smoke control regulations require combustion of solid fuels in devices demonstrated to meet specific emission requirements. It consequently seems likely that the UK urban emissions affecting sites such as EROS and London North Kensington comprise a mixture of wood burning in open fireplaces and closed devices which are likely to be relatively rich in levoglucosan relative to potassium. On the other hand more advanced domestic boilers and non-

domestic boilers having a much lower levoglucosan to potassium ratio as exemplified by the modern devices tests by Kelz et al. (2010). Consequently, it is difficult to propose a specific factor between either levoglucosan or K_{ws} concentrations and the mass of wood smoke particles.

Puxbaum et al. (2007), based on a review of literature data available at the time, propose use of the relationship:

$$\text{Biomass smoke OC} = \text{levoglucosan} \times 7.35$$

According to the studies reviewed by Puxbaum et al. (2007), OC represents between 51.3 and 100 % of PM from biomass combustion. Using the data for fireplaces and wood stoves presented by Puxbaum et al. (2007), the relationship between biomass smoke mass and levoglucosan can be derived and the various studies reviewed there give a range of 4.51 to 24.6 with a mean value of 11.2. If this value is adopted and applied to the measured levoglucosan data, the following average concentrations are derived:

Birmingham, EROS (May 2009–March 2010) $0.23\,\mu g\,m^{-3}$

London, North Kensington (June 2010 and February/March 2011) $0.33\,\mu g\,m^{-3}$

Budbrooke, Warwickshire (November 2009–April 2010) $0.42\,\mu g\,m^{-3}$

The literature is far less clear on the relationships between K_{ws} and biomass smoke mass. However, given the very high ratios between K_{ws} and levoglucosan, we think it likely that there is a separate source category of highly efficient larger biomass burning installations generating emissions with a very low organic content which would have little influence upon airborne levoglucosan. Judging from the work of Kelz et al. (2010), it is quite feasible for K_{ws} to make up around 50 % of the mass of such aerosol which would imply an additional contribution of biomass burning to $PM_{2.5}$ at

the urban locations with very low levoglucosan/K_{ws} ratios as follows:

Birmingham, EROS (May 2009–March 2010) $0.08\,\mu g\,m^{-3}$

London, North Kensington (June 2010 and February/March 2011) $0.30\,\mu g\,m^{-3}$

If there are two distinct source types, i.e. on the one hand wood burning in relatively
5 inefficient small appliances such as wood stoves and fireplaces, and on the other hand
combustion in larger more efficient installations, then the two sets of concentrations
should be approximately additive for a given location.

4.4 Comparison with earlier UK data

There are two sets of earlier data for the Birmingham, EROS site with which it is rele-
10 vant to intercompare. Yin et al. (2010), using a Chemical Mass Balance (CMB) model,
calculated source contribution estimates for wood smoke as a contributor to $PM_{2.5}$ as
$0.07\,\mu g\,m^{-3}$ in summer and $0.08\,\mu g\,m^{-3}$ in winter. In the light of our current data, the
lack of an obvious seasonal variation is surprising but the sampling regime involved
sampling for only five days of each month as opposed to daily in the current work and
15 it is possible that the concentrations measured were not wholly representative of the
overall period of sampling. Nonetheless, the two datasets are not strongly out of line in
terms of the magnitude of concentration. More recent unpublished data also using the
CMB method have shown higher concentrations at North Kensington of $0.23\,\mu g\,m^{-3}$
(summer campaign) and $0.36\,\mu g\,m^{-3}$ (winter campaign) which are far closer to the cur-
20 rent dataset.

Further indirect evidence is available from the study of Heal et al. (2011) in which
^{14}C was measured in aerosol collected at the EROS site in 2007/08. In all, some 75
$PM_{2.5}$ samples were collected between June to September 2007 and January to May
2008 and a subset of 26 samples were analysed for their "fraction modern carbon". The
25 contemporary elemental carbon is interpreted as arising from biomass combustion and
this again showed rather little seasonal variation which could once again be an artefact
of the days selected. The average contemporary elemental carbon was only 2 % of
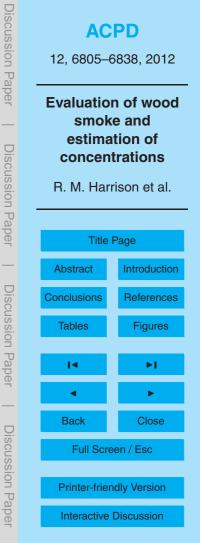
total carbon equating to around 0.1 µg m^{-3}. If this is assumed to be 20 % of biomass smoke (Schmidl et al., 2008), this equates to a concentration of 0.5 µg m^{-3} of biomass smoke. The figure of EC being 20 % of biomass smoke is highly uncertain – for example Minguillon et al. (2011) assume a ratio of EC/OC = 0.3 in biomass smoke which implies an EC content of below 14 %. The value of 0.5 µg m^{-3} of biomass smoke, although very uncertain due to the difficulty of achieving an accurate split between elemental and organic carbon in the samples and the uncertainty surrounding the %EC in biomass smoke, is not strongly out of line with the estimate for EROS derived from levoglucosan analysis above.

The only other UK data of which we are aware are from Fuller et al. (2011) who reported mean winter levoglucosan in London of 176 ng m^{-3}, almost four times the mean concentrations measured in our study in winter at North Kensington. They equate this to about 3 µg m^{-3} of wood smoke, which would cause a marked seasonal variation in PM$_{10}$ which is not observed.

5   Conclusions

This work is suggesting that annual mean concentrations of biomass smoke at UK sites are very low at well below 1 µg m^{-3} and very much lower than some other estimates for the UK (Fuller et al., 2011). It is recognised that these concentrations are far below those measured elsewhere in Europe (e.g. Puxbaum et al., 2007, Bari et al., 2010) but there may be good reasons for this. Most modern urban dwellings in the UK no longer have fireplaces and the fitting of wood stoves is very difficult due to the lack of a flue and chimney. Most UK urban areas are subject to smoke control orders which prohibit the use of many kinds of solid fuel other than in devices designed to burn them smokelessly. There are very limited supplies of logs and wood products available within urban areas for purchase by domestic consumers for combustion. Consequently, burning of wood in domestic premises is largely limited to rural areas of the UK and the fact that levoglucosan concentrations correlate so strongly between the Marylebone Road and

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North Kensington sites may imply an advected rural source of wood smoke. While the datasets from Birmingham, EROS and London, North Kensington show a marked seasonal variation, the estimated wood smoke masses are insufficient to cause a significant perturbation in $PM_{2.5}$ or PM_{10} concentrations such as would lead to a distinct seasonal pattern. Such a pattern is not observed at UK urban sites (see for example Bigi and Harrison (2010) for London, North Kensington), further supporting the contention that wood smoke concentrations in London and Birmingham are relatively low.

The multi-wavelength aethalometer calculations suggest higher concentrations but we discount these on the basis of their inconsistency with the levoglucosan data and the fact that the aethalometer method when applied to UK locations will be subject to interference from HULIS (humic-like substances) within secondary organic aerosol. Since this interference is variable, there is no correction for it and therefore the method may strongly over-estimate concentrations. The fact that the relationship between Delta-C and levoglucosan in our measurements is similar to that obtained by Wang et al. (2011) in the United States suggests that we can place trust in the levoglucosan data and ignore the more extreme concentration estimates deriving from application of the method of Sandradewi et al. (2008a,b) which was based originally upon data collected in a Swiss Valley where wood smoke and traffic emissions were expected to be the only significant sources of absorbing aerosol.

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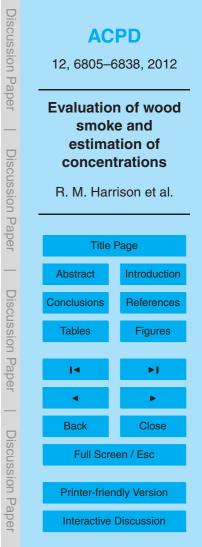
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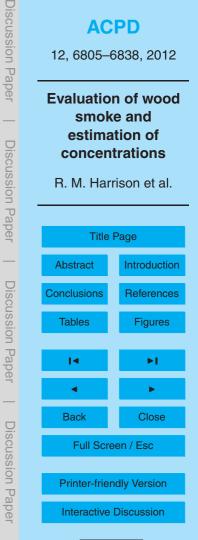
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Table 1. Summary of levoglucosan and potassium (K_{ws}) data ($\mu g\,m^{-3}$) and levoglucosan/K_{ws} ratios.

Site	Season*	Levoglucosan 10th percentile/90th percentile	Mean	Median	K_{ws} 10th percentile/90th percentile	Mean	Median	Levoglucosan/K_{ws} 10th percentile/90th percentile	Mean	Median
EROS	Winter	0.008/0.058	0.031	0.026	−0.011/0.177	0.074	0.075	−0.534/1.074	0.295	0.331
	Summer	0.005/0.02	0.010	0.009	−0.018/0.037	0.007	0.002	−2.115/2.26	0.477	0.281
North Ken.	Winter	0.022/0.069	0.045	0.044	0.073/0.444	0.251	0.236	0.111/0.455	0.356	0.165
	Summer	0.007/0.023	0.014	0.012	0.02/0.092	0.049	0.045	0.146/0.517	0.307	0.316
Marylebone Rd	Winter	0.024/0.085	0.050	0.046	0.039/0.362	0.204	0.221	0.141/1.202	1.349	0.249
Budbrooke	Winter	0.012/0.14	0.059	0.039	−0.001/0.165	0.076	0.053	−0.459/2.346	0.168	0.656
	Summer	0.007/0.029	0.016	0.013	0.017/0.057	0.037	0.035	0.326/0.723	0.495	0.427

* Winter: November to March

Summer: April to October

Fig. 1. Monthly variation of levoglucosan measured at EROS between 23/06/2008 and 31/03/2010.

$Y = 0.34_{(\pm0.02)} X + 0.009_{(\pm0.002)}$ $r^2 = 0.44$

$Y = 0.15_{(\pm0.01)} X + 0.007_{(\pm0.003)}$ $r^2 = 0.68$

$Y = 0.74_{(\pm0.06)} X + 0.002_{(\pm0.007)}$ $r^2 = 0.27$

$Y = 0.18_{(\pm0.03)} X + 0.013_{(\pm0.008)}$ $r^2 = 0.32$

Fig. 2. Relationship between levoglucosan and K_{ws} measured at EROS (04/05/2009–31/03/2010) (top left plot) and Budbrooke (19/11/2009–08/04/2010) (bottom left plot) and North Kensington (filled circles represent winter) (16/02/2011–15/03/2011) and open circles represent summer (03/06/2010–29/06/2010) (top right plot) and Marylebone Road (16/02/2011–15/03/2011) (bottom right plot). All are fitted using a RMA regression.

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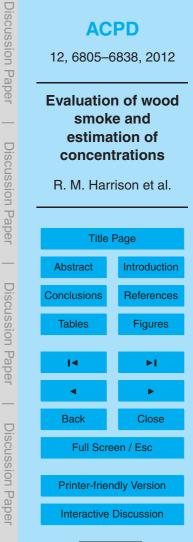◄ | ►

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Fig. 3. Diurnal variation of PMwb and PMtraf measured at EROS (04/05/2009–31/03/2010) (left panel) , North Kensington (16/02/2011–15/03/2011) (middle panel) and Budbrooke (20/11/2009–08/04/2010) (right panel). (Top panel plot: average day including both week and weekends; middle panel plot: average week day; bottom panel plot: average week end day)
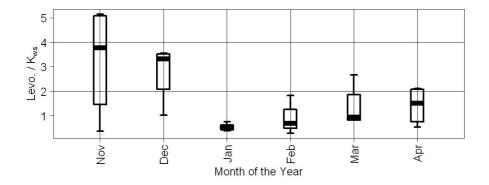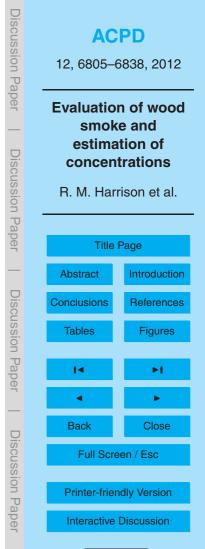
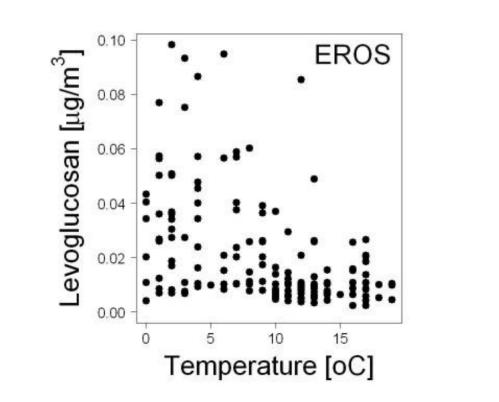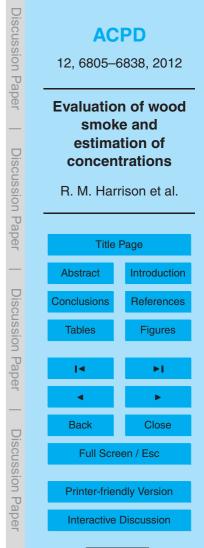**Fig. 4.** Ratio of levoglucosan: K_{ws} at Budbrooke according to month of observation.

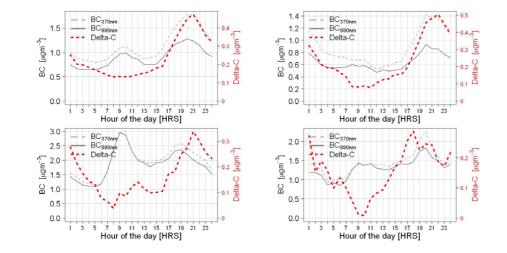**Fig. 5.** Variation of levoglucosan with temperature at EROS site.

Fig. 6. Average diurnal variation in black carbon estimated at 370 and 880 nm with the aethalometer and Delta-C at Budbrooke (top panel) and North Kensington, winter (bottom panel). Left plot: weekday; right plot: weekend.