Airborne particles from wood burning in UK cities

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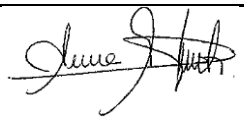
Lead authors	Anna Font		March & July 2017
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Summary

It is believed that domestic wood burning in the UK has been systematically underestimated by a factor of three in the national emissions inventory (Waters 2016) and there is therefore an important need to properly quantify its contribution to the urban atmosphere. This study aimed to quantify particulate matter (PM) from wood smoke in cities in the UK using aethalometer data from Defra's black carbon network from 2009 to 2016.

In addition to measuring black carbon concentrations, aethalometers provide information on the light absorption of the collected particles at two different wavelengths. These measurements were processed to quantify the particle matter from wood burning.

As expected air pollution from wood burning was greatest in winter and almost absent in summer. Mean wintertime PM from wood burning varied between cities, ranging between 0.2 and 2.7 µg m^{-3}. In Cardiff, 13% of PM_{10} in winter 2009/10 came from wood burning.

On annual basis wood burning in $PM_{2.5}$ ranged between 4 to 6% as an average across rural areas; and between 6 to 9% averaged across urban areas. It should be remembered that the majority of PM_{10} and $PM_{2.5}$ in urban and rural areas is not from primary emissions. Instead the majority comes from reactions between other gaseous pollutants forming secondary particles. It was estimated that wood burning was between 23 and 31% of the urban derived $PM_{2.5}$ in London and Birmingham, making control of wood burning an important urban issue.

On the whole the proportion of wood burning in PM_{10} did not increase on days when the EU limit value was breached. Instead wood burning was more important in determining mean concentrations. From a policy perspective the ambient impacts from wood burning will have greatest effects on the public health framework indicators, based on average $PM_{2.5}$, and on the $PM_{2.5}$ exposure reduction enshrined EU directives. It will also affect progress towards the attainment of WHO annual mean guidelines for these pollutants.

In most cities wood burning PM concentrations were greater in evenings, indicating residential combustion, and greater at weekends. Coupled with the poor correlation with daily temperature (R^2 = 0.12- 0.57) this suggested that current urban wood burning was in large part decorative and was not being used primary heating.

Between 2009 and 2015 trends in PM from wood burning were slightly downwards at -0.03 (-0.05, -0.01) µg m^{-3} $year^{-1}$. This was unexpected given anticipated growth in biomass combustion due to policy initiatives aimed at increasing production of heating and electricity from renewable and low-carbon energy sources. UK Industry data also suggests stove sales are running at between 150,000 and 200,000 units per year with over one million stoves sold between in 2010 and 2015. One possible explanation for the observed trend is the replacement of high emission fireplaces with newer, and lower emission wood stoves, balancing an increase in total wood heating.

There is no reference method for estimating PM from wood burning in ambient air. Several methods exist. They are generally well correlated in international studies and this was borne out in comparisons carried out in London. We can therefore have good confidence in attribution of wood burning to be from residential heating. Similarly we can be confident in the relative spatial distributions. Although our method gives results that were close to the mean of other methods the

spread between the concentration estimates is wide at +- 70%. This needs to be considered when comparing concentrations to emissions inventory estimates.

Looking forward, large increases in biomass burning are projected from energy scenarios over the next two decades (Williams et al., 2017). An increase in the coverage of the aethalometer network would enhance our ability to track the impact of these changes. A surveillance programme would also need to include long-term measurements with multiple methods at key locations.

1. Background

While wood burning is a common practice of residential heating in rural areas, it is starting to become a generalized practice in urban areas contributing to ambient particulate matter (PM) concentrations. In the United Kingdom (UK) a substantial growth in biomass combustion is anticipated due to policy initiatives aimed at increasing production of heating and electricity from renewable and low-carbon energy sources. These policies include the Renewable Heat Incentive, Feed-in Tariffs and the Merton Rule for on-site renewable heating in new buildings. Biomass fuels were also included in the European Commissions' strategy for reaching the "202020" targets (20% reduction in greenhouse gas emissions; 20% use of renewable energy sources; and 20% increase in energy efficiency by 2020 (ECF, 2010). The adopted policies for renewable energy sources are expected to double biomass use by 2030 compared to 2005 in the EU-28 (Amann et al. 2014). A recent UK government survey (Waters 2016) found that 7.5% of UK homes now burn wood. However, there is considerable uncertainty about the frequency of fire use, the balance between stoves and fireplaces and therefore the total emission.

Smoke from wood burning is made up of a mixture of gases such as carbon monoxide (CO) and nitrogen oxides (NO_X) and fine particles. Earlier work in London found that the PM_{10} (PM with aerodynamic diameter < 10 μm) from home wood burning in 2010 represented ~10% of the winter PM_{10} averages in inner London exceeding the PM_{10} reductions from the first two phases of the London Low Emission Zone (Fuller et al 2014). Wood smoke also contain several toxic air pollutants including benzene, formaldhyde, acrolein and polycyclic aromatic hydrocarbons (PAHs) such as Benzo[a]pyrene (B[a]P), contributing to the deterioration of urban air quality. The National Atmospheric Emissions Inventory (NAEI; naei.defra.gov.uk) estimates that residential and commercial combustion has been the main emitter of B[a]P to the atmosphere since 1999. This includes the use of solid fuel, such as wood for domestic heating (Tompkins et al. 2016).

Epidemiological studies have evidenced the link between short-term exposures to PM from wood smoke and adverse health effects (such as asthma, respiratory symptoms, daily mortality, and lung function) and there is no evidence than particles from wood smoke are less toxic than other particles from other sources (Boman, Forsberg, and Järvholm 2003; Naeher et al. 2007). The toxicity of wood smoke particles seems to strongly depend on the organic fraction, and it is probably associated with organic components other than PAHs (Bolling et al. 2012). An estimated 61,000 premature deaths in Europe in 2010 were attributable to outdoor $PM_{2.5}$ from residential heating with solid fuels (either wood or coal) (WHO, 2015). Also, wood smoke also impacts on ecosystem and the environment, reducing visibility (haze) and creating environmental and aesthetic damage.

It is believed that domestic wood burning in the UK has been systematically underestimated by a factor of three in the national emissions inventory (Waters 2016) and there is therefore an important need to properly quantify its contribution to the urban atmosphere. This study aims to quantify the wood smoke mass concentrations in cities in the UK using aethalometer data from Defra's black carbon network from 2009 to 2016. This is the first time that the quantification of wood burning mass concentration has done systematically across different cities in a given country over several years.

2. Methods

2.1 Monitoring sites and temporal cover

PM concentration from wood burning (C_{wood}) was calculated for the urban background, suburban and rural sites belonging to the Defra's black carbon network (Butterfield et al., 2013; 2015). This comprises a total of 21 sites distributed in 16 cities and 4 rural areas. Additionally, the kerbside site measurements undertaken at Marylebone Road site in London were also considered to parameterise the model (see section 2.2).

The analysis comprises the period between 1st January 2009 and 29th February 2016. However, the time span with available measurements for each site does not cover the whole period due to a restructuration of the network in 2012 (Table 1). Winter averages comprised the month of December and the following January and February (DJF).

Table 1. Site information of monitoring sites belonging to Defra's black carbon network.

Name	*Lon*	*Lat*	*Type*	*Start Year*	*End Year*	*Additional measurements*
Auchencorth Moss	-3.245	55.793	Rural	2012	--	PM_{10}, $PM_{2.5}$, NO_X, BaP
Bath	-2.355	51.389	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Cardiff	-3.164	51.489	Urban Background	2009	2015	
Dudley Central	-2.097	52.512	Urban Background	2009	2012	
Edinburgh St Leonards	-3.182	55.946	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Goonhilly Cornwall	-5.185	50.046	Rural	2012	2013	
Glasgow Centre	-4.256	55.858	Suburban	2009	2012	PM_{10}, $PM_{2.5}$, NO_X, BaP
Glasgow Townhead	-4.244	55.866	Urban Background	2013	--	PM_{10}, $PM_{2.5}$, NO_X, BaP
Woolwich	0.073	51.473	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Harwell	-1.327	51.571	Rural	2009	2016	PM_{10}, $PM_{2.5}$, NO_X, BaP
Halifax	-1.863	53.713	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Birmingham Tyburn	-1.827	52.513	Urban Background	2009	--	PM_{10}, $PM_{2.5}$, NO_X, BaP
South Kirkby	-1.312	53.594	Urban Background	2009	2012	
Manchester Piccadilly	-2.238	53.481	Urban Background	2009	2012	
London North Kensington	-0.214	51.521	Urban Background	2009	--	PM_{10}, $PM_{2.5}$, NO_X
Nottingham Centre	-1.147	52.955	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Norwich Centre	1.303	52.615	Urban Background	2009	2013	
Sunderland	-1.389	54.920	Urban Background	2009	2012	
Stoke-on-Trent Centre	-2.175	53.028	Urban Background	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Folkestone - Cheriton	1.160	51.087	Suburban	2009	2012	PM_{10}, $PM_{2.5}$, NO_X
Maidstone - Detling	0.582	51.308	Rural	2012	--	PM_{10}, $PM_{2.5}$, NO_X
London Marylebone Rd	-0.155	51.522	Kerbside	2009	--	PM_{10}, $PM_{2.5}$, NO_X, BaP

2.2 The aethalomter model to calculate wood burning mass aeorosols

The PM concentration from wood burning (C_{wood}) was calculated using dual wavelength Magee Scientific Aethalometers (AE-22). The aethalometer model is based on the light absorption behaviours of wood burning and traffic fossil fuel combustion aerosols, with biomass aerosols absorbing more at shorter wavelengths (Kirchstetter et al., 2004; Sandradewi et al., 2008).

The Magee AE-22 aethalometer samples air at 4 l min^{-1} and collects $PM_{2.5}$ particles onto a quartz tape. The light attenuation through the sampled filter at 370 nm (in the UV) and at 880 nm (in the IR) was measured as:

$$ATN_{(\lambda)} = -\ln (I_{(\lambda)} / I_{0(\lambda)}) \quad (1)$$

where I is the intensity of light that passes through the sampled spot on the filter and I_0 is the intensity of light through a clean filter and λ the wavelength.

The aerosol absorption coefficients for each of the wavelengths, $b_{abs}(370)$ and $b_{abs}(880)$, were obtained from attenuation values (*ATN*):

$$b_{abs} = A/Q \cdot \Delta ATN/\Delta t \quad (2)$$

where Q is the flow rate, the t is the sampling time and A the filter spot size (A = 1.67 cm^2). Prior to 2012 the sampling time was 15 minutes; after 2012, at sampling was done at 5 minute intervals. Raw data was corrected for the effect the filter loading using the model developed by Virkkula et al (2007). Ratified and reported data was available as hourly means.

According to the Beer-Lambert law, the absorption of light is dependent on the wavelength (λ):

$$b_{abs} \propto \lambda^{-\alpha} \quad (3)$$

where α is the Ängström exponent. The Ängström exponent describes the wavelength dependency of the absorption coefficient and it can provide information about the predominant aerosol type of source (e.g. Lewis et al., 2008; Cazorla et al., 2013). Black carbon aerosols derived from diesel traffic sources have α ~ 1 (Schnaiter et al. 2003). Wood smoke has a large content of organic material when compared with traffic aerosols and therefore absorbs significantly more radiation in the UV part of the spectrum. This is reflected by higher Ängstrom coefficients for wood smoke (α ~ 2) (Kirchstetter et al., 2004; Sandradewi et al., 2008).

Ängström coefficients were calculated by solving the equation system using both absorption coefficients in the UV and IR channels measured in the 2 wavelength aethalometer:

$$\alpha = \frac{-log\left(\frac{b_{abs(370)}}{b_{abs(880)}}\right)}{log\left(\frac{370}{880}\right)} \quad (4)$$

Assuming that only two sources of aerosols (traffic and wood burning) are present in the sampled atmosphere, the aerosol absorption coefficient at a given wavelength, $b_{abs}(\lambda)$, can be expressed as the sum of the light absorption of aerosols emitted by this two sources.

$$b_{abs}(\lambda) = b_{abs}(\lambda)_{traffic} + b_{abs}(\lambda)_{wood} \quad (5)$$

Both quantities can be obtained from light absorption measurements if the Ängström coefficients from both sources are known. With given $\alpha_{traffic}$ and α_{wood} and the two wavelengths, the following equations can be applied:

$$b_{abs}(370)_{traffic} / b_{abs}(880)_{traffic} = (370/880)^{-\alpha\ traffic} \quad (6)$$

$$b_{abs}(370)_{wood} / b_{abs}(880)_{wood} = (370/880)^{-\alpha\ wood} \quad (7)$$

The carbonaceous material (CM) equals the sum of organic matter (OM) and black carbon (BC), that it can be, on its turn, expressed as the mass concentration from traffic and wood burning aerosols:

$$CM = OM + BC = C_{traffic} + C_{wood} \quad (8)$$

Sandradewi et al. (2008) regressed the mass concentration of total carbonaceous matter against $b_{abs}(880)_{traffic}$ and $b_{abs}(470)_{wood}$ for the determination of the contribution of traffic and wood burning to CM:

$$CM = C_1 * b_{abs}(880)_{traffic} + C_2 * b_{abs}(370)_{wood} + C_3 \quad (9)$$

with C_1 and C_2 relating the aerosol absorption coefficient to the total carbonaceous mass concentration; and C_3 a residual factor corresponding to the amount of non-combustion organic aerosol (Favez et al., 2010).

Equation (9) can be solved when combining with equations (5) - (8) to calculate C_{wood}. For that, we used $\alpha_{traffic} = 0.96$ as in Fuller et al. (2014); and $\alpha_{wood} = 2$ as in Sandradewi et al. (2008); Favez et al. (2010). These values are closer those suggested for dual wavelength aethalometers in a recent Swiss-wide study (Zotter et al. 2016) ($\alpha_{traffic} = 0.9$ and $\alpha_{wood} = 2.09$). $C_2 = 418323$ mg Mm^{-2} calculated as the average used in the studies of Favez et al. (2009, 2010), Sandrewi et al. (2008) and Sciare et al. (2011) and transformed from the 470 nm channel to 370 nm.

2.2.2. Comparative measurements quantifying C_{wood} concentrations

Daily levoglucosan measurements were available for the London North Kensington in winter 2010 between 18th January and 28th February. The sampling and analytic methodology and the results were first reported in Fuller et al., 2014. A factor of 11 was applied to convert levoglucosan measurements to C_{wood} (Fuller et al., 2014). This was in accordance with the conversion factor of 10.7 suggested in Schmidl et al. (2008) and applied elsewhere: Flanders (Maenhaut et al. 2012); Leicester (Cordell et al. 2016); etc.

Hourly measurements from a quadruple aerosol mass spectrometer (AMS, Aerodyne Research Inc., Massachusetts, USA) were available during the ClearFlo campaign (Bohnenstengel et al. 2015) from London North Kensington for the period between 11th January 2012 and 23rd January 2013. AMS provides online measurements of the chemical composition (organic aerosols, nitrate, sulphate, chloride and ammonium) and mass size distribution of the non-refractory fraction of submicrometer aerosols (size range < 1µm). Particles are vaporized at 600°C. Positive matrix factorization analysis (PMF) is widely used in the AMS community for aerosol apportionment using the mass spectra. PMF is a least-squares approach based on a receptor-only multivariate factor analytic model (Paatero and Tapper 1994). Young et al. (2015) applied PMF to the mass spectra measured by the AMS and apportioned the organic aerosols (OA) in hydrocarbon-like OA (HOA), solid fuel OA (SFOA), and oxygenated OA (OOA). SFOA was identified to be likely emitted from domestic space heating given its higher concentrations in the evening and in urban environments such as London, SFOA concentrations were assumed to be from wood burning. SFOA factor was then compared to levoglucosan measurements collocated in the same site.

A latest generation seven wavelength aethalometer (Magee Scientific, A33) was collocated at London North Kensington between 7th December 2015 and 19th February 2016. This new instrument benefits from real-time spot-loading correction. Wood burning concentrations were calculated using the aethalometer method formulation using the absorbance values at 370 and 880 nm.

2.3 Additional data sets

Hourly PM_{10}, $PM_{2.5}$ and NO_X concentrations were extracted from Defra's Automatic Urban and Rural Network (AURN) for the sites with collocated measurements (Table 1). PM_{10} and $PM_{2.5}$ were measured by TEOM-FDMS (Tapered Element Oscillating Microbalance - Filter Dynamics Measurement System) considered equivalent to the EU reference method, which is based on 24-hour

sampling and gravimetric analysis. NO_X (NO + NO_2) was measured by chemi-luminiscence and fortnightly calibrations enabled the traceability of measurements to national metrological standards. All instruments were subject to twice yearly audit tests by the National Physical Laboratory or Ricardo AEA. Benzo[a]pyrene (B[a]P) measurements were available as monthly means as part of Defra's Polycyclic Aromatic Hydrocarbons (PAH). Particles were collected using a Digitel DHA-80 high volume aerosol sampler on glass fibre filters for daily and subsequent analyzed by gas chromatography/mass spectroscopy (GC/MS) of pooled samples. Details about the network and analytical procedures can be found in Tompkins et al. (2016).

Surface air temperature and wind speed was extracted from the nearest meteorological site belonging to the NOAA Integrated Surface Database (ISD) network (https://www.ncdc.noaa.gov/isd).

2.4 C_{wood} as a fraction of particulate matter concentrations and urban derived particle matter.

The contribution of wood smoke mass concentrations to particulate matter was quantified in two ways.

First, for all sites with collocated measurements and with a minimum data capture of 75%, annual and winter C_{wood}/PM_{10} and $C_{wood}/PM_{2.5}$ ratios were calculated.

Second, for those sites with paired urban background – rural observations (sites apart < 100 km), the wood burning contribution to the urban derived PM was calculated by subtracting the rural concentration and the C_{wood} contribution to urban pollution as:

$$C_{wood}/\text{urbanPM} = C_{wood}\ \text{urban} / (\text{PM urban} - \text{PM rural}) \quad (10)$$

$$C_{wood}/\text{urbanPM*} = C_{wood}\ \text{urban} / (\text{PM urban} - \text{PM rural} - C_{wood}\ \text{rural}) \quad (11)$$

2.5 Trends in time

Trends in C_{wood} were calculated using the TheilSen estimator, available from the R-openair package (Carslaw and Ropkins, 2012). Linear trends were calculated from monthly means with a minimum of 75% data capture. Trends were calculated without deseasonalizing (marked as "All data") and deseasonalizing the time series (marked as "All data – deseason"). Since C_{wood} was expected to be more prominent during the winter months, also trends in winter C_{wood} were calculated. For this a linear-square model was fitted to winter concentrations (average of December, January and February). In order to check the robustness of the trends to weather conditions, trends in C_{wood} * wind speed were calculated for winter months. Taking a simple box model approach, the product C_{wood} * wind speed should act as a crude tool to remove the wind speed effects.

The overall trend for each variable across the UK was calculated by fitting the linear Random-Effects Model "DerSimonian-Laird estimator" following the methodology developed in Font and Fuller (2016). The Random-Effects (RE) fit assumes that there are two sources of variation in the data set: the within-site estimation variance (variability in the trend calculated for one site as expressed by the confidence intervals) and between sites (variability of trends among the population of sites) (Borenstein et al. 2010). The graphical representation of the distribution of trends along with individual confidence intervals and the overall trend was done through "Forest plots". Only those sites with at least 75% data capture for the whole period were considered to calculate the overall trends.

A statistically significant trend was assumed when $p < 0.1$ (represented with a '+' symbol), meaning that the trend was not random at a 90% chance; $p < 0.05$, $p < 0.01$ and $p < 0.001$ (marked by '*', '**' and '***', respectively) indicate very high significant trends; and $p > 0.1$ indicate insignificant trends.

The trends in C_{wood} were calculated for two different time spans: from the beginning of January 2009 to end of 2011; and from the beginning of January 2009 to end of 2015. This was done to include all sites that stopped the monitoring with the restructure of the network at the beginning of 2012.

3. Results

3.1 Ångström coefficients

The Ångström coefficients derived from the aethalometer at London Marylebone Road showed median annual values of ~ 1 (annual medians ranging from 1.02 to 1.11 between 2009 and 2015). These values are in accordance with α~1 associated with traffic aerosols from diesel engines as expected due to the proximity of the monitoring site to the road (Figure 1A).

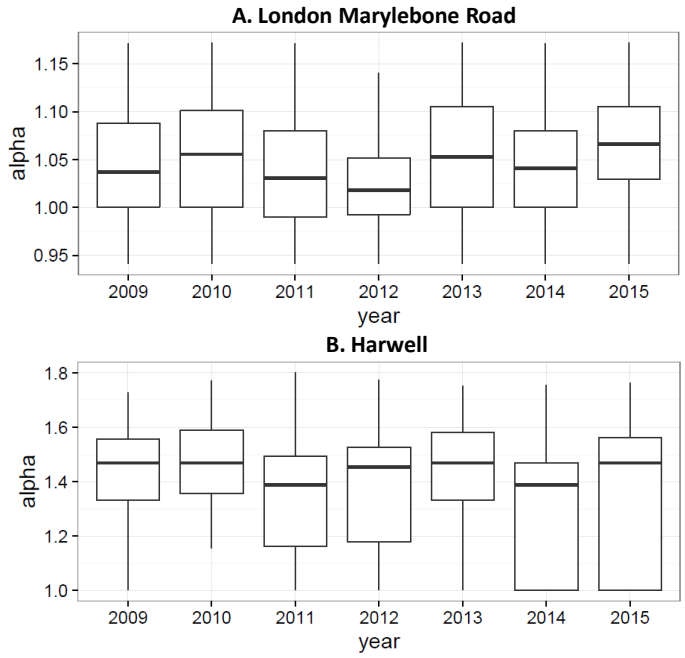

Figure 1. Distribution of hourly Ångstrom coefficients each year calculated from London Marylebone Road (all year) and Harwell (only winter months).

In contrast with Marylebone Road, the Ångström coefficients at the rural site of Harwell during the winter months were ~ 1.3-1.5 indicating a mixture of both biomass and traffic aerosol sources (Figure 1B).Table 2 summarizes the distribution of winter Ångstrom coefficients for each individual site. The 95th percentiles values were close to α~2 at rural sites: Detling (α=1.96), Goonhilly (α=1.91), Harwell and Auchencorth Moss (α=1.89), indicating episodes dominated by wood smoke aerosol were present. The suburban sites of Folkstone and Norwich also observed Ångstrom coefficients near to 2 (α=1.86) during the winter months.

Table 2. Median of annual median, 5th and 95th percentiles of winter Ångstrom coefficients for the rural and urban background sites of the Defra's black smoke network across the sampling years.

Site	*Years*	*Median*	*5th percentile*	*95th percentile*
Auchencorth Moss	2012-2015	1.40	1.00	1.89
Bath	2009-2012	1.22	0.97	1.74
Cardiff	2009-2015	1.32	1.00	1.75
Dudley Central	2009-2012	1.25	1.00	1.63
Edinburgh St Leonards	2009-2012	1.21	1.00	1.51
Goonhilly	2012-2013	1.51	0.76	1.91
Glasgow Centre	2009-2012	1.15	1.00	1.40
Glasgow Townhead	2013-2015	1.17	1.00	1.80
Woolwich	2009-2012	1.37	1.00	1.80
Harwell	2009-2015	1.47	1.00	1.89
Halifax	2009-2012	1.27	1.00	1.67
Birmingham Tyburn	2009-2015	1.21	1.00	1.55
South Kirkby	2009-2012	1.23	0.99	1.66
Manchester Piccadilly	2009-2012	1.20	1.00	1.43
London North Kensington	2011-2015	1.26	1.00	1.61
Nottingham Centre	2009-2012	1.25	1.00	1.54
Norwich Centre	2009-2013	1.42	1.00	1.86
Sunderland	2009-2012	1.33	1.00	1.80
Stoke-on-Trent Centre	2009-2012	1.25	1.00	1.60
Folkestone - Cheriton	2009-2012	1.45	1.00	1.86
Maidstone - Detling	2012-2015	1.45	1.00	1.96

3.2 Winter C_{wood} concentrations in the UK

Winter concentrations (December, January and February, DJF) from 2009/10 to 2015/16 are shown in Figure 2. Concentrations ranged from 0.23 µg m^{-3} (measured in Goonhilly in DJF 2012/13) to 2.69 µg m^{-3} (measured in Cardiff in DJF 2010/11). Higher concentrations were generally measured in DJF 2010/11; more noticeably in Wales (Cardiff measured C_{wood} mean winter concentration of 2.69 µg m^{-3}) and in the Midlands (with an average concentration ~ 1.8 µg m^{-3}). A general decreased was observed across the network in the subsequent winters. The highest winter concentration in DJF 2015/16 was 1.1 µg m^{-3} measured in London North Kensington. However caution is asked when interpreting the results in DJF 2015/16 as only provisional data was available when writing this report.

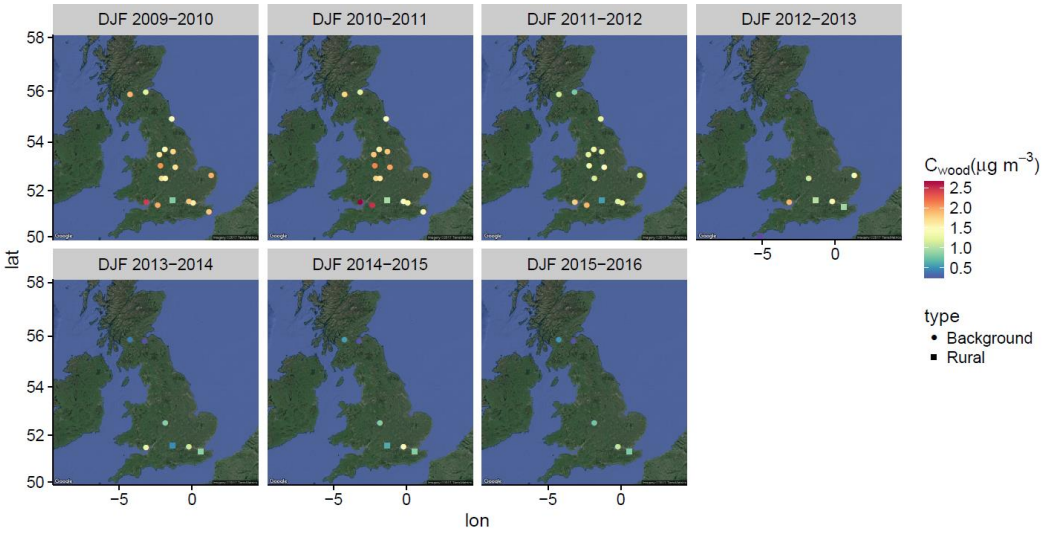

Figure 2. Winter wood burning aerosol mass concentration measured in rural and urban background sites in the UK.

3.3 Time dynamics of C_{wood} concentrations in the UK

The higher concentrations in C_{wood} prevail in winter months, from November to February, with average concentrations of ~1.5-2.0 µg m^{-3}. Lower concentrations were observed in summer (May to August) with average concentrations <0.5 µg m^{-3} (Figure 3).

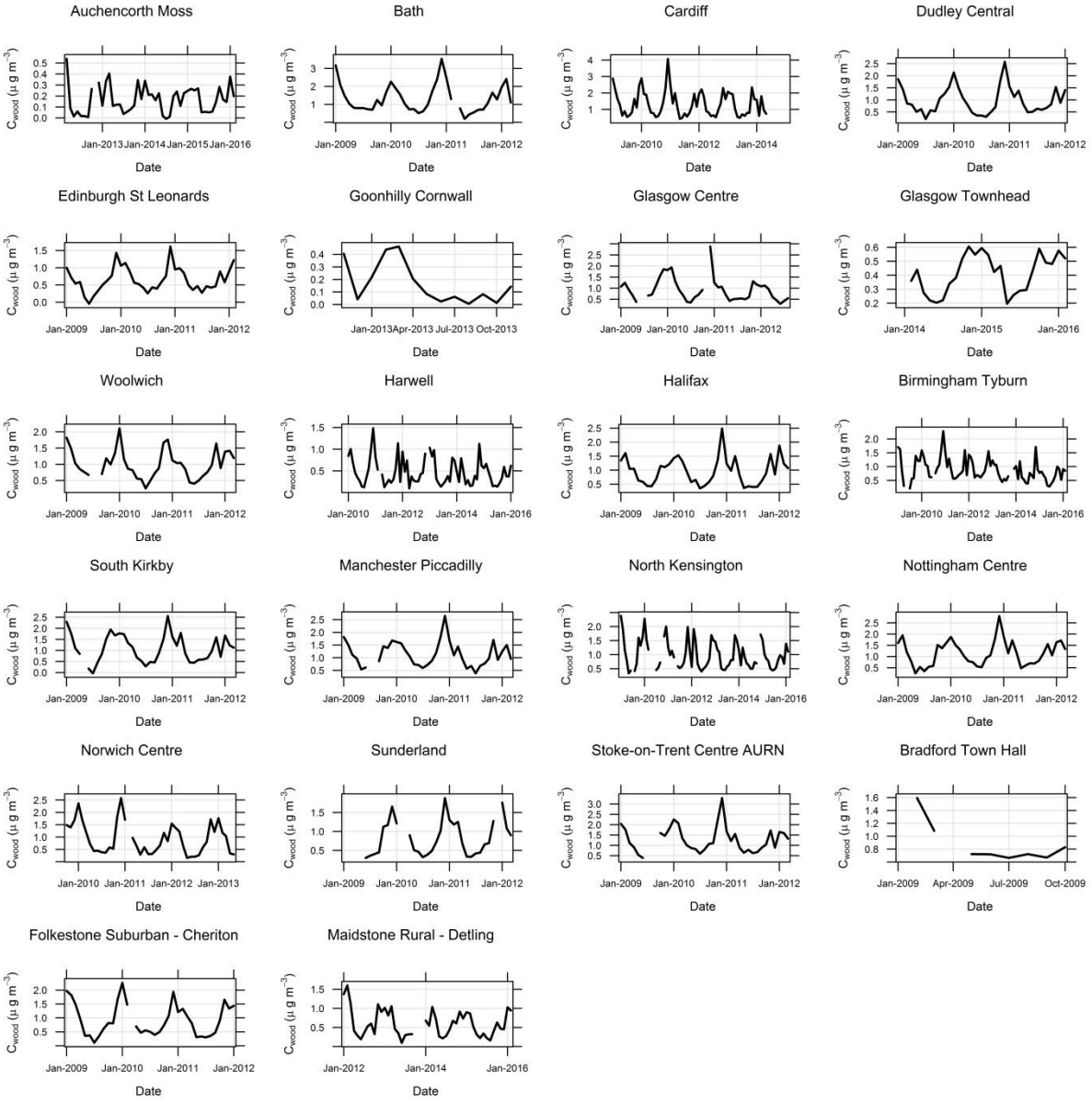

Figure 3. Monthly mean C_{wood} concentrations for the rural, suburban and urban background sites.

The temporal dynamics, i.e. mean hourly variation, mean hourly variation per day of the week and monthly variation, in C_{wood} concentrations at three urban locations from a north to south gradient are shown: Glasgow Centre (Figure 4), Manchester Piccadilly (Figure 5) and London North Kensington (Figure 6); and at a rural site (Harwell; Figure 7. Despite all three sites showing a similar seasonal variation, the diurnal and weekly patterns differed between sites. The mean C_{wood} concentration dipped to ~0.5 µg m^{-3} from 1 to 5 am at both Glasgow Centre and Manchester Piccadilly to increase later attaining values ~1 µg m^{-3} that remained constant for most of day. A slight enhancement between 8 pm and midnight was observed at Manchester Piccadilly.

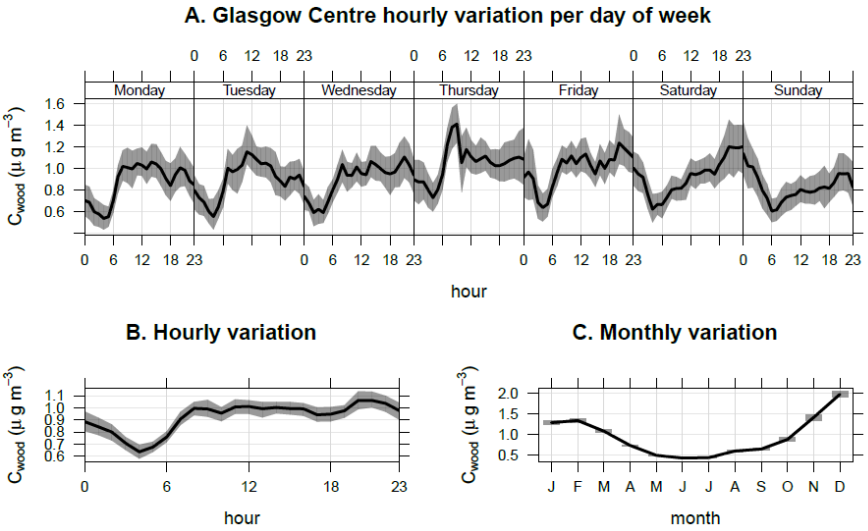

Figure 4. Mean hourly variation per day of the week (A), mean hourly variation (B) and mean monthly variation (C) of C_{wood} calculated for Glasgow Centre.

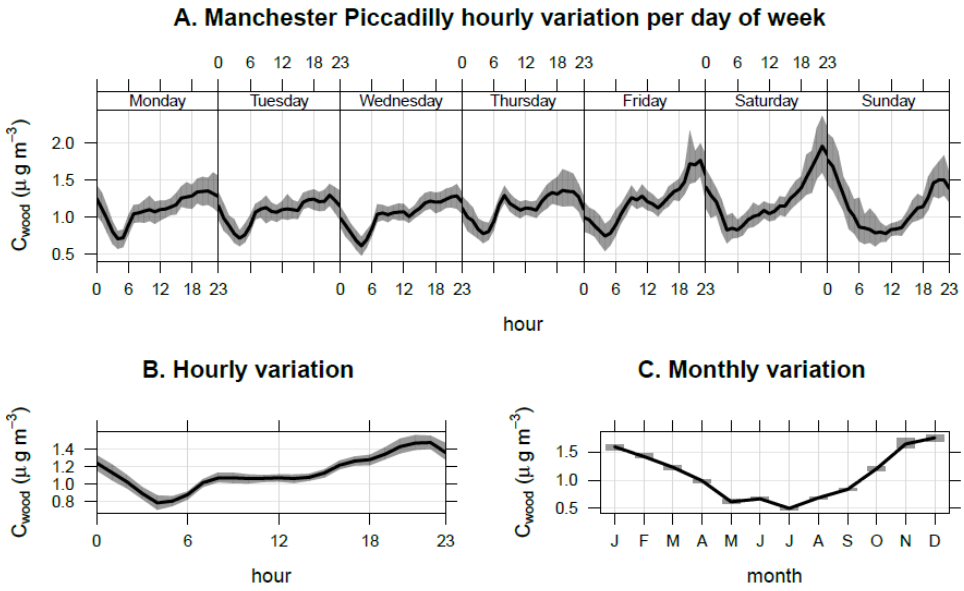

Figure 5. Same as Figure 4 for Manchester Piccadilly.

London North Kensington observed low and declining mean concentrations in the early hours of the day (~1 µg m^{-3}) as peak concentrations dissipated from the previous night. This was followed by a peak between 6 and 7 am mainly during the weekdays. Concentrations dipped during the central hours of the day (0.6-0.8 µg m^{-3}) followed by an enhancement of C_{wood} concentrations from 8 pm to midnight with mean concentrations 1.4-1.5 µg m^{-3} during weekdays and up to 2 µg m^{-3} at the weekend.

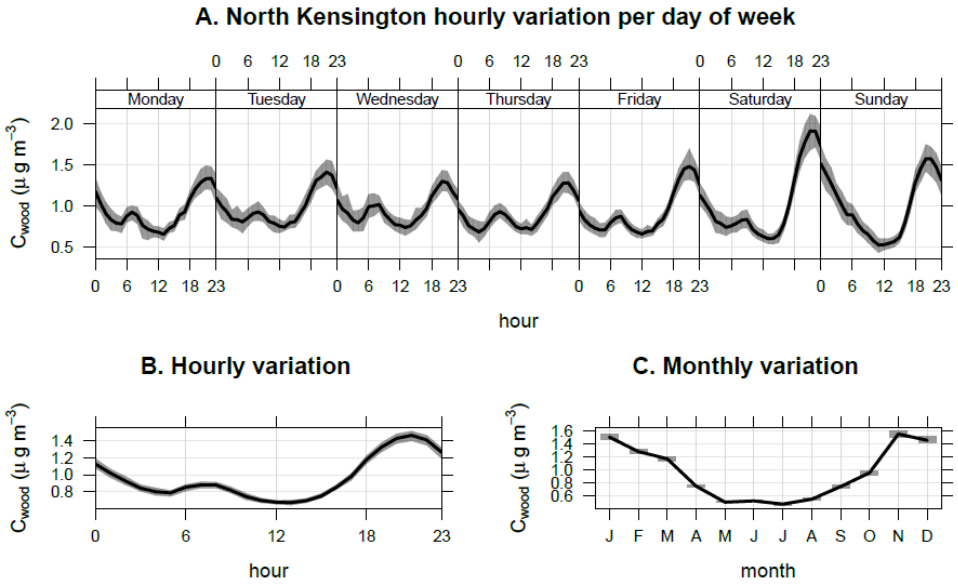

Figure 6. Same as Figure 4 for London North Kensington.

The weekly pattern for C_{wood} at Harwell (a rural site in south England) showed a similar pattern to that at London North Kensington, although with a difference in magnitude, almost 1 µg m^{-3} lower at Harwell compared to North Kensington. C_{wood} was enhanced during the evenings (6 pm to midnight) and especially at the weekend (Figure 7).

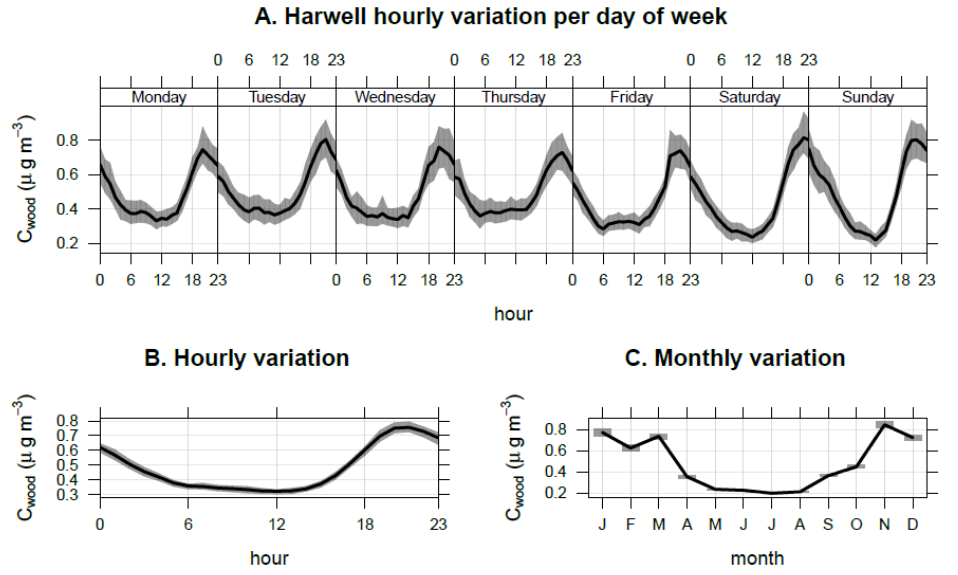

Figure 7. Same as Figure 4 for Harwell.

The weekend/weekday ratios can be used as a metric to explore if wood burning was used as a main source of heating or used as a recreational activity. Only evening hours were considered (6-11 pm) to calculate the ratio. The average ratio in the UK was 1.16 (median of 1.09); ranging from 0.56 (Detling, DJF 2015/16) to 1.90 (Norwich Centre; DJF 2011/12). The distribution of the weekend/weekday ratios per each winter is shown in Figure 8. There was a generalized weekend/weekday ratio > 1 during winter 2011/12 with some locations in the south-east with ratios

>1.5. Overall, locations in the south showed ratios > 1 (with the exception of DJF 2015/16) while in the Midlands and Scotland ratios close to 1.

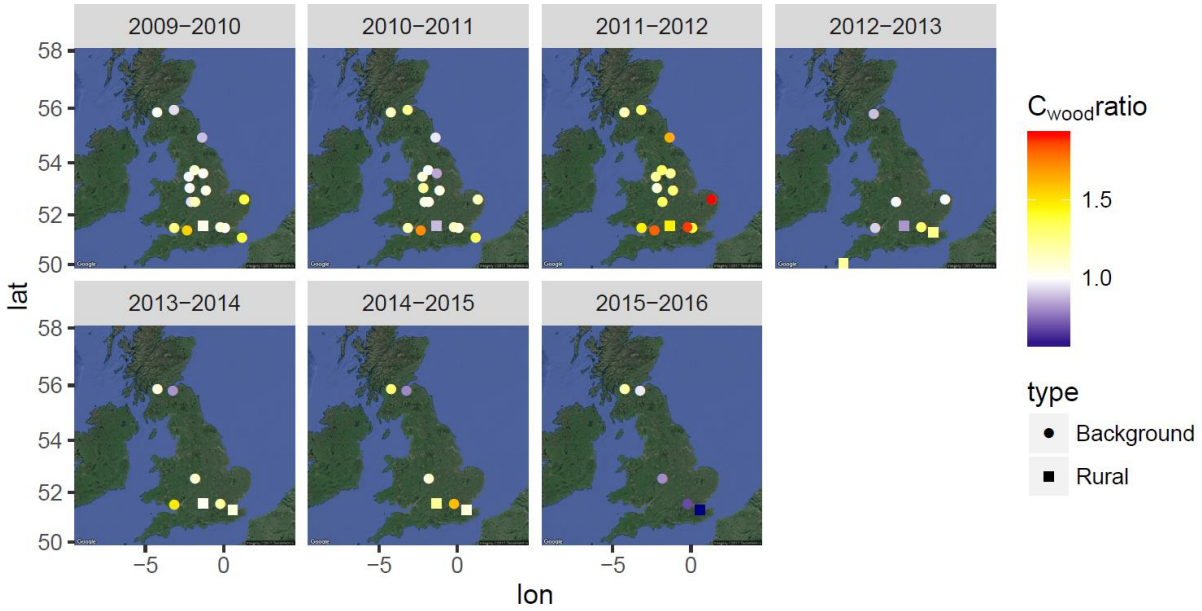

Figure 8. Distribution of the weekend/weekday evening winter ratios across the network. Background sites indicate either urban background or suburban sites.

3.4 Percentage of C_{wood} in PM

There was a large spatial variability in C_{wood}/PM_{10} across the network, in part due to spatial gradients in $PM_{2.5}$ (Air Quality Expert Group 2012). The largest C_{wood}/PM_{10} ratios were observed in DJF 2009/10 in Cardiff and Stroke-on-Trent with ratios >10%. Cardiff measured ratios ~8-9% in the following winters. The lowest ratios (3-4%) were measured in the rural sites at Harwell and Auchencorth Moss (Figure 9).

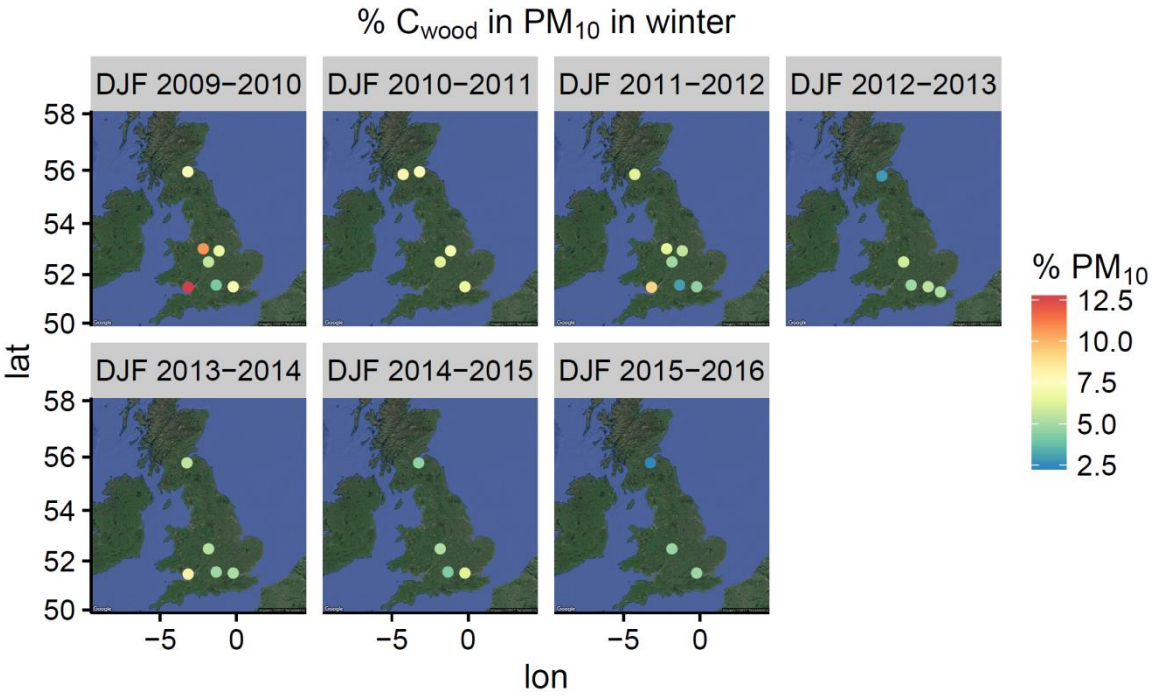

Figure 9. C_{wood} /PM_{10} ratios in winter for sites with a data capture greater than 75%.

On annual basis, C_{wood} in PM_{10} ranged between 2 and 4% in rural sites; and between 4% to 6% in urban sites (Figure 10, Table 3).

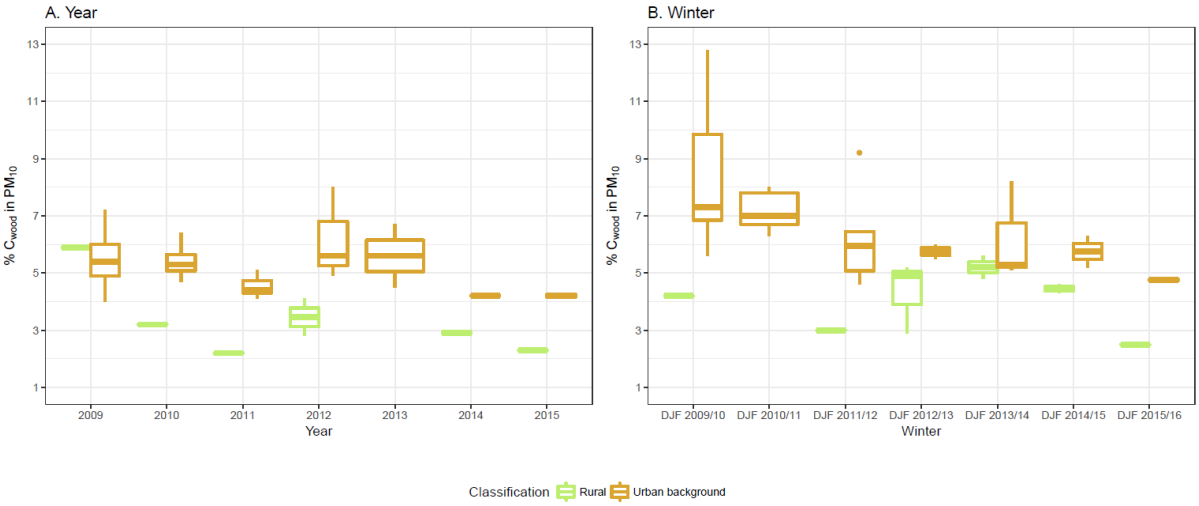

Figure 10. C_{wood} / PM_{10} ratios observed across the network split by type of site. Only sites with data capture greater than 75% were considered.

A decrease in the ratios of C_{wood} in PM_{10} in winter was observed through the study years (Figure 10B). While the first two winters in the time series C_{wood} represented ~ 7% of PM_{10} (calculated as median across the network), it decreased to 5% in the following ones. It should be noted that the number of monitoring sites also changed along the study period. This might have an impact on this calculation and also in the reduction of variability observed in the boxplots.

Table 3. Annual mean C_{wood}/PM_{10} for type of site (in µg µg^{-1}).

Type of site	*Year*	*C_{wood}/ PM_{10}*	*N sites*
Rural	2010	3%	1
Rural	2011	2%	1
Rural	2012	4%	2
Rural	2014	3%	1
Rural	2015	2%	1
Urban background/Suburban	2009	6%	5
Urban background/Suburban	2010	5%	4
Urban background/Suburban	2011	5%	6
Urban background/Suburban	2012	6%	3
Urban background/Suburban	2013	6%	2
Urban background/Suburban	2014	4%	1
Urban background/Suburban	2015	4%	2

Cardiff was the location where C_{wood} represented the largest of $PM_{2.5}$ (17%-13%), followed by Nottingham and Glasgow Centre (12-13%). As with PM_{10} this in part reflects the spatial distribution of $PM_{2.5}$ in the UK. The lowest $C_{wood}/PM_{2.5}$ ratios were measured at rural sites: Harwell and Auchencorth Moss (Figure 11).

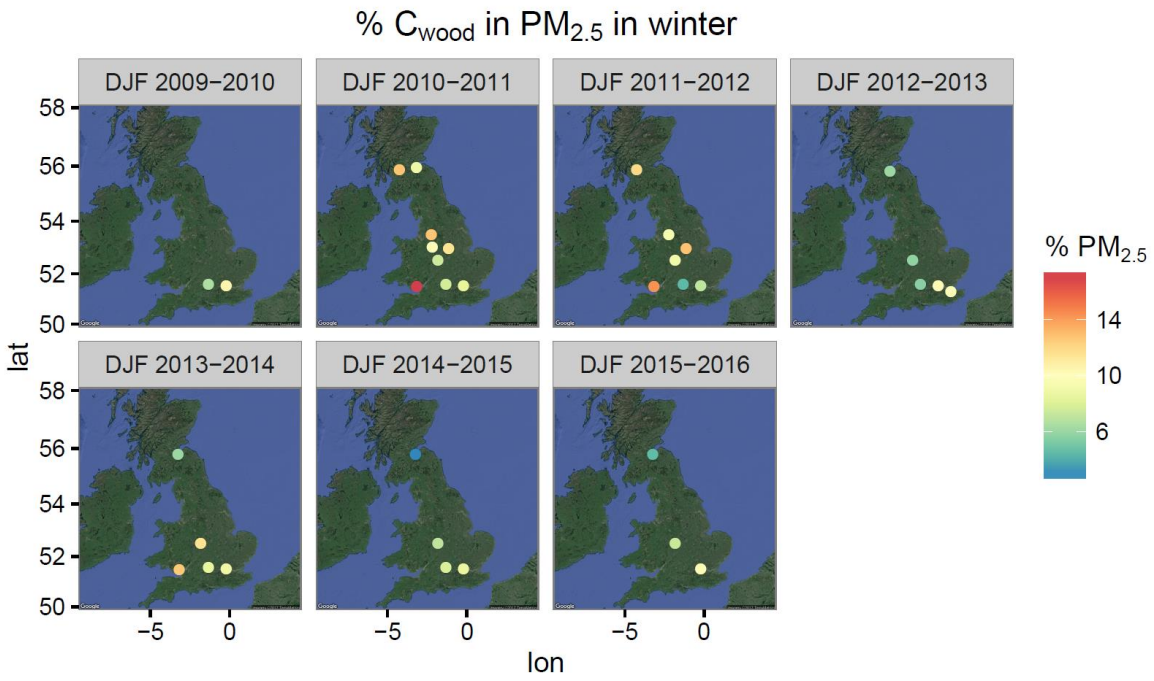

Figure 11. C_{wood} /$PM_{2.5}$ ratios in winter for sites with a data capture greater than 75%.

On annual basis C_{wood} in $PM_{2.5}$ ranged between 4 to 6% in rural sites; and between 6% to 9% in urban sites (Figure 12; Table 4). Winter C_{wood} in $PM_{2.5}$ showed a decreased ratios at the end of the time series, with medians ~10% in DJF2009/10 and DJF2011/12; to ratios ~8% in the last two winters (Figure 12B) largely caused by a decrease at rural sites.

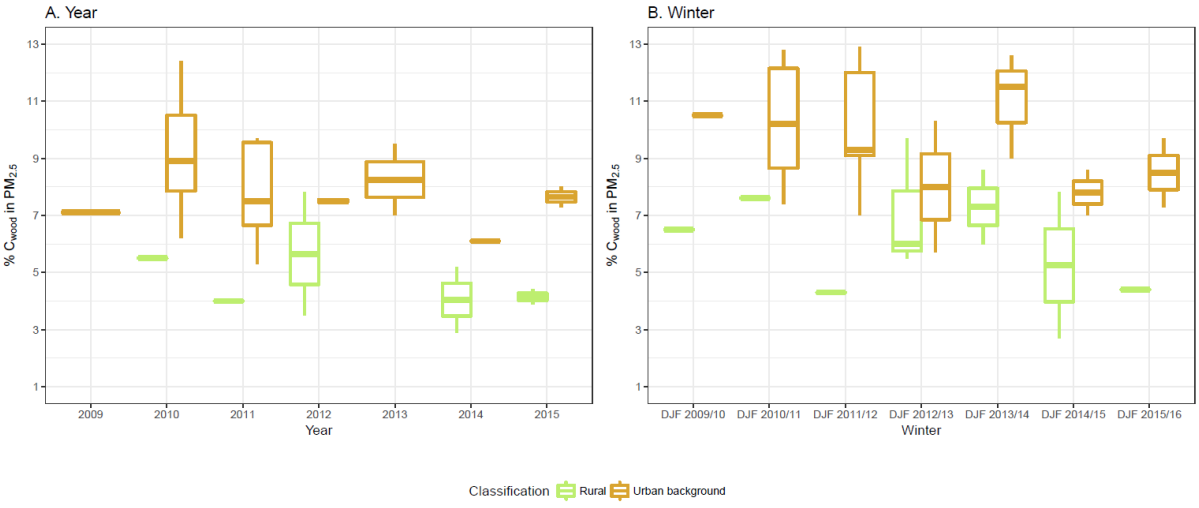

Figure 12. C_{wood} / $PM_{2.5}$ ratios observed across the network split by type of site. Only sites with data capture greater than 75% were considered.

Table 4. Annual mean $C_{wood}/PM_{2.5}$ for type of site (in µg µg^{-1}).

Type of site	Year	C_{wood}/ $PM_{2.5}$	*N* sites
Rural	2010	6%	1
Rural	2011	4%	1
Rural	2012	6%	2
Rural	2014	4%	2
Rural	2015	4%	2
Urban background/Suburban	2009	7%	1
Urban background/Suburban	2010	9%	7
Urban background/Suburban	2011	8%	7
Urban background/Suburban	2012	8%	2
Urban background/Suburban	2013	8%	2
Urban background/Suburban	2014	6%	1
Urban background/Suburban	2015	8%	2

Table 5 summarizes the contribution from wood smoke to winter daily PM_{10} concentrations for the days when the EU limit value was exceeded (daily PM_{10} > 50 µg m^{-3}). Wood burning ranged between 3 % (Harwell) to 10% (Cardiff) of PM_{10} during exceedance days. The contribution of C_{wood} to stratified by concentrations of daily winter PM_{10} is shown in the Supplementary Material for individual sites.

Table 5. Percentage of the contribution from C_{wood} to daily PM_{10} concentration for those days when daily PM_{10} > 50 µg m^{-3} during winter.

Site	C_{wood} / PM_{10} (%)	*N days*
Edinburgh St Leonard's	--	0
Folkestone, Kent Network	--	0
Maidstone, Detling	--	0
Auchencorth Moss	--	0
Manchester Piccadilly	6	1
Harwell	3	4
Cardiff	10	5
Stoke Centre	8	7
Birmingham Tyburn	5	13
North Kensington	7	14
Nottingham Centre	6	18
Glasgow Centre	4	22

The contribution of C_{wood} to urban derived $PM_{2.5}$ was calculated for those sites with rural observations within100 km. The rural sites were used to estimate a regional background that could then be subtracted from the urban measurements to focus on the additional $PM_{2.5}$ arising within the urban area. These pairs were: London North Kensington (urban) and Harwell (rural); London North Kensington and Detling; Birmingham Tyburn (urban) and Harwell (rural); Cardiff (urban) and Harwell (rural); Glasgow Townhead (urban) and Auchencorth Moss (rural). However, some of the site combinations resulted in very small urban PM increments (less than 1 µg m^{-3}), very close to the expected precision of FDMS-TEOM measurements, inducing a high degree of uncertainty into the calculation. This was the case for Glasgow and Cardiff. The results for London and Birmingham are shown in Table 6. Depending on calculation method and time period, it was estimated C_{wood} was between 23 and 31% of the urban derived $PM_{2.5}$ in London and Birmingham.

Table 6. C_{wood} contribution to urban $PM_{2.5}$ increments.

Urban site	Rural site	Time period	C_{wood}/urban PM(%)	C_{wood}/urban PM(%)
London North Kensington	Harwell	Dec'09–Dec'15	23	26
London North Kensington	Detling	Jan'12–Feb'13	25	31
Birmingham Tyburn	Harwell	Jan'10–Dec'15	25	29

3.5 C_{wood} / B[a]P and C_{wood} / NO_X ratios

In order to provide information for the National Atmospheric Emission Inventory (NAEI) verification and modelling to evaluate the spatial and temporal prediction of primary C_{wood} concentrations, the C_{wood}/B[a]P and C_{wood}/NO_X ratios were calculated for those sites with collocated measurements. Only sites with 75% data capture were reported.

C_{wood}/B[a]P ratios ranged from 0.39 to 0.11 (median value = 0.18). Auchencorth Moss observed a decrease of the ratio with time, from 0.39 to 0.11 from 2012 to 2015. Such decrease in the ratios was not observed anywhere else in the network. Birmingham Tyburn showed a median ratio of 0.23, similar to the one observed in Glasgow Centre in 2011 (0.20). The rural site at Harwell observed the lowest ratio, a median of 0.15.

Table 7. Annual C_{wood} / B[a]P ratios (in µg µg^{-1}) calculated from monthly means. Only sites with 75% data capture are reported.

Site	*Year*	*C_{wood} / B[a]P*	*N months*
Auchencorth Moss	2012	0.39	10
Auchencorth Moss	2013	0.18	12
Auchencorth Moss	2014	0.16	12
Auchencorth Moss	2015	0.11	12
Birmingham Tyburn	2009	0.29	12
Birmingham Tyburn	2010	0.23	12
Birmingham Tyburn	2011	0.19	12
Birmingham Tyburn	2012	0.24	12
Birmingham Tyburn	2013	0.22	12
Birmingham Tyburn	2014	0.23	12
Glasgow Centre	2011	0.20	12
Harwell	2010	0.15	12
Harwell	2011	0.18	12
Harwell	2013	0.15	12
Harwell	2014	0.12	12
Harwell	2015	0.15	12

C_{wood}/NO_X ratios in rural areas ranged from 0.039 to 0.060 µg µg^{-1}, depending on the year. In urban areas, the annual mean ratios were lower, ranging from 0.017 to 0.025 µg µg^{-1}, indicating a much richer NO_X environment than rural areas. Ratios per each individual site per year can be found in Supplementary Material (SM4).

Table 8. Annual mean C_{wood}/NO_X for type of site (in µg µg^{-1}).

Type of site	*Year*	*C_{wood}/ NO_X*	*N sites*
Rural	2009	0.039	1
Rural	2010	0.049	1
Rural	2011	0.038	1
Rural	2012	0.040	2
Rural	2013	0.060	2
Rural	2014	0.041	1
Rural	2015	0.040	1
Urban background/Suburban	2009	0.019	8
Urban background/Suburban	2010	0.021	8
Urban background/Suburban	2011	0.020	8
Urban background/Suburban	2012	0.025	3
Urban background/Suburban	2013	0.024	3
Urban background/Suburban	2014	0.017	2
Urban background/Suburban	2015	0.017	2

3.6 Correlations with temperature and wind speed

To explore possible factors determining C_{wood} concentrations, correlations between C_{wood} and daily temperature and C_{wood} and wind speed were explored. To inspect the shape of relation between variables the median concentration of C_{wood} was calculated for each 0.5°C in temperature and for each 1 m s^{-1} in wind speed (Figure 13). C_{wood} and temperature showed a negative linear relation with less C_{wood} concentration as temperature rises. Correlation coefficients ranged between 0.22 and 0.94 (Table 9). C_{wood} and wind speed showed a quadratic relation with C_{wood} decreasing as wind speed increases. Correlation coefficients of the quadratic relation ranged 0.40-0.99.

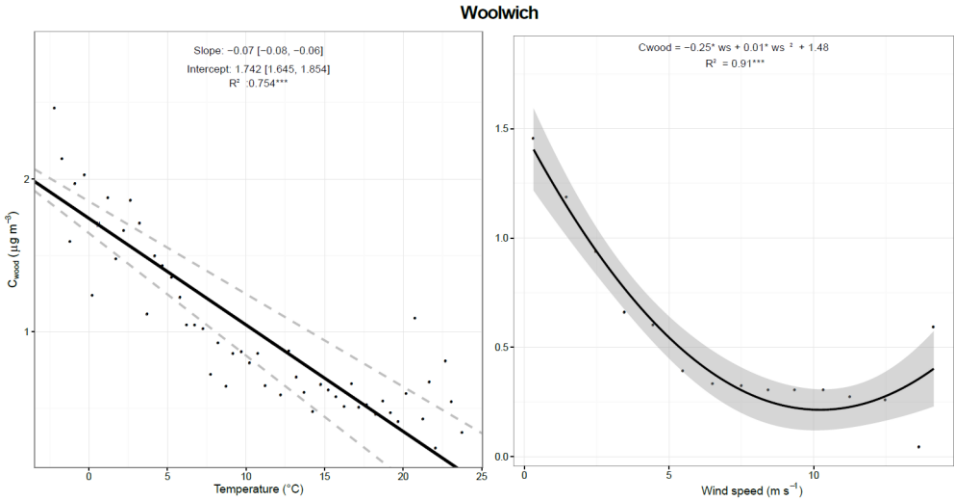

Figure 13. Correlation of daily C_{wood} vs daily temperature grouped by 0.5°C; and C_{wood} against wind speed at Woolwich site.

Table 9. Correlation coefficient (R^2) from correlating daily wood smoke mass concentrations against daily air temperature.

	Temperature **Daily values, grouped by 0.5°C**	**Wind speed** **Hourly values, grouped by 1 m s^{-1}**
All	0.22 - 0.79	0.40-0.95
Winter	0.25 - 0.94	0.49-0.99
Winter weekends	0.35 - 0.87	0.48-0.99

Using all available data the daily C_{wood} concentrations showed a moderate correlation with daily mean temperatures (R^2 = 0.09 - 0.43). A spatial gradient (lower-higher) was observed north-south in the country. Better correlation coefficients were observed when considering only winter months (R^2 = 0.12 - 0.57) and winter weekends (R^2 = 0.12 - 0.76). The highest correlation was observed at the Goonhilly rural site (R^2 = 0.76). However, only data for one winter was available at this site (DJF 2012/13).

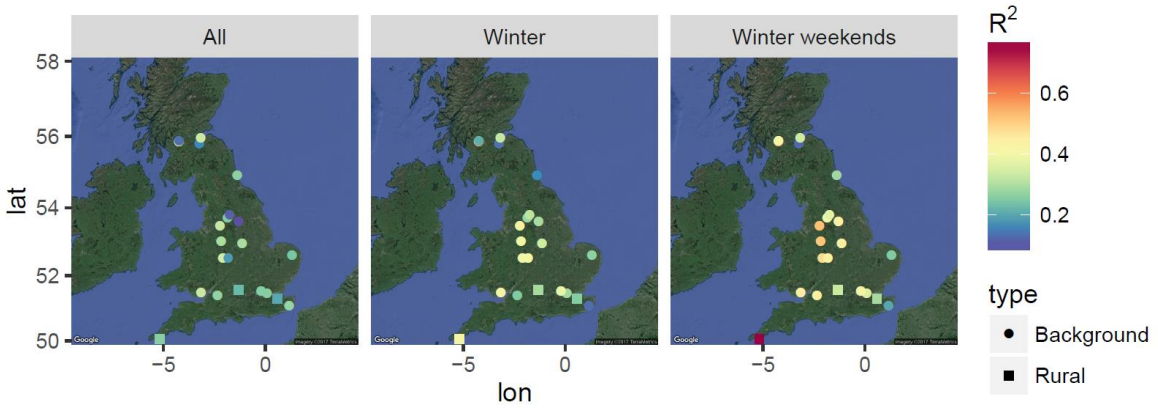

Figure 14. Distribution of correlation coefficients between daily C_{wood} and daily temperature; all year-round, winter days and winter weekends data considered.

Different temperature metrics were correlated against daily C_{wood} concentrations: daily maximum and minimum temperature; night maximum and minimum temperature (defined from 6 pm to midnight). The range of R^2 values are shown in Table 10; those (R^2: 0.03 -0.74) are within the range of values obtained comparing C_{wood} concentrations with mean daily concentrations (R^2: 0.12 -0.76).

Table 10. Correlation coefficient (R^2) from correlating daily wood smoke mass concentrations against maximum and minimum daily air temperature.

	Maximum daily temperature	Minimum daily temperature	Maximum night temperature	Minimum night temperature
All	0.07-0.27	0.12-0.44	0.07-0.74	0.07-0.31
Winter	0.06-0.35	0.06-0.65	0.08-0.29	0.03-0.73
Winter weekends	0.03-0.73	0.15-0.34	0.07-0.37	0.11-0.74

The correlation between C_{wood} and wind speed at hourly basis was very weak: R^2 = 0 – 0.31 (all data); R^2 = 0.05 – 0.34 (winter); and R^2 = 0.05-0.43 (winter weekends). The lowest correlation coefficients were observed in the south-eastern part of the UK whereas the highest were observed in the Midlands, Glasgow and London (R^2~0.3-0.4) (Figure 15).

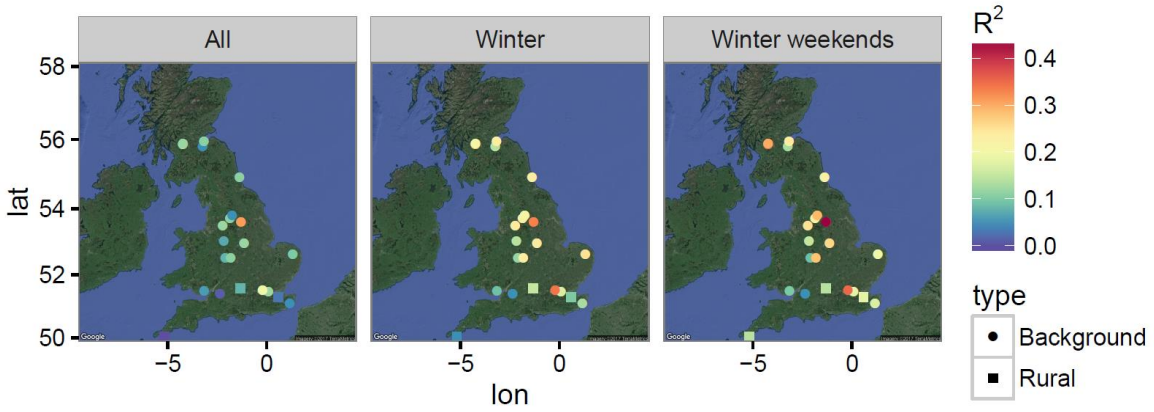

Figure 15. Distribution of correlation coefficients between hourly C_{wood} and hourly wind speed; all year-round, winter days and winter weekends data considered.

3.7 Trends in winter C_{wood}

Sixteen of the 21 sites could be used to calculate trends in the period 2009 – 2011.The majority of the monitoring sites observed a flat trend in C_{wood} when using the raw time series; and when looking at the network, the overall trend was slightly negative, at a rate of -0.07 (-0.12, -0.02) µg m^{-3} $year^{-1}$. When applying the deseason algorithm, five of the 16 sites observed statistically significant downward trend; one site observed a positive upward trend. Looking at the whole of the network, there was an overall downward trend in C_{wood} at a rate of -0.04 (-0.08, 0) µg m^{-3} $year^{-1}$, statistically significant (p<0.1) (Figure 16).

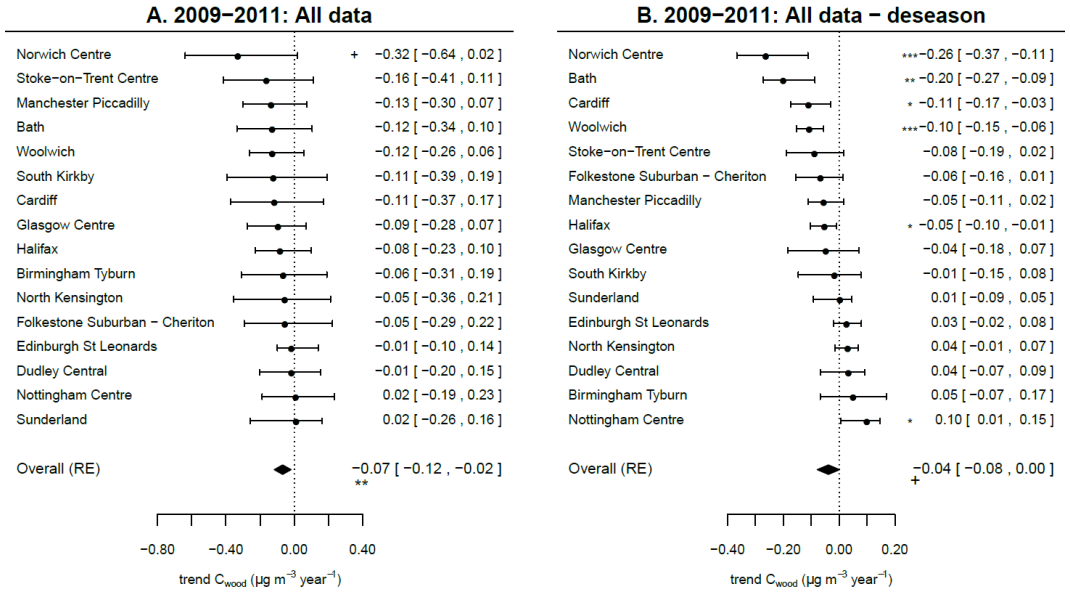

Figure 16. Forest plots for the trends in C_{wood} between January 2009 and December 2011. A. All data; B. All data – deseasonalized time series. * significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.**

Trends in C_{wood} using winter concentrations for the period 2009-2011 also showed no significant trends at the individual sites with the exception of South Kirby; but a significant downward trend network-wide at a rate of −0.21 (−0.25, −0.16) µg m^{-3} $winter^{-1}$. For the same period of time, winter temperature and winter wind speed increased at a rate of 1.30 (1.16, 1.44) ºC $winter^{-1}$ and 0.40 (0.27, 0.52) m s^{-1} $winter^{-1}$, respectively (Figure 17). Trends in C_{wood} * wind speed also showed a negative trend when looking at the overall trend for the whole network: −0.39 (−0.59, −0.19) µg m^{-2} $winter^{-1}$.

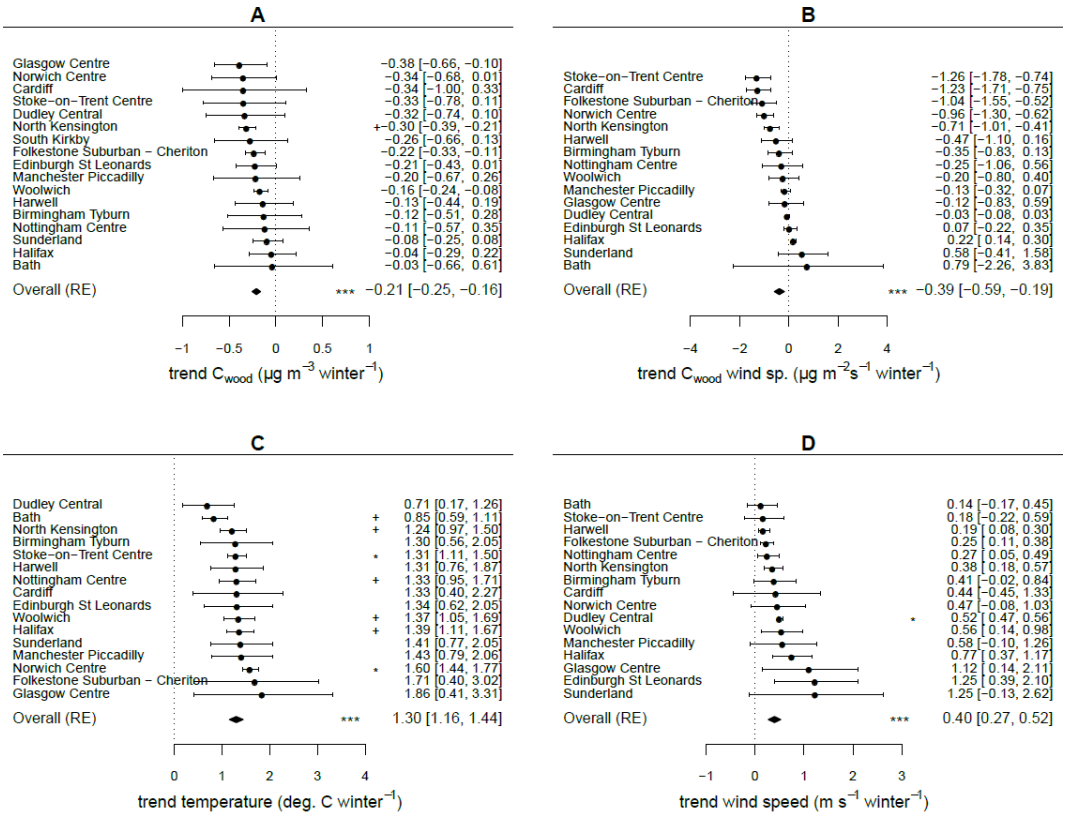

Figure 17. Forest plots for the trends in winter C_{wood} (A), C_{wood} * wind speed (B), winter temperature (C) and winter wind speed (D) between January 2009 and February 2012. * significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.**

Only four monitoring sites fulfilled the data capture threshold for the calculation of the trends in the whole period between 2009 and 2015 (Figure 18). The overall trend without deseasonalizing the time series was -0.04 (-0.08, 0.00) µg m^{-3} $year^{-1}$, statistically significant (p<0.01) although individual site trends were not (Figure 18). Deseasonalizing the time series resulted in statistically significant trends in three of the four available sites. The overall trend was -0.03 (-0.05, -0.01) µg m^{-3} $year^{-1}$, also statistically significant at the p<0.01 level.

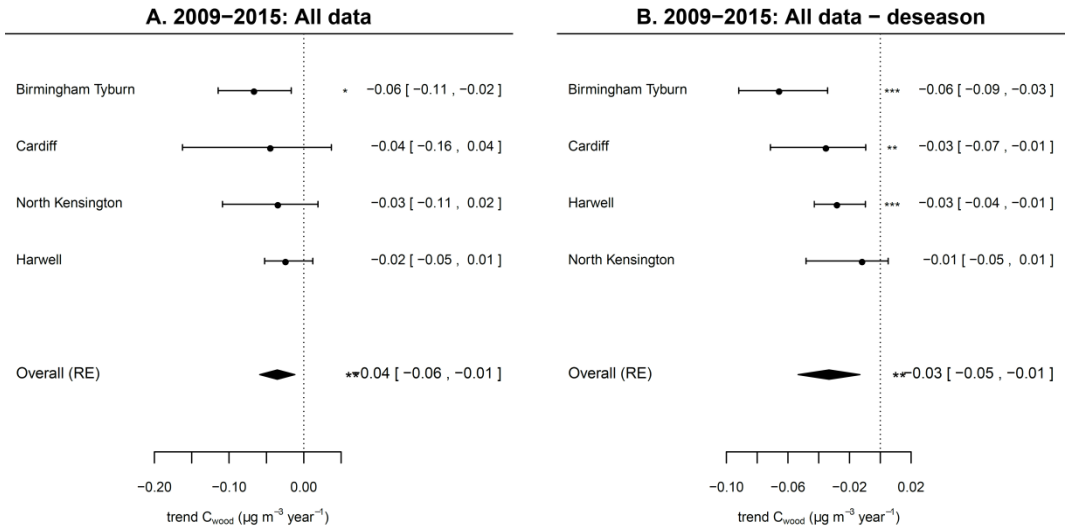

Figure 18. Forest plots for the trends in C_{wood} between January 2009 and January 2016. A. All data; B. All data – deseasonalized time series. * significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.**

Trends in winter C_{wood} in the period DJF 2009/11 to DJF 2015/16 showed also a negative trend of -0.14 (-0.23, -0.05) µg m^{-3} $winter^{-1}$, significant at $p<0.01$. Winters were also milder, with temperatures increasing at a rate of 0.63 (0.42, 0.82) ºC $winter^{-1}$; and windier, with wind speed increasing at a rate of 0.19 (0.12, 0.26) m s^{-1} $winter^{-1}$.

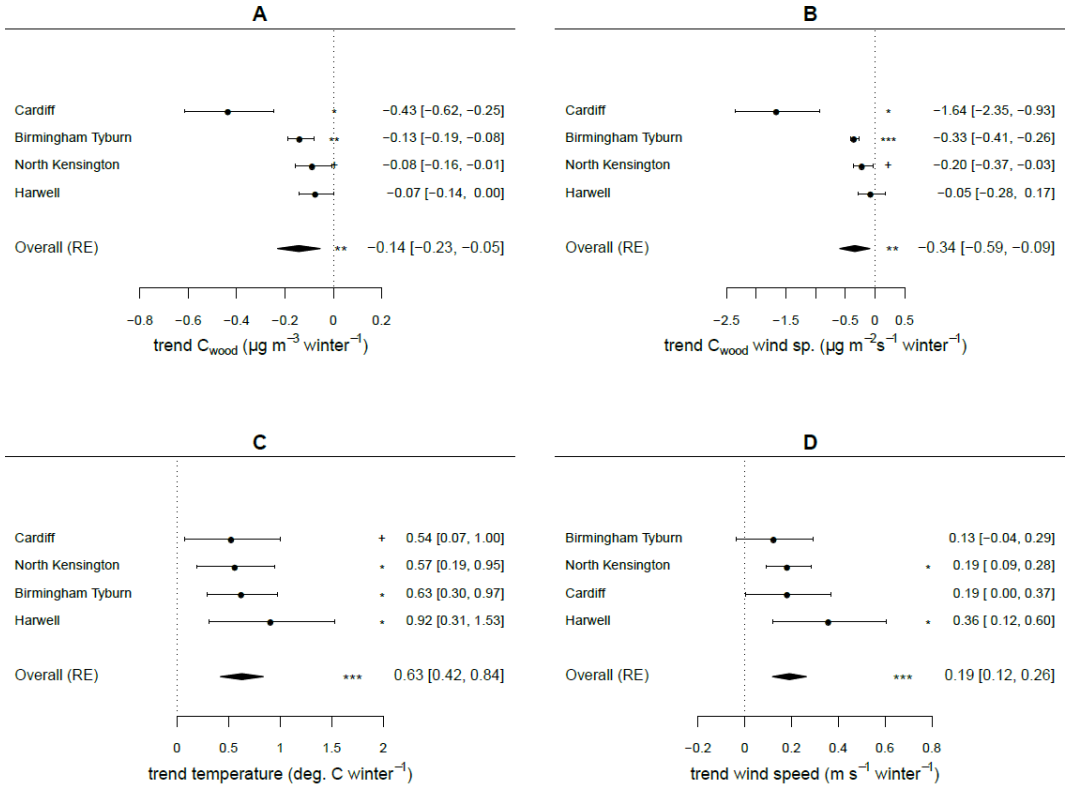

Figure 19. Forest plots for the trends in winter C_{wood} (A), C_{wood} * wind speed (B), winter temperature (C) and winter wind speed (D) between January 2009 and February 2016. * significant at the 0.001 level; ** significant at the 0.01 level; * significant at the 0.05 level; + significant at the 0.1 level; (blank) not statistically significant. Overall (RE) refers to the mean trend for all sites.**

The summary of the trends in C_{wood} using the different approaches is presented in Table 10. The overall trends calculated for the two periods of time were quite similar, with or without deseasonalizing. The downward trends ranged between -0.07 to -0.03 µg m^{-3} $year^{-1}$. The rate of decrease was faster when computing trends using only the winter concentrations; however, no significant differences were observed for trends between 2009-2012 (with an overall trend of -0.21 µg m^{-3} $winter^{-1}$) and 2009-2016 (-0.14 µg m^{-3} $year^{-1}$).

Table 11. Summary of the trends in C_{wood} according the different approaches.

	2009 – 2011	2009 – 2015
C_{wood} all data (µg m^{-3} $year^{-1}$)	−0.07 (−0.12 , −0.02)**	−0.04 (−0.06 , −0.01)**
C_{wood} all data – deseasonalized (µg m^{-3} $year^{-1}$)	−0.04 (−0.08 , 0.00)$^{+}$	−0.03 (−0.05 , −0.01)**
C_{wood} winter (µg m^{-3} $winter^{-1}$)*	−0.21 (−0.25, −0.16)***	−0.14 (−0.23, −0.05)**
C_{wood} x wind speed winter (µg m^{-3} $winter^{-1}$)*	−0.39 (−0.59, −0.19)***	−0.34 (−0.59, −0.09)**

* winter trends comprises the periods DJF 2009/10- DJF 2011/12 and DJF 2009/10- DJF 2015/16

4. Discussion

4.1 Proxies to calculate wood burning mass concentrations

Currently there is no reference method for C_{wood} quantification. Although there are several chemicals used as tracers for wood burning (i.e. levoglucosan, fine potassium, etc.) and the optical characteristics of carbonaceous material from biomass burning differ from those from diesel soot, the contribution of biomass burning emissions to air quality is difficult to quantify precisely. All the methods have their own uncertainties and interferences that make difficult the precise quantification of aerosol from wood burning emissions. The tracer methods such as levoglucosan and fine potassium require a conversion factor to estimate the quantity of biomass aerosol. This is assumed to be constant but it is known that it depends on combustion conditions and wood composition (Schmidl et al. 2008). These two methods are also constrained by their low time resolution (daily) and their sampling and analysis cost, meaning that most of the available measurements are campaign based.

Aerosol mass spectrometers (AMS) are able to determine organic aerosols from wood burning. To do this the organic aerosol fragments detected by the instrument are clustered using statistical techniques such as Positive Matrix Factorisation (PMF) which seeks sources that vary together in time and identifies sources, for instance wood burning, using key tracer species (such as fragments from levoglucosan). This approach can provide solutions that vary from site to site dependent on the local pollutant mixture and it can also struggle to resolve sources that change together over time or are subject to similar meteorology. It is therefore subject to both statistical and analytical uncertainties that can affect specificity and quantification. The application of PMF has limitations when source emissions have a strong temporal correlation, or when meteorology has a strong impact on PM variability. In these cases, the resulted factors are mixed source profiles and consequently misrepresenting the real-world sources (Gianini et al., 2012). Difficulties are also found in identifying sources that make a small contribution to PM mass concentrations (Ulbrich et al., 2009; Crippa et al., 2014).

The quantification of the wood aerosol mass concentrations by the aethalometer method is constrained by different factors. The first is that only one source of solid fuel should be present and preferentially absorbing light in the UV channel and thereby increasing the Ånsgstrom coefficient from unity. That was the main reason why the methodology was not been applied to sites in Northern Ireland given the

extensive use of coal as a domestic heating because many locations are off the natural gas grid. On the basis of PAH speciation by Brown and Brown (2012), coal burning is thought to make a negligible contribution to mainland UK PM in contrast to Northern Ireland, where impacts are clear. Another variable that affects the quantification of wood burning through the aethalometer method is the dependence on the $\alpha_{traffic}$ and α_{wood} values. In this study we used $\alpha_{traffic} = 0.96$ and $\alpha_{wood} = 2$. There is evidence in the literature that α values from wood burning from individual fires can vary over a quite large range (e.g. Lewis et al., 2008; Harrison et al., 2013). Harrison et al. (2013) estimated the α values using a 7-wavelength aethalometer when burning different types of wood (*Fagus sylvatica*, *Populus nigra* and *Quercus pyrenaica*) in a traditional cast iron stove and found α ranging from <1 to 3, 2-3 and 1.5-2.5, respectively, over the span of three hours. However this variation will be samller over a town or a city and the average α_{wood} of 2 has been used in many studies.

Sandradewi et al. (2008), Favez et al. (2010), Zotter et al. (2016), among other, have proved the reliability of the aethalometer model to quantify wood burning aerosol mass concentrations in alpine valleys and cities such as Grenoble and Zurich. Harrison et al. (2013) raised questions about the ability of the aethlometer to correctly track seasonal variability on wood burning in London. However, in our study the seasonal variation in wood burning was found to be more than twice that of traffic dominated black carbon suggesting a separation of traffic from wood burning aerosols. Several diurnal profiles (notably Dudley, Stoke, Glasgow and Edinburgh) showed fluctuations around the time of the morning traffic peak which could indicate interference from traffic black carbon in the method but this was not present in all cities. Fuller et al. (2014) showed that the two-wavelength aethalometer could be used to quantify wood burning mass concentrations in London given its good correlation to levoglucosan measurements.

The comparison between C_{wood} calculations for the three methods available in this study is presented in the Supplementary Material (SM1); a summary is shown in Table 12 . All the three methods compare well with R^2 values > 0.78 (p<0.01). Despite good correlation coefficients quantification of C_{wood} differed between methods. The aethalometer model and the levoglucosan method presented the best comparison. However, mean C_{wood} concentrations reported in Crilley et al. (2015) from levoglucosan measurements at London North Kensington during the winter Intensive Observational Period (IOP) for the ClearFlo project (12th January-10th February 2012) were 39% lower than C_{wood} from the aethalometer model for the same period of time.

Higher wood smoke concentrations were obtained from the 2 wavelength aethalometer compared to AMS-PMF by 70%. Similar values have been reported between the 7 wavelength aethalometer and AMS-PMF in Paris (Crippa et al., 2013). Favez et al. (2010) also found that the aethalometer model predicted higher concentrations (~30%) than AMS-PMF for 15 day campaign in Grenoble, in the French Alps.

Some authors have pointed that the aethalometer model can underestimate C_{wood} concentrations in relatively clean atmospheres. That it is due to the loss of semi-volatile light absorbing organics from the aethalometer filter tape during long-time sampling on a given "spot" (Favez et al. 2010). Therefore the quantification of wood burning mass aerosols in the rural areas might suffer from this effect.

Clear advantages reside in the use of the aethalometer model to calculate C_{wood}: low cost; reliability; and high-time resolution. Long-time series of absorbance data are then available allowing the characterization of temporal dynamics and long-term trends.

Table 12. Comparison of C_{wood} concentrations estimated from different methods.

Compared methods	R^2	*Comparison*	*Reference*
2wl AeM *vs* levoglucosan	0.84	13% lower	This study
2wl AeM *vs* levoglucosan	--	39% higher	This study *vs* Crilley et al. (2015)
2wl AeM *vs* AMS-PMF	0.77	70% higher	This study
7wl AeM *vs* AMS-PMF	0.85	30% higher	Favez et al. (2010)
7wl AeM *vs* AMS-PMF	0.73	68% lower	Crippa et al. (2013)
7wl AeM *vs* AMS-PMF	0.59	72% lower	Crippa et al. (2013)
2wl AeM *vs* 7wlA	0.82	41% lower	This study

4.2 Contribution of C_{wood} to PM pollution

Table 12 shows the C_{wood} contribution to PM across different cities in Europe. The values reported in the present study are in accordance but it has been quantified in other cities across Europe.

Table 13. Contribution of C_{wood} in PM in different studies undertaken in urban background locations in Europe.

City (country)	*Year*	*C_{wood}/PM_{10}*	*$C_{wood}/PM_{2.5}$*	*Method*	*Reference*
Copenhagen (Denmark)	1999-2004	15% (annual)		Daily filters, COPREM	Andersen et al., (2007)
Athens (Greece)	2002		15% (annual)	Daily filters, PMF 7 factor solution)	Karanasiou et al. (2009)
Zaragoza (Spain)	2003-2004	6% (annual)		Weekly filters, PMF	Callén et al. (2009)
Vienna (Austria)	2004	10% (winter)		Levoglucosan	Caseiro et al. (2009)
Milano (Italy)	2006	1% (summer) 14% (winter)		4 h samples, PMF (7 factor solution)	Bernardoni et al.(2013)
Milano (Italy)	2006-2009		8% (spring) 1% (summer) 30% (autumn) 25% (winter)	Daily filters, CMB	Perrone et al. (2013)
Marseille (France)	2008		0.5% (summer)	12-h filters, CMB	El Haddad et al. (2011)
Zurich (Switzerland)	2008-2009	13% (annual)		Daily filters, PMF (6 factor solution)	Gianini et al. (2013)
Barcelona (Spain)	2009		3 % (annual)	Daily filter, PMF (9 factor solution)	Reche et al. (2012)
Paris (France)	2009-2010		12% (annual)	Daily filter, PMF (7 factor solution)	Bressi et al. (2013)
Various locations (UK)	2009-2015	4-6 % (annual)	6-9% (annual)	Aethalometer	This study
Barcelona (Spain)	2010		5% (autumn)	Hourly composition, PMF (9 factor solution)	Dall'Osto et al. (2013)
London (UK)	2010	10% (winter)		Aethalometer	Fuller et al. (2014)
Flanders (Belgium) (6 cities)	2010-11	8.6–11.3% (winter) 4.8–6.3% (annual)		Levoglucosan	Maenhaut et al. (2012)
Barcelona (Spain)	2011		8% (winter)	Daily filter, PMF (4 factor solution)	Viana et al. (2013)
Leicester (UK)	2013-2015	5.1% (winter) 3.1% (year)		Levoglucosan	Cordell et al. (2016)
Lille (France)	2013	11.6% (winter)		Levoglucosan	Cordell et al. (2016)

4.3 Trends in wood burning aerosol in the UK

Some authors have pointed out probable increases in biomass burning derived PM_{10} pollution in years to come (Cordell et al. 2016). However, downward trends in C_{wood} concentrations were observed in almost all sites in the UK despite the low or absent statistical significance. Overall trends based on the network-wide tendency confirmed the slight downward trend. No significant changes were observed when different approaches were used to calculate trends, therefore results are statistically robust.

Winter downward C_{wood} trends were accompanied by upward trends in winter temperature (milder winters) and positive trends in wind speed (windier conditions). However, there was no a clear correlation between trends in C_{wood} and trends in temperature and wind speed (Figure 20Figure 20) despite the general tendency.

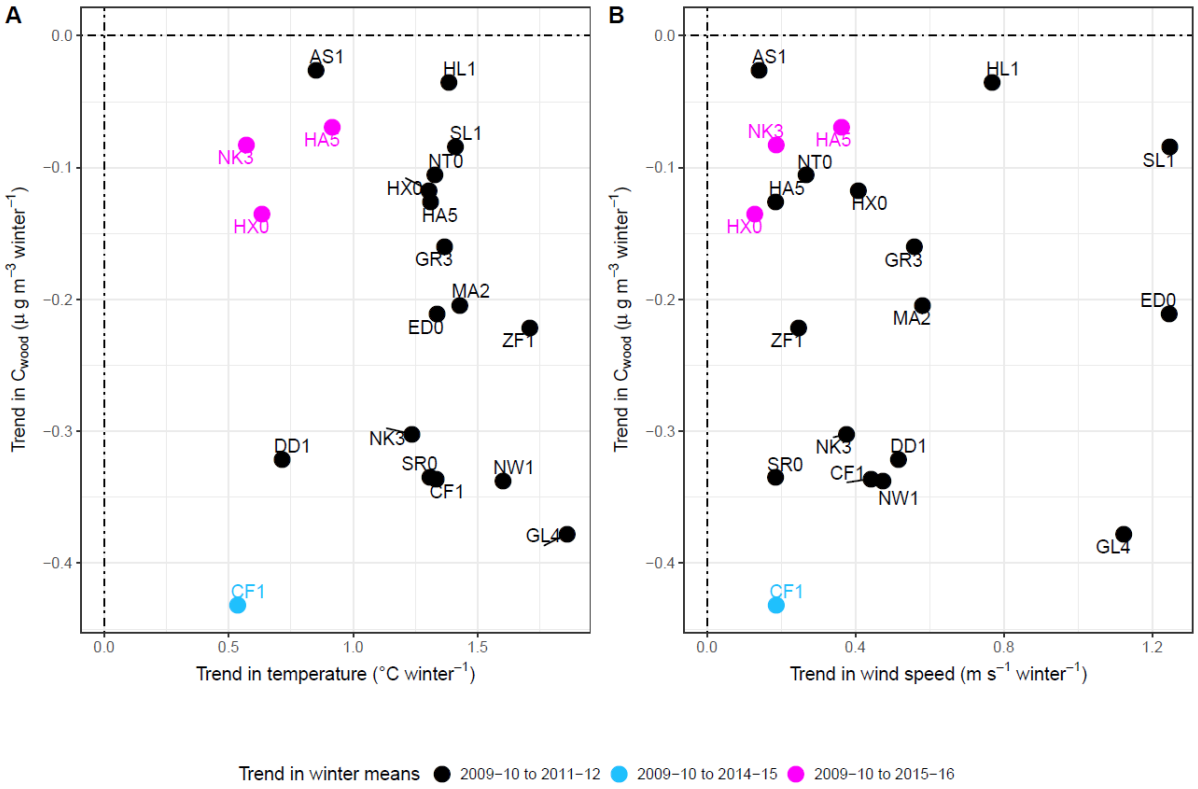

Figure 20. Trends in winter C_{wood} against trends in winter temperature (A) and against winter wind speed (B) coloured by the time span that trends were calculated for.

Downwards trend in C_{wood} were unexpected given the policy context incentivizing the use of biomass as source of domestic heating. Data from the Stove Industries Alliance shows an increase in the number of sales of wood stoves in the United Kingdom (Figure 21) (Milligan, personal communication) peaking in 2014 with an estimate 210,000 new appliances sold. However, the number of Defra exempt new appliances also increased between 2009 and 2015 from 5% of the new appliances in 2009 to 30% in 2015. Previous studies have highlighted the disparity of emissions from open fires to new residential woodstoves and boilers with higher combustion efficiencies. Emission tests undertaken in biomass combustion facilities in Portugal found that particle emissions from fireplaces were ~3, ~12 and ~15-fold higher than those from traditional woodstove, eco-labelled appliance and pellet stove, respectively (AIRUSE-ActionB4, 2015). Therefore, despite the increase in the use of wood burning, downward trends in C_{wood} might be also partly explained by the

improvement of the emission standards of new appliances and the replacement of old higher emission fire places with lower emission stoves.

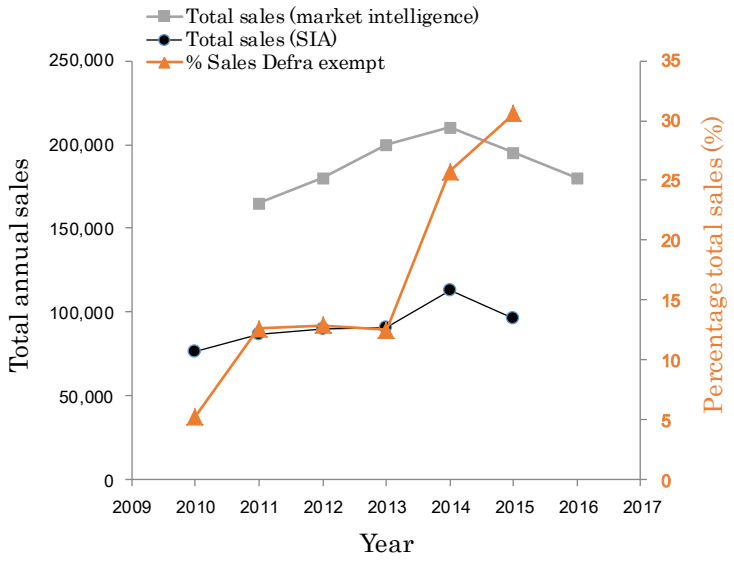

Figure 21. Total estimated wood stove sales, sales by the Stove Industry Alliance (SIA) and percentage of Defra Smoke Exempt Appliance in the United Kingdom5. Conclusions

5. Conclusions

This is the first systematic quantification of wood burning mass concentration across different cities in a given country over multiple years. Previous studies have characterized the wood burning concentrations based on short-time campaigns. Here, using the long-term data series from aethalometers from the Defra black carbon network we estimated PM from wood burning in 16 cities and 4 rural locations between 2009 and 2016.

As expected air pollution from wood burning was greatest in winter and almost absent in summer. Mean wintertime PM from wood burning varied between cities, ranging between 0.2 and 2.7 μg m^{-3}. In Cardiff, 13% of PM_{10} in winter 2009/10 came from wood burning. On annual basis wood burning in $PM_{2.5}$ ranged between 4 to 6% averaged across rural areas; and between 6 to 9% averaged across urban areas. It should be remembered that the majority of PM_{10} and $PM_{2.5}$ in urban and rural areas is not from primary emissions (e.g. Gelencsér et al., 2007). Instead the majority comes from reactions between other gaseous pollutants forming secondary particles. The proportion of wood burning in primary emissions will be greater than the percentages quoted here. It was estimated that wood burning was between 23 and 31% of the urban derived $PM_{2.5}$ in London and Birmingham, making control of wood burning an important urban issue. Similar proportions would be expected in other urban areas. On the whole the proportion of wood burning in PM_{10} did not increase on days when the EU limit value was breached. Instead wood burning is more important in determining mean concentrations. From a policy perspective the ambient impacts from wood burn will have greatest effects on the public health framework indicators, based on average $PM_{2.5}$, and on the $PM_{2.5}$ exposure reduction enshrined EU directives. It will also affect progress towards the attainment of WHO annual mean guidelines for these pollutants.

In most cities wood burning PM concentrations were greater in evenings, indicating residential combustion, and greater at weekends. Coupled with the poor correlation with daily temperature (R^2 = 0.12- 0.57) this suggests that current urban wood burning is in large part decorative and is not being used primary heating. Reis et al (2009) suggest that the timing and location of wood burning is a major factor in population exposure; put simply people burn wood in residential areas at the times of day when residential populations are greatest. Fuller et al. (2013) showed how indoor black carbon concentrations were affected by wood burning in neighbouring homes.

Between 2009 and 2015 trends in PM from wood burning were slightly downwards at -0.03 (-0.05, -0.01) µg m^{-3} $year^{-1}$. This was unexpected given anticipated growth in biomass combustion due to policy initiatives aimed at increasing production of heating and electricity from renewable and low-carbon energy sources. These policies include the Renewable Heat Incentive, Feed-in Tariffs and the so-called Merton Rule. UK Industry data also suggests stove sales are running at between 150,000 and 200,000 units per year with over one million stoves sold between in 2010 and 2015. A number of alternative methods were used to calculate the trends. These included tests to allow for changes in the severity of winters over the study period; which became warmer and windier at the end of the study. Although the different methods made slight changes to the magnitude of the calculated trend, the conclusion that trends were slightly downwards was found to be robust. One possible explanation is the replacement of high emission fireplaces with newer, and lower emission, wood stoves balancing an increase in total wood heating.

From an emissions perspective wood burning has an impact on requirements to reduce total UK primary emissions of PM under Göteborg protocol and the National Emissions Ceiling Directive. To support inventory verification this study produced ratios of wood burning PM to other ambient air pollutants.

There is no reference method for estimating PM from wood burning in ambient air. Several methods exist. They are generally well correlated in international studies and this was borne out in comparisons carried out in London. We can therefore have good confidence in attribution of wood burning to be residential, and to some extent recreational based on the daily and weekly variation in concentrations, correlations with temperature and also in concluding that wood burning is a winter source. Similarly we can be confident in the relative spatial distributions. It is possible that a change in the way wood is being burnt, due to increasing stove use, is affecting the type of particle emissions and therefore sensitivity of our technique to detect trend. Any interferences in the apportionment method from traffic black carbon, which has fallen substantially in recent years (Font and Fuller 2016), could also depress any trend. However, given the good correlations between methods in comparisons during 2010 and then again 2012 / 2013 such effect is likely to be small. Although our method gives results that were close to the mean of other methods (mean bias of – 7% from 8 comparisons) the spread between the concentration estimates is wide at +- 70%. This needs to be considered when comparing concentrations to emissions inventory estimates.

Looking forward, large increases in biomass burning are projected from energy scenarios over the next two decades (Williams et al. 2017). An increase in the coverage of the aethalometer network would enhance our ability to track the impact of these changes. A surveillance programme would also need to include long-term measurements with multiple methods at key locations.

New laboratory studies (Bruns et al., 2015, 2016 and references therein) are suggesting that secondary particles also form from wood smoke over time periods of four hours or less suggesting impacts near burning source areas. The methods used here to trace primary emissions might therefore underestimate the full contribution of wood burning to ambient air and developments in this emerging evidence base should be followed closely.

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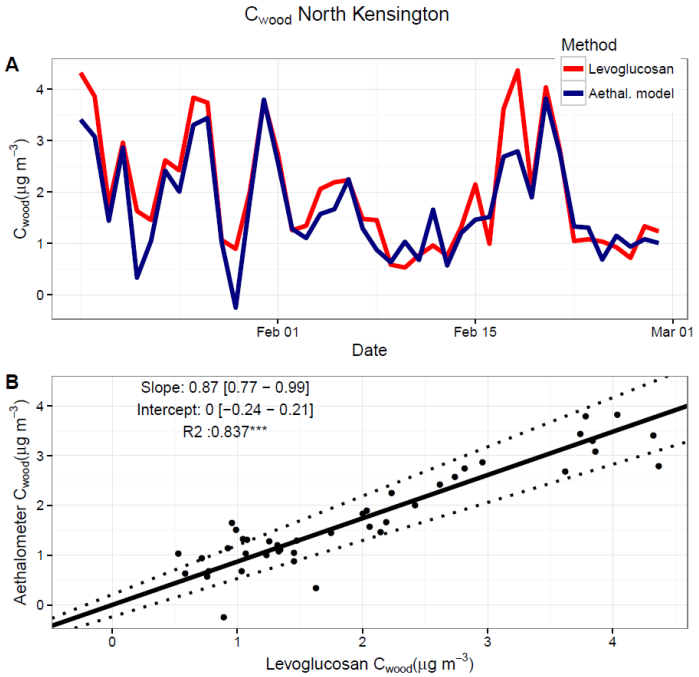
Supplementary Material

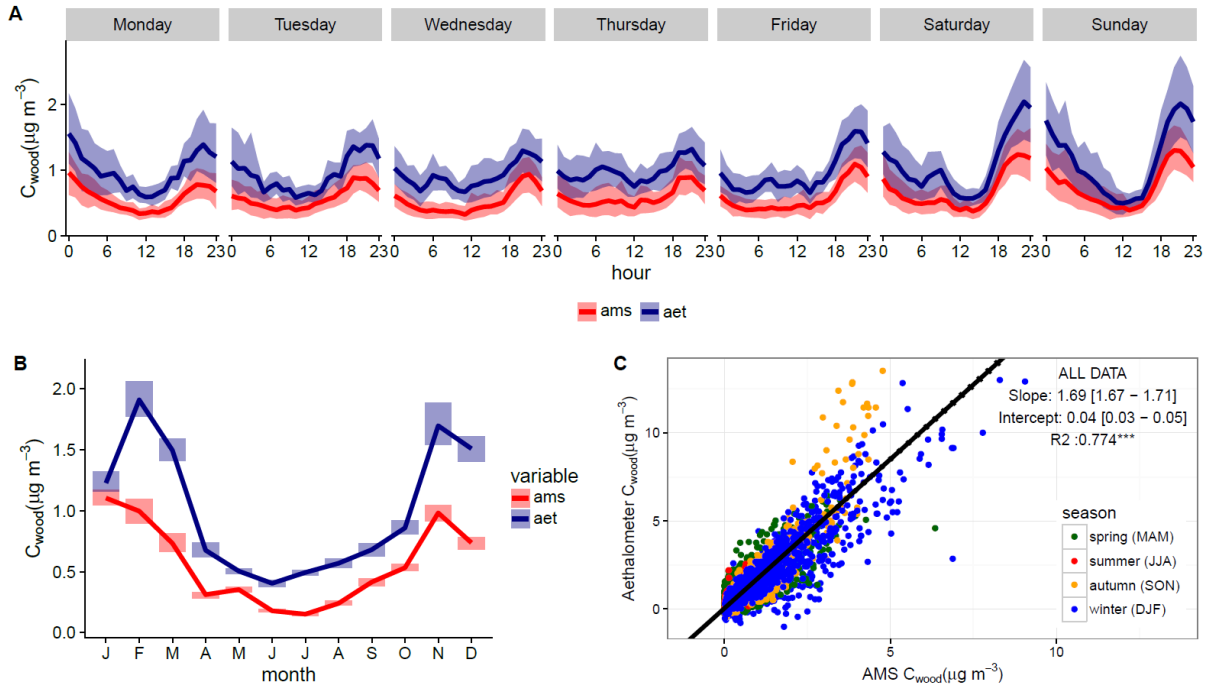
SM1. Wood burning: comparison between methods in London

C_{wood} data calculated from the aethalomter method used in this study was compared with three collocated techniques at London North Kensington. Wood burning derived from levoglucosan measurements and the aethalometer method agreed well in London from January to March 2010 (Fuller et al., 2014), showing the same day-to-day variability (R^2 = 0.84, p<0.01). However, C_{wood} derived from the aethalometer slightly underpredicted concentrations compared to levoglucosan measurements (slope of 0.87 as calculated from reduced-major-axis regression; Supplementary Figure 1).



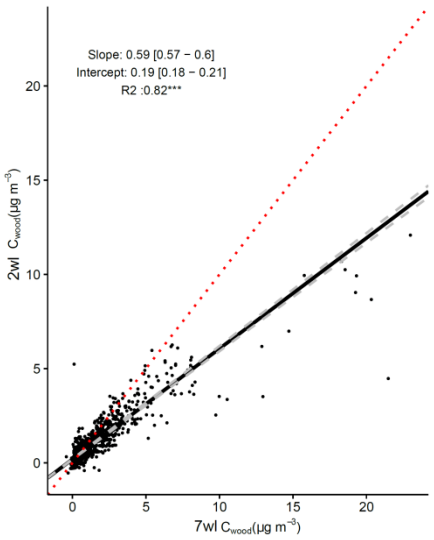
Supplementary Figure 1. Time series of wood burning derived from levoglucosan measurements and the aethalometer model at North Kensington between January and March 2010 (A) and comparison of concentrations from the two methods (B). Regression values were calculated with reduced-major axis regression.

C_{wood} concentrations from AMS-PMF between January 2012 and January 2013 and the aethalometer model followed the same temporal dynamics during the collocation at London North Kensington, with higher concentrations at nights and more noticeable during the evening hours at the weekends evenings (Supplementary Figure 2A, B) (R^2 = 0.77, p<0.01). However, the aethalometer method measured higher concentrations, 69% on average (58% higher for the winter months; R^2 = 0.79, p<0.01) (Supplementary Figure 2C). The overestimation of C_{wood} from the aethalometer method might be partly explained by the misrepresentation of biomass burning in the SFOA factor (Young et al., 2015).



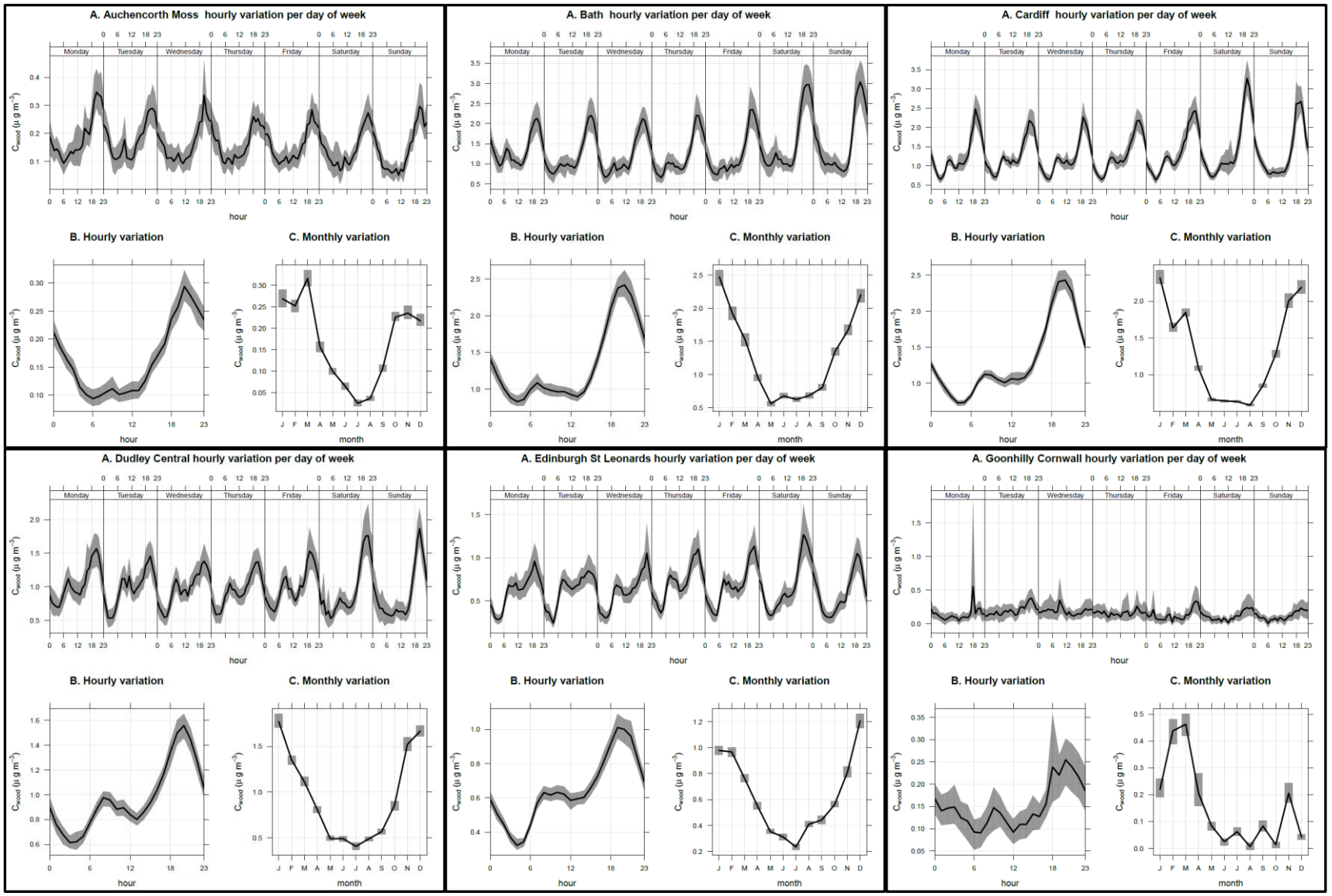
Supplementary Figure 2. Mean weekly (A) and mean hourly variations (B) for the wood burning concentrations as derived from the AMS-PMF and the aethalometer model at North Kensington; comparison of concentrations from the two methods. Regression values were calculated with reduced-major axis regression.

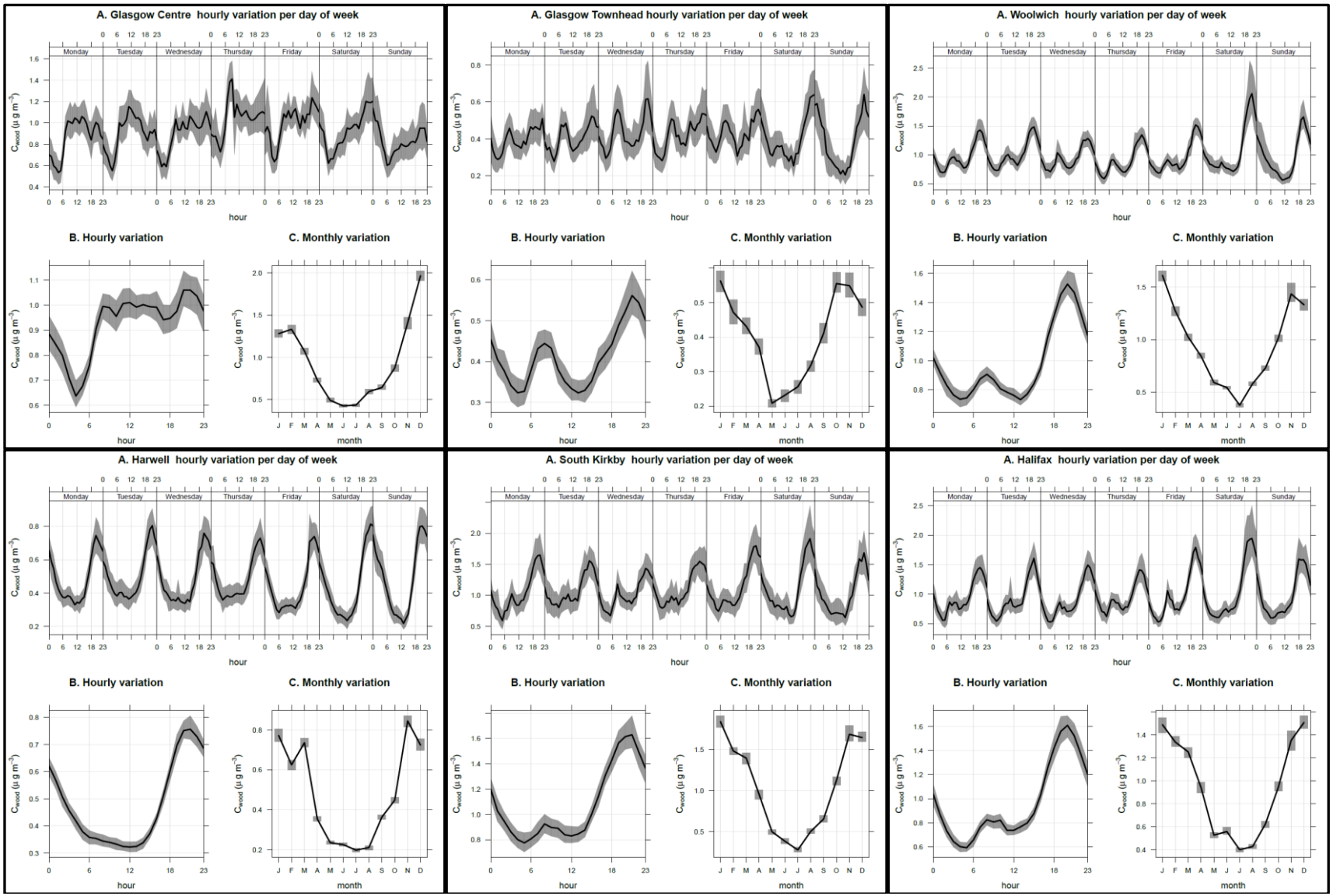
In winter 2015/16, an aethalometer A33 (7 wavelength aethalometer) was collocated at London North Kensington. C_{wood} was then derived using the same wavelengths as for the A22 aethalometer (370 and 880 nm). Lower wood burning mass concentrations (~40% lower) were derived from the 2 wavelength aethalometer compared with those calculated from the 7 wavelength one (R^2 = 0.82, p<0.01) (Supplementary Figure 3).

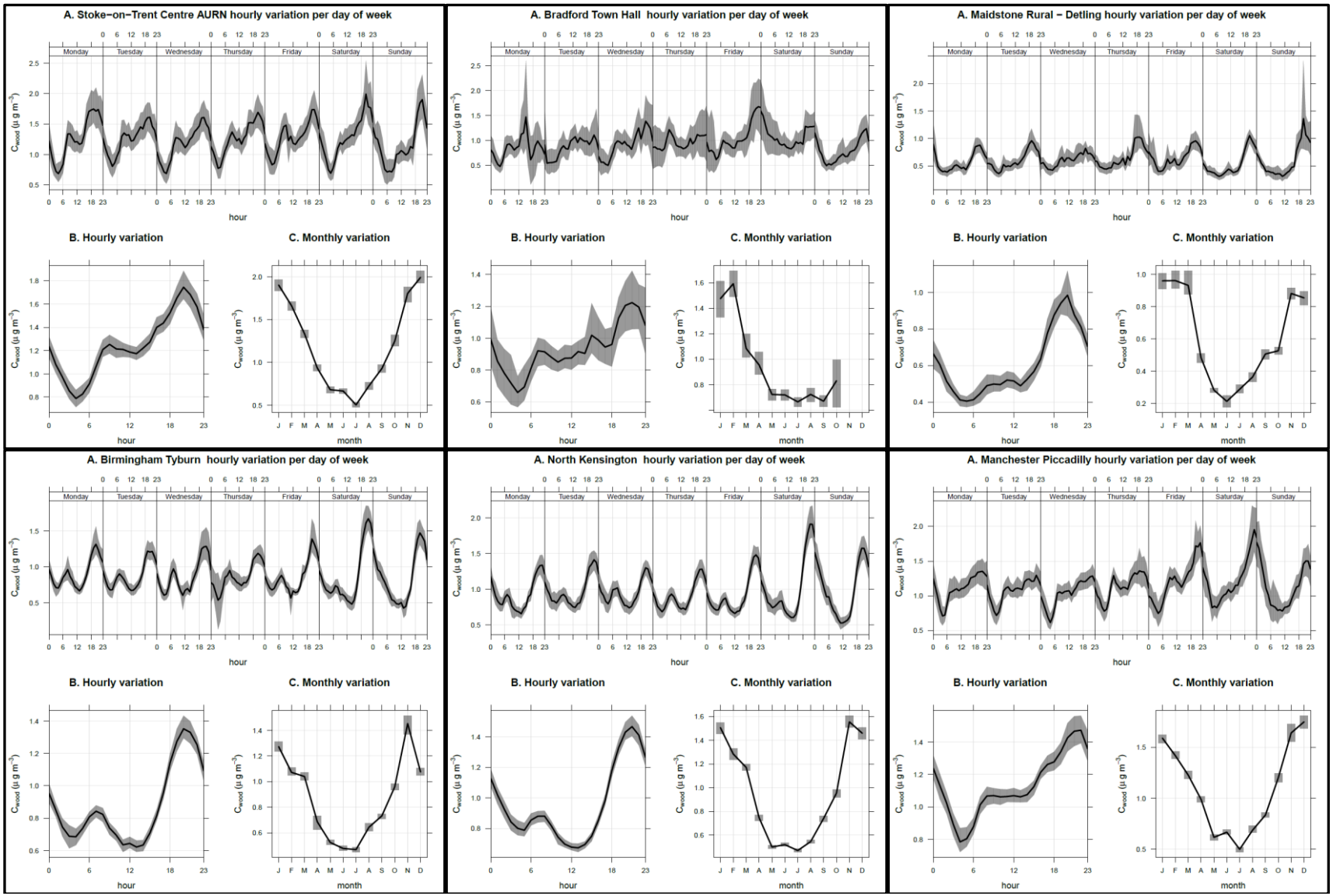


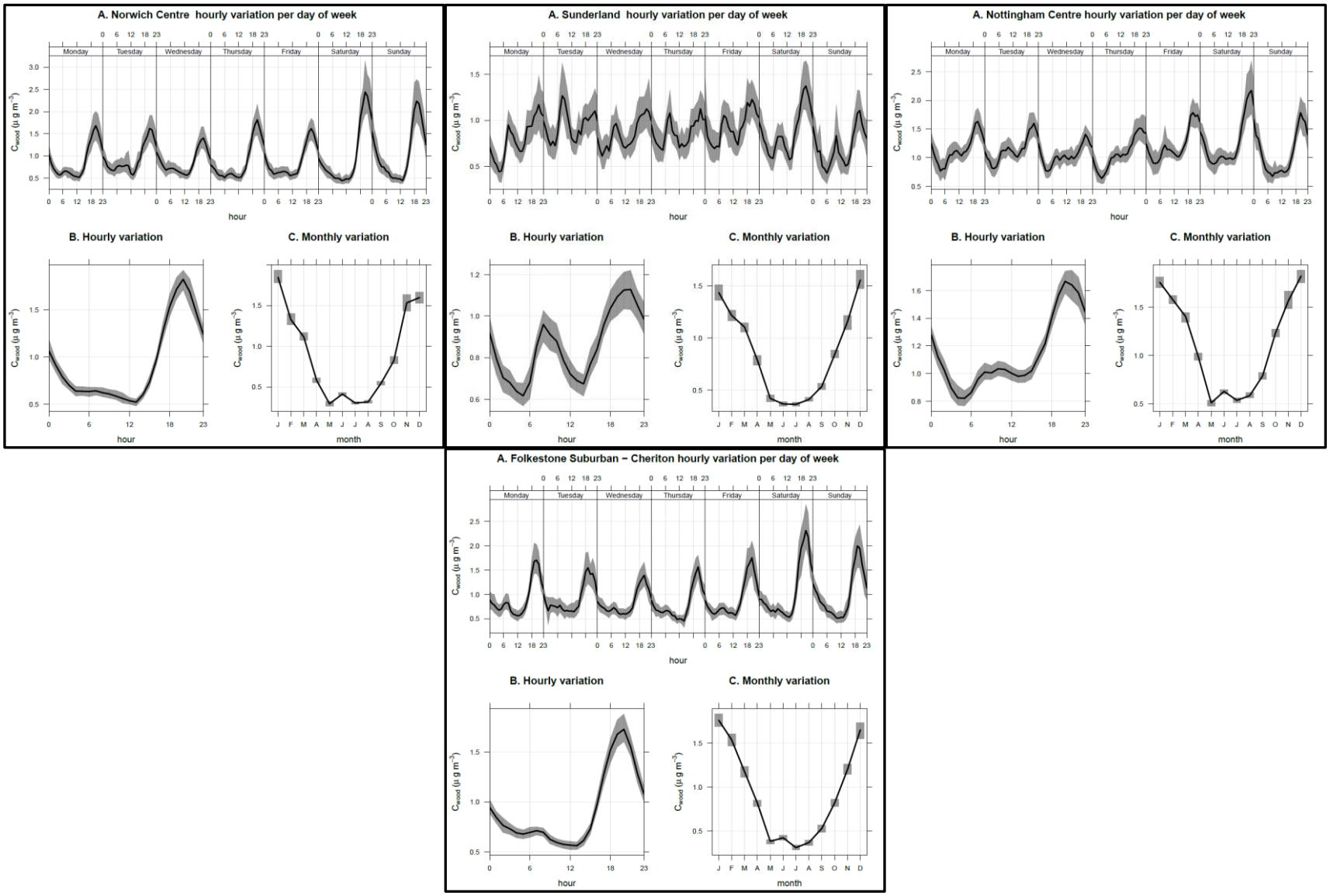
Supplementary Figure 3. Comparison of wood burning concentrations (expressed in µg m^{-3}) as derived from the 7 and 2 wavelength aethalometers collocated at North Kensington in Dec'15-Feb'16. Red line indicates the 1:1 line. Regression values were

SM2. Time dynamics of C_{wood}




A. Glasgow Centre hourly variation per day of week
A. Glasgow Townhead hourly variation per day of week
A. Woolwich hourly variation per day of week
A. Harwell hourly variation per day of week
A. South Kirkby hourly variation per day of week
A. Halifax hourly variation per day of week
Monday
Tuesday
Wednesday
Thursday
Friday
Saturday
Sunday
hour
B. Hourly variation
C. Monthly variation
month
J F M A M J J A S O N D


A. Stoke−on−Trent Centre AURN hourly variation per day of week
A. Bradford Town Hall hourly variation per day of week
A. Maidstone Rural − Detling hourly variation per day of week
A. Birmingham Tyburn hourly variation per day of week
A. North Kensington hourly variation per day of week
A. Manchester Piccadilly hourly variation per day of week
Monday
Tuesday
Wednesday
Thursday
Friday
Saturday
Sunday
hour
B. Hourly variation
C. Monthly variation
month
J F M A M J J A S O N D
C_{wood} (μ g m^{-3})


A. Norwich Centre hourly variation per day of week
Monday
Tuesday
Wednesday
Thursday
Friday
Saturday
Sunday
hour
B. Hourly variation
C. Monthly variation
month
A. Sunderland hourly variation per day of week
A. Nottingham Centre hourly variation per day of week
A. Folkestone Suburban – Cheriton hourly variation per day of week

SM3. C_{wood} in Particulate Matter

Supplementary Table 1. Annual C_{wood} / PM_{10} ratios for sites with data capture > 75%.

Site	Year	C_{wood} / PM_{10}	Data capture (%)
Cardiff	2009	7%	83
Birmingham Tyburn	2009	4%	84
London North Kensington	2009	5%	82
Stoke-on-Trent Centre	2009	6%	88
Folkestone - Cheriton	2009	6%	85
Edinburgh St Leonards	2010	5%	94
Harwell	2010	3%	76
Birmingham Tyburn	2010	5%	85
Nottingham Centre	2010	5%	89
Stoke-on-Trent Centre	2010	6%	94
Edinburgh St Leonards	2011	4%	97
Glasgow Centre	2011	5%	88
Harwell	2011	2%	78
Birmingham Tyburn	2011	5%	76
London North Kensington	2011	4%	85
Nottingham Centre	2011	4%	93
Stoke-on-Trent Centre	2011	5%	89
Cardiff	2012	8%	78
Harwell	2012	3%	94
Birmingham Tyburn	2012	6%	96
London North Kensington	2012	5%	78
Maidstone - Detling	2012	4%	92
Cardiff	2013	7%	91
Birmingham Tyburn	2013	5%	88
Auchencorth Moss	2014	3%	91
Birmingham Tyburn	2014	4%	82
Harwell	2015	2%	88
Birmingham Tyburn	2015	4%	94
London North Kensington	2015	4%	95

Supplementary Table 2. Winter C_{wood} / PM_{10} ratios for sites with data capture > 75%.

Site	Winter	C_{wood} / PM_{10}	Data capture (%)
Cardiff	DJF 2009-2010	13%	80
Edinburgh St Leonards	DJF 2009-2010	7%	77
Harwell	DJF 2009-2010	4%	79
Birmingham Tyburn	DJF 2009-2010	6%	96
London North Kensington	DJF 2009-2010	7%	97
Nottingham Centre	DJF 2009-2010	7%	100
Stoke-on-Trent Centre	DJF 2009-2010	11%	87
Edinburgh St Leonards	DJF 2010-2011	8%	97
Glasgow Centre	DJF 2010-2011	8%	97
Birmingham Tyburn	DJF 2010-2011	6%	99
London North Kensington	DJF 2010-2011	7%	98
Nottingham Centre	DJF 2010-2011	7%	94
Cardiff	DJF 2011-2012	9%	95
Glasgow Centre	DJF 2011-2012	6%	89
Harwell	DJF 2011-2012	3%	98
Birmingham Tyburn	DJF 2011-2012	5%	96
London North Kensington	DJF 2011-2012	5%	90
Nottingham Centre	DJF 2011-2012	6%	100
Stoke-on-Trent Centre	DJF 2011-2012	7%	97
Auchencorth Moss	DJF 2012-2013	3%	95
Harwell	DJF 2012-2013	5%	78
Birmingham Tyburn	DJF 2012-2013	6%	97
London North Kensington	DJF 2012-2013	6%	96
Maidstone - Detling	DJF 2012-2013	5%	99
Auchencorth Moss	DJF 2013-2014	6%	89
Cardiff	DJF 2013-2014	8%	92
Harwell	DJF 2013-2014	5%	89
Birmingham Tyburn	DJF 2013-2014	5%	94
London North Kensington	DJF 2013-2014	5%	87
Auchencorth Moss	DJF 2014-2015	5%	95
Harwell	DJF 2014-2015	4%	92
Birmingham Tyburn	DJF 2014-2015	5%	96
London North Kensington	DJF 2014-2015	6%	90
Auchencorth Moss	DJF 2015-2016	3%	78
Birmingham Tyburn	DJF 2015-2016	5%	84
London North Kensington	DJF 2015-2016	5%	94

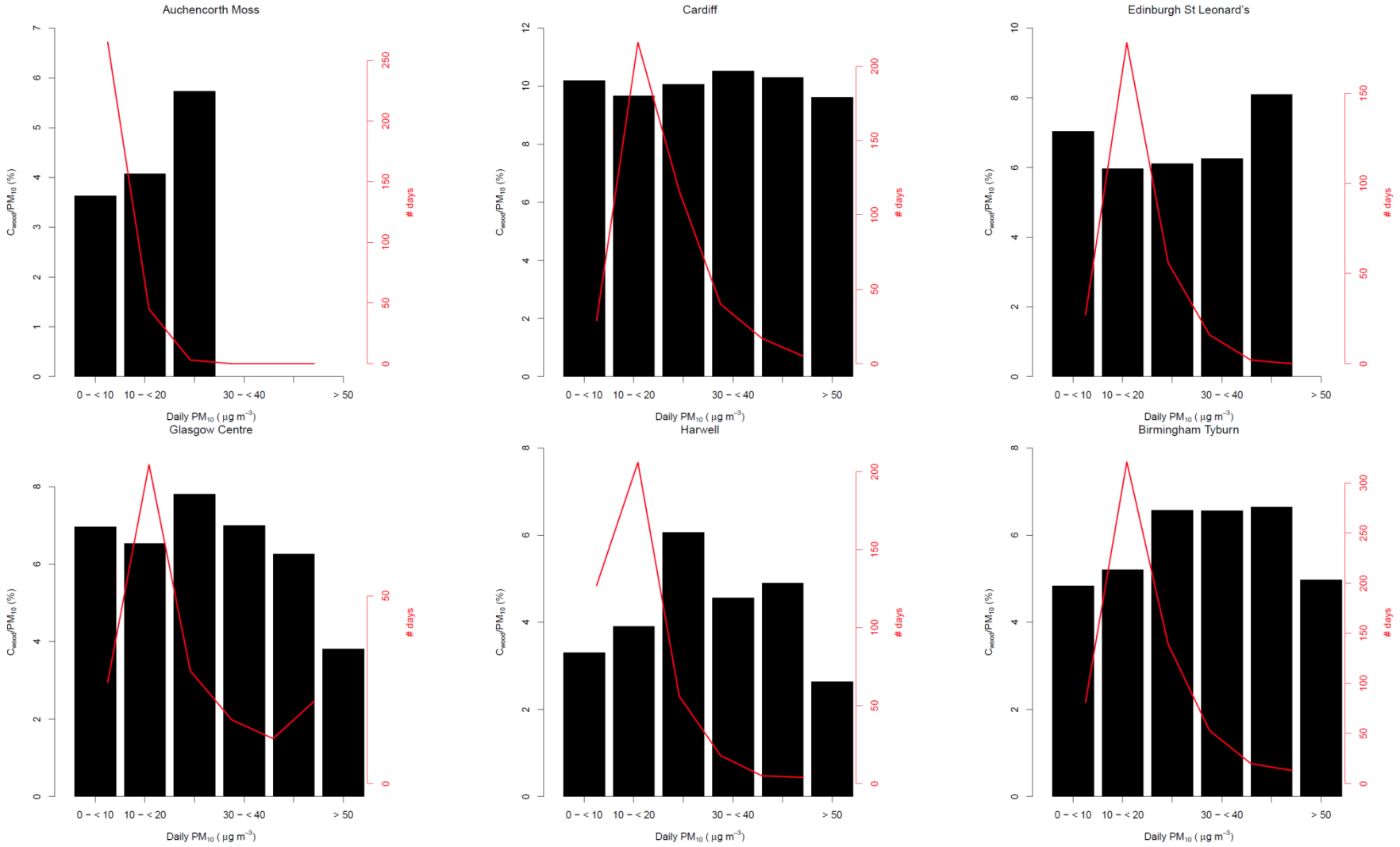
Supplementary Table 3. Annual C_{wood} / $PM_{2.5}$ ratios for sites with data capture > 75%.

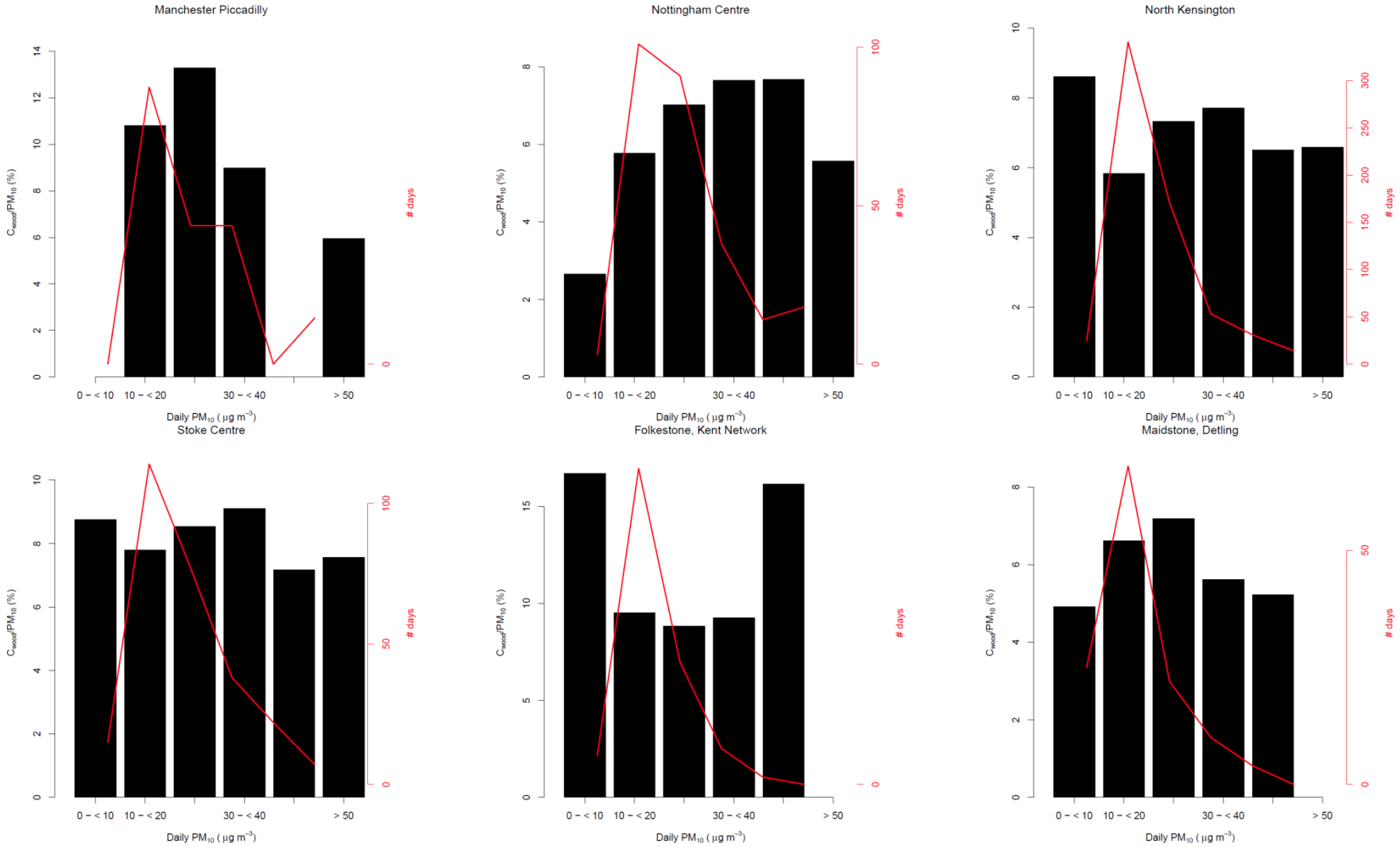
Site	Year	C_{wood} / $PM_{2.5}$	Data capture (%)
London North Kensington	2009	7%	93
Cardiff	2010	12%	85
Edinburgh St Leonards	2010	8%	92
Glasgow Centre	2010	11%	94
Harwell	2010	6%	98
Birmingham Tyburn	2010	6%	90
London North Kensington	2010	8%	81
Nottingham Centre	2010	10%	76
Stoke-on-Trent Centre	2010	9%	96
Edinburgh St Leonards	2011	5%	97
Glasgow Centre	2011	10%	92
Harwell	2011	4%	92
Birmingham Tyburn	2011	8%	79
Manchester Piccadilly	2011	10%	87
London North Kensington	2011	6%	91
Nottingham Centre	2011	10%	91
Stoke-on-Trent Centre	2011	7%	94
Harwell	2012	4%	94
Birmingham Tyburn	2012	7%	97
London North Kensington	2012	8%	86
Maidstone - Detling	2012	8%	87
Cardiff	2013	10%	91
London North Kensington	2013	7%	94
Auchencorth Moss	2014	3%	82
Harwell	2014	5%	96
Birmingham Tyburn	2014	6%	93
Auchencorth Moss	2015	4%	95
Harwell	2015	4%	95
Birmingham Tyburn	2015	7%	93
London North Kensington	2015	8%	94

Supplementary Table 4. Winter C_{wood} / $PM_{2.5}$ ratios for sites with data capture > 75%.

Site	Winter	C_{wood} / $PM_{2.5}$	Data capture (%)
Harwell	DJF 2009-2010	7%	86
London North Kensington	DJF 2009-2010	11%	78
Cardiff	DJF 2010-2011	17%	89
Edinburgh St Leonards	DJF 2010-2011	9%	97
Glasgow Centre	DJF 2010-2011	13%	97
Harwell	DJF 2010-2011	8%	96
Birmingham Tyburn	DJF 2010-2011	7%	96
Manchester Piccadilly	DJF 2010-2011	13%	94
London North Kensington	DJF 2010-2011	8%	93
Nottingham Centre	DJF 2010-2011	12%	78
Stoke-on-Trent Centre	DJF 2010-2011	10%	89
Cardiff	DJF 2011-2012	14%	98
Glasgow Centre	DJF 2011-2012	12%	97
Harwell	DJF 2011-2012	4%	99
Birmingham Tyburn	DJF 2011-2012	9%	99
Manchester Piccadilly	DJF 2011-2012	9%	93
London North Kensington	DJF 2011-2012	7%	95
Nottingham Centre	DJF 2011-2012	13%	100
Auchencorth Moss	DJF 2012-2013	6%	95
Harwell	DJF 2012-2013	6%	82
Birmingham Tyburn	DJF 2012-2013	6%	99
London North Kensington	DJF 2012-2013	10%	97
Maidstone - Detling	DJF 2012-2013	10%	99
Auchencorth Moss	DJF 2013-2014	6%	77
Cardiff	DJF 2013-2014	13%	92
Harwell	DJF 2013-2014	9%	91
Birmingham Tyburn	DJF 2013-2014	12%	94
London North Kensington	DJF 2013-2014	9%	90
Auchencorth Moss	DJF 2014-2015	3%	94
Harwell	DJF 2014-2015	8%	92
Birmingham Tyburn	DJF 2014-2015	7%	96
London North Kensington	DJF 2014-2015	9%	86
Auchencorth Moss	DJF 2015-2016	4%	76
Birmingham Tyburn	DJF 2015-2016	7%	96
London North Kensington	DJF 2015-2016	10%	92

SM4. C_{wood}/PM_{10} for daily PM_{10} concentrations





Supplementary Figure 4. Winter C_{wood}/PM_{10} for different daily PM_{10} concentrations. The red line indicates the number of days available for each PM bin.

SM5. C_{wood} / NO_X ratios

Supplementary Table 5. Annual C_{wood} / NO_X ratios for sites with data capture > 75%.

Site	Year	C_{wood} / NOx (µg µg^{-1})	Data capture (%)
Cardiff	2009	0.031	98
Cardiff	2010	0.034	100
Cardiff	2011	0.033	98
Cardiff	2012	0.036	99
Cardiff	2013	0.035	99
Edinburgh St Leonards	2009	0.016	97
Edinburgh St Leonards	2010	0.017	99
Edinburgh St Leonards	2011	0.019	99
Glasgow Centre	2009	0.014	88
Glasgow Centre	2010	0.014	95
Glasgow Centre	2011	0.016	98
Harwell	2010	0.049	100
Harwell	2011	0.038	98
Harwell	2012	0.041	96
Harwell	2013	0.045	99
Harwell	2014	0.041	100
Harwell	2015	0.040	99
Birmingham Tyburn	2009	0.015	90
Birmingham Tyburn	2010	0.023	94
Birmingham Tyburn	2011	0.020	100
Birmingham Tyburn	2012	0.022	98
Birmingham Tyburn	2013	0.020	90
Birmingham Tyburn	2014	0.016	96
Birmingham Tyburn	2015	0.015	98
Manchester Piccadilly	2009	0.015	90
Manchester Piccadilly	2010	0.015	100
Manchester Piccadilly	2011	0.013	98
London North Kensington	2009	0.019	95
London North Kensington	2010	0.021	88
London North Kensington	2011	0.020	95
London North Kensington	2012	0.017	97
London North Kensington	2013	0.018	99
London North Kensington	2014	0.018	89
London North Kensington	2015	0.020	97
Nottingham Centre	2009	0.017	96
Nottingham Centre	2010	0.018	100
Nottingham Centre	2011	0.019	100
Stoke-on-Trent Centre	2009	0.023	91
Stoke-on-Trent Centre	2010	0.024	100
Stoke-on-Trent Centre	2011	0.022	99
Folkestone - Cheriton	2009	0.039	97
Maidstone - Detling	2012	0.039	94
Maidstone - Detling	2013	0.074	88

Supplementary Table 6. Winter C_{wood} / NO_X ratios for sites with data capture > 75%.

Site	Winter	C_{wood} / NOx (ng µg^{-1})	Data capture (%)
Cardiff	DJF 2009-2010	0.038	95
Cardiff	DJF 2010-2011	0.042	93
Cardiff	DJF 2011-2012	0.040	94
Cardiff	DJF 2013-2014	0.028	95
Edinburgh St Leonards	DJF 2009-2010	0.022	94
Edinburgh St Leonards	DJF 2010-2011	0.018	95
Glasgow Centre	DJF 2009-2010	0.014	86
Glasgow Centre	DJF 2010-2011	0.015	91
Glasgow Centre	DJF 2011-2012	0.016	94
Harwell	DJF 2009-2010	0.043	84
Harwell	DJF 2010-2011	0.045	96
Harwell	DJF 2011-2012	0.039	94
Harwell	DJF 2012-2013	0.055	78
Harwell	DJF 2013-2014	0.039	93
Harwell	DJF 2014-2015	0.068	95
Birmingham Tyburn	DJF 2009-2010	0.023	96
Birmingham Tyburn	DJF 2010-2011	0.015	96
Birmingham Tyburn	DJF 2011-2012	0.019	98
Birmingham Tyburn	DJF 2012-2013	0.020	97
Birmingham Tyburn	DJF 2013-2014	0.018	95
Birmingham Tyburn	DJF 2014-2015	0.014	97
Birmingham Tyburn	DJF 2015-2016	0.019	97
Manchester Piccadilly	DJF 2009-2010	0.014	91
Manchester Piccadilly	DJF 2010-2011	0.014	95
Manchester Piccadilly	DJF 2011-2012	0.014	97
London North Kensington	DJF 2010-2011	0.020	96
London North Kensington	DJF 2011-2012	0.018	96
London North Kensington	DJF 2012-2013	0.020	96
London North Kensington	DJF 2013-2014	0.018	95
London North Kensington	DJF 2014-2015	0.021	90
London North Kensington	DJF 2015-2016	0.021	97
Nottingham Centre	DJF 2009-2010	0.019	98
Nottingham Centre	DJF 2010-2011	0.020	94
Nottingham Centre	DJF 2011-2012	0.019	97
Stoke-on-Trent Centre	DJF 2009-2010	0.030	95
Stoke-on-Trent Centre	DJF 2010-2011	0.026	92
Stoke-on-Trent Centre	DJF 2011-2012	0.026	96

SM6. Trends in C_{wood}

Supplementary Table 7. Trends in C_{wood} between January 2009 to December 2011.

TRENDS 2009 - 2011								
Site	**Start Date**	**End Date**	**Method**	**Slope ($\mu g\ m^{-3}\ year^{-1}$)**	**Lower ($\mu g\ m^{-3}\ year^{-1}$)**	**Upper ($\mu g\ m^{-3}\ year^{-1}$)**	**Slope (% $year^{-1}$)**	**N**
Bath	01-Jan-09	01-Jan-12	All data	-0.12	-0.34	0.10	-10.05	36
Bath	01-Jan-09	01-Jan-12	All data - deseason	-0.20	-0.27	-0.09	-12.53 **	36
Bath	DJF 2009-10	DJF 2011-12	Winter	-0.03	-0.66	0.61	-0.90	3
Cardiff	01-Jan-09	01-Jan-12	All data	-0.11	-0.37	0.17	-8.15	36
Cardiff	01-Jan-09	01-Jan-12	All data - deseason	-0.11	-0.17	-0.03	-6.64 *	36
Cardiff	DJF 2009-10	DJF 2011-12	Winter	-0.34	-1.00	0.33	-9.18	3
Dudley Central	01-Jan-09	01-Jan-12	All data	-0.01	-0.20	0.15	-1.09	36
Dudley Central	01-Jan-09	01-Jan-12	All data - deseason	0.04	-0.07	0.09	4.27	36
Dudley Central	DJF 2009-10	DJF 2011-12	Winter	-0.32	-0.74	0.10	-12.42	3
Edinburgh St Leonards	01-Jan-09	01-Jan-12	All data	-0.01	-0.10	0.14	-1.64	36
Edinburgh St Leonards	01-Jan-09	01-Jan-12	All data - deseason	0.03	-0.02	0.08	4.95	36
Edinburgh St Leonards	DJF 2009-10	DJF 2011-12	Winter	-0.21	-0.43	0.01	-11.61	3
Glasgow Centre	01-Jan-09	01-Jan-12	All data	-0.09	-0.28	0.07	-8.33	36
Glasgow Centre	01-Jan-09	01-Jan-12	All data - deseason	-0.04	-0.18	0.07	-4.44	36
Glasgow Centre	DJF 2009-10	DJF 2011-12	Winter	-0.38	-0.66	-0.10	-13.44	3
Woolwich	01-Jan-09	01-Jan-12	All data	-0.12	-0.26	0.06	-11.43	36
Woolwich	01-Jan-09	01-Jan-12	All data - deseason	-0.10	-0.15	-0.06	-9.33 ***	36
Woolwich	DJF 2009-10	DJF 2011-12	Winter	-0.16	-0.24	-0.08	-6.87	3
Harwell	01-Jan-09	01-Jan-12	All data	-0.11	-0.37	0.11	-21.88	24
Harwell	01-Jan-09	01-Jan-12	All data - deseason	-0.11	-0.37	0.11	-21.88	24
Harwell	DJF 2009-10	DJF 2011-12	Winter	-0.13	-0.44	0.19	-10.35	3
Halifax	01-Jan-09	01-Jan-12	All data	-0.08	-0.23	0.10	-7.76	36
Halifax	01-Jan-09	01-Jan-12	All data - deseason	-0.05	-0.10	-0.01	-4.74 *	36
Halifax	DJF 2009-10	DJF 2011-12	Winter	-0.04	-0.29	0.22	-1.68	3
Birmingham Tyburn	01-Jan-09	01-Jan-12	All data	-0.06	-0.31	0.19	-5.47	36
Birmingham Tyburn	01-Jan-09	01-Jan-12	All data - deseason	0.05	-0.07	0.17	5.94	36

Birmingham Tyburn	DJF 2009-10	DJF 2011-12	Winter	-0.12	-0.51	0.28	-5.63		3
South Kirkby	01-Jan-09	01-Jan-12	All data	-0.11	-0.39	0.19	-10.89		36
South Kirkby	01-Jan-09	01-Jan-12	All data - deseason	-0.01	-0.15	0.08	-1.17		36
South Kirkby	DJF 2009-10	DJF 2011-12	Winter	-0.26	-0.66	0.13	-10.14		3
Manchester Piccadilly	01-Jan-09	01-Jan-12	All data	-0.13	-0.30	0.07	-10.58		36
Manchester Piccadilly	01-Jan-09	01-Jan-12	All data - deseason	-0.05	-0.11	0.02	-4.26		36
Manchester Piccadilly	DJF 2009-10	DJF 2011-12	Winter	-0.20	-0.67	0.26	-8.37		3
London North Kensington	01-Jan-09	01-Jan-12	All data	-0.05	-0.36	0.21	-4.11		36
London North Kensington	01-Jan-09	01-Jan-12	All data - deseason	0.04	-0.01	0.07	3.47		36
London North Kensington	DJF 2009-10	DJF 2011-12	Winter	-0.30	-0.39	-0.21	-11.11	+	3
Nottingham Centre	01-Jan-09	01-Jan-12	All data	0.02	-0.19	0.23	1.54		36
Nottingham Centre	01-Jan-09	01-Jan-12	All data - deseason	0.10	0.01	0.15	10.86	*	36
Nottingham Centre	DJF 2009-10	DJF 2011-12	Winter	-0.11	-0.57	0.35	-4.20		3
Norwich Centre	01-Jan-09	01-Jan-12	All data	-0.32	-0.64	0.02	-31.35	+	27
Norwich Centre	01-Jan-09	01-Jan-12	All data - deseason	-0.26	-0.37	-0.11	-21.91	***	27
Norwich Centre	DJF 2009-10	DJF 2011-12	Winter	-0.34	-0.68	0.01	-11.71		3
Sunderland	01-Jan-09	01-Jan-12	All data	0.02	-0.26	0.16	2.89		35
Sunderland	01-Jan-09	01-Jan-12	All data - deseason	0.01	-0.09	0.05	0.88		35
Sunderland	DJF 2009-10	DJF 2011-12	Winter	-0.08	-0.25	0.08	-3.99		3
Stoke-on-Trent Centre	01-Jan-09	01-Jan-12	All data	-0.16	-0.41	0.11	-11.78		36
Stoke-on-Trent Centre	01-Jan-09	01-Jan-12	All data - deseason	-0.08	-0.19	0.02	-6.37		36
Stoke-on-Trent Centre	DJF 2009-10	DJF 2011-12	Winter	-0.33	-0.78	0.11	-10.93		3
Folkestone - Cheriton	01-Jan-09	01-Jan-12	All data	-0.05	-0.29	0.22	-5.40		36
Folkestone - Cheriton	01-Jan-09	01-Jan-12	All data - deseason	-0.06	-0.16	0.01	-6.57		36
Folkestone - Cheriton	DJF 2009-10	DJF 2011-12	Winter	-0.22	-0.33	-0.11	-8.13		3

Supplementary Table 8. Trends in C_{wood} between January 2009 to December 2015.

TRENDS 2009 - 2015								
Site	**Start Date**	**End Date**	**Method**	**Slope ($\mu g\ m^{-3}\ year^{-1}$)**	**Lower ($\mu g\ m^{-3}\ year^{-1}$)**	**Upper ($\mu g\ m^{-3}\ year^{-1}$)**	**Slope (% $year^{-1}$)**		**N**
Cardiff	01-Jan-09	01-Jan-16	All data	-0.04	-0.16	0.04	-3.44		65
Cardiff	01-Jan-09	01-Jan-16	All data - deseason	-0.03	-0.07	-0.01	-2.40	**	65
Cardiff	DJF 2009-10	DJF 2014-15	Winter	-0.43	-0.62	-0.25	-15.06	*	6
Harwell	01-Jan-09	01-Jan-16	All data	-0.03	-0.06	0.01	-5.57		72
Harwell	01-Jan-09	01-Jan-16	All data - deseason	-0.02	-0.04	-0.01	-4.36	*	72
Harwell	DJF 2009-10	DJF 2015-16	Winter	-0.07	-0.14	0.00	-6.97		7
Birmingham Tyburn	01-Jan-09	01-Jan-16	All data	-0.07	-0.12	-0.02	-6.87	**	84
Birmingham Tyburn	01-Jan-09	01-Jan-16	All data - deseason	-0.06	-0.09	-0.03	-5.77	***	84
Birmingham Tyburn	DJF 2009-10	DJF 2015-16	Winter	-0.14	-0.19	-0.08	-6.47	**	7
London North Kensington	01-Jan-09	01-Jan-16	All data	-0.04	-0.11	0.02	-3.86		84
London North Kensington	01-Jan-09	01-Jan-16	All data - deseason	-0.01	-0.05	0.01	-1.36		84
London North Kensington	DJF 2009-10	DJF 2015-16	Winter	-0.08	-0.16	-0.01	-5.97	+	7





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