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Anna Font, K Ciupek, D Butterfield, G W Fuller

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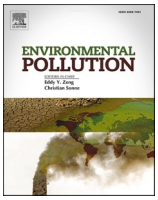

Long-term trends in particulate matter from wood burning in the United Kingdom: Dependence on weather and social factors☆

A. Font [a,*,1], K. Ciupek [a,b], D. Butterfield [b], G.W. Fuller [a]

[a] MRC Centre for Environment and Health, Environmental Research Group, Imperial College, London, UK
[b] Air Quality and Aerosol Metrology Group, National Physical Laboratory, Teddington, UK

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ABSTRACT

Particulate matter from wood burning emissions (C_{wood}) was quantified at five locations in the United Kingdom (UK), comprising three rural and two urban sites between 2009 and 2021. The aethalometer method was used. Mean winter C_{wood} concentrations ranged from 0.26 μg m^{-3} (in rural Scotland) to 1.30 μg m^{-3} (London), which represented on average 4% (in rural environments) and 5% (urban) of PM_{10} concentrations; and 8% of $PM_{2.5}$. Concentrations were greatest in the evenings in winter months, with larger evening concentrations in the weekends at the urban sites. Random-forest (RF) machine learning regression models were used to reconstruct C_{wood} concentrations using both meteorological and temporal explanatory variables at each site. The partial dependency plots indicated that temperature and wind speed were the meteorological variables explaining the greatest variability in C_{wood}, with larger concentrations during cold and calm conditions. Peaks of C_{wood} concentrations took place during and after events that are celebrated with bonfires. These were Guy Fawkes events in the urban areas and on New Year's Day at the rural sites; the later probably related to long-range transport. Time series were built using the RF. Having removed weather influences, long-term trends of C_{wood} were estimated using the Theil Sen method. Trends for 2015–2021 were downward at three of the locations (London, Glasgow and rural Scotland), with rates ranging from −5.5% $year^{-1}$ to −2.5% $year^{-1}$. The replacement of old fireplaces with lower emission wood stoves might explain the decrease in C_{wood} especially at the urban sites The two rural sites in England observed positive trends for the same period but this was not statistically significant.

1. Introduction

Biomass burning from domestic heating releases harmful air pollutants to the atmosphere including gases such as carbon monoxide (CO) and nitrogen oxides (NO_X) alongside primary fine particulate matter ($PM_{2.5}$). Fresh particles include toxic air pollutants such as benzene, formaldehyde, acrolein and polycyclic aromatic hydrocarbons (PAHs) including Benzo[a]pyrene (B[a]P) (Kirchsteiger et al., 2021). Particles emitted during wood combustion also include both elemental and organic carbon and inorganic ions (e.g., fine potassium) (Fine et al., 2004a, 2004b; 2001). The proportion of organic to elemental carbon depends on the conditions of combustion, with more organics emitted in suboptimal combustion processes. Secondary organic aerosols formed from atmospheric oxidation reactions of wood burning emissions represent a big proportion of particles related to wood burning emissions (Bruns et al., 2015; Bruns et al., 2017).

There is substantial evidence that links chronic exposure to biomass smoke with adverse health effects, notably chronic obstructive pulmonary disease (COPD) (Capistrano et al., 2017). 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 Regional Office for Europe, 2015). The toxicity of wood smoke particles seems to strongly depend on the organic fraction and probably associated with the organic components (Bolling et al., 2012). In a study performed in Athens (Greece), PAH from wood burning was associated with an increased health risk, accounting for almost half the annual PAH carcinogenic potential (Tsiodra et al., 2021). Wood smoke also impacts on ecosystems reducing visibility (haze) and creating environmental and aesthetic damage.

Further evidence on the health impacts of air pollution from wood

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* Corresponding author.
E-mail address: anna.font@imt-nord-europe.fr (A. Font).
[1] now at: IMT Nord Europe, Centre d'Enseignement, de Recherche et d'Innovation Énergie Environnement (CERI EE), Douai, France.

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burning arises from intervention studies where wood burning emissions were decreased in communities. In Launceston (Tasmania), an education programme and financial incentives decreased wood burning usage from 66% to 30% of households. Winter mean PM_{10} fell by 40%; wintertime respiratory deaths reduced by 28% and heart deaths by 20% (Johnston et al., 2013). In Libby (Montana, United States) a program to replace old wood stoves with new ones improved wintertime $PM_{2.5}$ by 30%. The decrease in $PM_{2.5}$ from wood smoke was associated with decreased reports of childhood wheeze and other respiratory health conditions, and also less illness-related school absences, although this latter finding was not consistent across all age-groups (Noonan et al., 2011). In the San Joaquin Valley (California, United States) bans on wood burning on the most polluted days reduced wintertime ambient $PM_{2.5}$ levels by 11–15% and decreased hospital admissions for heart disease among people aged 65 years and older (Yap and Garcia, 2015).

Emission rates of $PM_{2.5}$ from wood burning are based on estimates of emission factors and consumption rates. Emission factors are taken from measurements on a variety of wood burning burners and wood types. Consumption rates can be calculated from either surveys or total wood consumption. In the UK, a survey by Waters (2016) found that 7.5% of UK homes burnt wood for heating. A follow up survey in 2018–2019 concluded that the proportion of homes burning wood was relatively stable. It found that UK householders were shifting to more efficient appliances but these may be used more on average (Kantar, 2020). According to the UK National Atmospheric Emissions Inventory (NAEI) domestic combustion was responsible for 19.8 kt of $PM_{2.5}$ in 2020, which represented almost a quarter of total emissions, with wood burning being responsible for 17%. The NAEI also estimated that emissions of $PM_{2.5}$ from domestic wood burning increased by 35% between 2010 and 2020 (GOV.UK, 2022) based on estimated energy consumption (BEIS, 2016) and an increase in the growth rate in stove installations of 50% each two to three years from 2003 onwards. Sources of data included the National Association of Chimney sweeps, stove and fireplace installers, an insurance assessor and the UK Forestry Commission.

In the UK, the Clean Air Act of 1956 established Smoke Control Areas (SCA) where emissions of smoke from domestic premises are banned except if using authorised fuel or using an exempt appliance. One effect of this is to ban wood and coal burning in open fires. The majority of local authorities in large English cities such as London and Birmingham are covered by SCAs. In Scotland, the Clean Air Act in 1993 defined many parts of Scotland as SCA. However, large increases in biomass burning are still projected from energy scenarios over the next two decades, due to increased use of renewable energy sources (Fuller et al., 2013; Williams et al., 2017). Regulations and frameworks exist to decrease emissions from new residential heating appliances. These include the European Directive 2009/125/EC with regard to eco-design requirements for solid fuel local space heaters. Additionally, the Approved J document for combustion appliances and fuel storage systems (HM-Government, 2015), and the Domestic Renewable Heat Incentive for England, Wales and Scotland (GOV.GOV.UK, 2013) place restrictions on the emissions from new wood heating systems. New bans on the sale of wet wood (Defra, 2021) also aim to reduce emissions.

Atmospheric $PM_{2.5}$ concentrations from wood burning can be estimated using different proxies including levoglucosan measurements (Bhattarai et al., 2019; Cordell et al., 2014; Puxbaum et al., 2007; Yttri et al., 2005); fine potassium (Andreae, 1983; Hsu et al., 2009; Pachon et al., 2013); the absorption characteristics of aerosols as measured by the aethalometer (Grange et al., 2020; Sandradewi et al., 2008b; Zotter et al., 2017); and/or from estimates using the Positive Matrix Factorization on multiple element measurements (Karagulian et al., 2015; Molnár and Sallsten, 2013) or organic carbon (Wagener et al., 2012; Yttri et al., 2021; Canonaco et al., 2013; Reyes-Villegas et al., 2016; Young et al., 2014; Zhang et al., 2019). In Europe, evidence of aerosols from wood burning have been found in both rural areas (Kristensson et al., 2020) and cities such as Paris (Favez et al., 2009), London (Fuller et al., 2014) and Berlin (Wagener et al., 2012). The proportion of wood burning aerosols varied from location to location and may also vary according to the proxy method used. In Paris, wood burning aerosols represented 20 ± 10% in $PM_{2.5}$ in winter 2005 (Favez et al., 2009) and ~10% of winter PM_{10} in London (Fuller et al., 2014). Viana et al. (2013) estimated that 8% of the $PM_{2.5}$ in winter 2011 in Barcelona was attributed to transport of biomass burning emissions from rural areas to the city. Wood burning was estimated to contribute to about 3.1% of the winter PM_{10} mass in Oporto, and to 3.7% in Copenhagen (Caseiro and Oliveira, 2012). Based on levoglucosan measurements, it was estimated that biomass burning contributed between 2.7% and 11.6% to winter PM_{10} in five cities in north-west Europe (Cordell et al., 2016). From Positive Matrix Factorization studies, $PM_{2.5}$ associated with domestic fuel combustion (including both wood burning and coal combustion) in western and north-western Europe was on average 15 and 22%, respectively (Karagulian et al., 2015).

Although these studies are useful to determine the contribution of wood burning to PM at a point in time, long-term observations are needed to track the impacts of changes in home heating and the impacts of renewable energy policies. In this study we report long-term trends in ambient particle concentrations from wood burning (C_{wood}) in different locations in the UK using the aethalometer method. We characterized the temporal dynamics of C_{wood} concentrations in the UK to determine the behavioural factors that influence wood heating, and the long-term trends, having removed possible confounding from variable meteorological conditions. In this way, changes in ambient C_{wood} concentrations were solely attributed to changes in emissions and not to changes in weather parameters and dispersion conditions.

2. Methods

2.1. Monitoring sites and temporal coverage

PM concentration from wood burning (C_{wood}) was calculated for those sites belonging to the Department for Environment, Food and Rural Affairs (Defra) black carbon network (Ciupek et al., 2021) that had more than five years of measurement in the period January 1, 2009 and June 30, 2021 (Fig. 1). Data from 2021 was subject to final quality assurance at the time of writing. Roadside sites were excluded from C_{wood} calculations and only data from urban background, suburban and rural sites were included. Sites in Northern Ireland were also excluded as other solid fuels such as peat and coal are burned here in large quantities (Brown & Brown, 2012; Kantar, 2020) and may therefore interfere with the aethalometer quantification (Brown et al., 2016). A total of five sites were selected: Glasgow Townhead (GLA; urban background site) and Auchencorth Moss (AUC; rural background) in Scotland; and London North Kensington (LNK; urban background), Chilbolton Observatory (CHO; rural) and Maidstone-Detling (DET; rural) in south-east England. A brief description of each site is given in the Supplementary Information. Additionally, data from London Marylebone Road (LMR), a kerbside site in central London, next to a 6-lane traffic road, was used to verify the distribution of the values for Ångström coefficients for road traffic.

2.2. The aethalometer model to calculate wood burning aerosols mass concentrations

The PM mass concentrations from wood burning (C_{wood}) was calculated using the aethalometer method (Sandradewi et al., 2008b) based on the light absorption behaviours of wood burning and traffic fossil fuel combustion aerosols; with biomass aerosols absorbing more at shorter wavelengths (Kirchstetter and Novakov, 2007; Sandradewi et al., 2008a). For a complete description of the method the reader is referred to Sandradewi et al. (2008b); and a detailed summary is presented in the Supplementary Information. Briefly, the method relies on the Ängström exponent (α) derived from the Beer-Lambert law and its

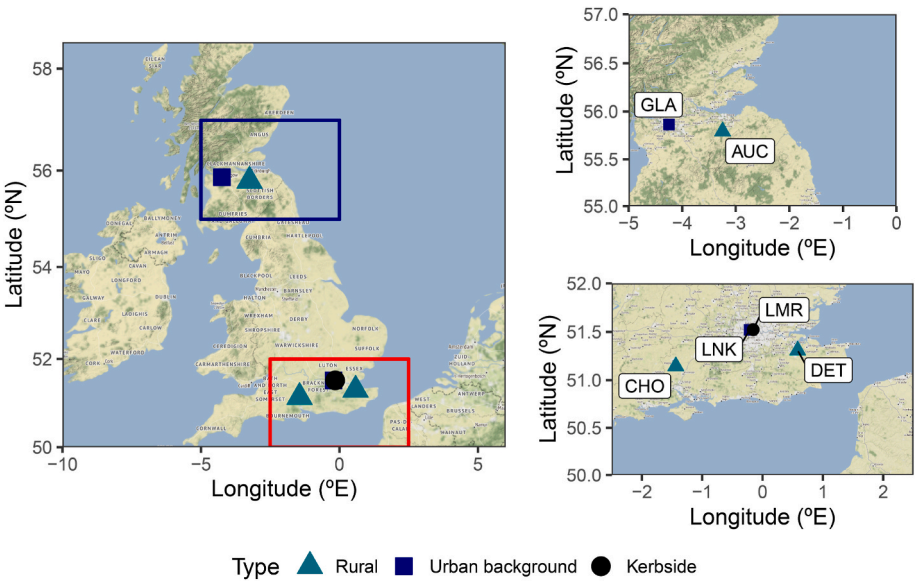

Fig. 1. Map with the aethalometer sites in the UK used in this study.

value can provide information about the predominant aerosol type of source (e.g. Cazorla et al., 2013; Lewis et al., 2008). It assumes that two varying sources of carbonaceous aerosols exist: traffic and wood burning. The α for traffic was obtained by comparing wood burning estimates in London from levoglucosan and those from the aethalometer method (Fuller et al., 2014). A similar approach was also taken by Diapouli et al. (2017) in Athens. The α value for wood burning was also selected based on Fuller et al. (2014). The α for wood burning aerosol will vary between individual sources (Harrison et al., 2012). However, our study looks to characterise sources in background locations where the influence of any individual source is diluted. The plausibility of these α values was tested prior to their application, as discussed in the results section.

Absorption data from the dual wavelength Magee Scientific aethalometer (AE-22) from 2009 was used until November 2019; and then from the seven wavelength Magee Scientific aethalometer AE-33. No evident step-change was observed in the C_{wood} calculations associated with the change of instrument (Supplementary Fig. 1). The Magee AE-22 aethalometer samples air at 4 l min^{-1} and the Magee AE-33 at 5 l min^{-1} and in this study, both collected $PM_{2.5}$ particles onto a filter tape. The light attenuation through the sampled filter at 370 nm (in the UV) and at 880 nm (in the IR) was measured.

2.3. Random forest modelling and de-weathering algorithm

A de-weathering algorithm based on the Random Forest (RF) machine learning modelling was applied to remove the possible influence of meteorological conditions in the temporal trends. This accounted for changes in meteorology over time so that observed trends can be associated with changes in emission patterns or chemistry, independent of changes in meteorology (Grange et al., 2018). To build the de-weathered time series, a decision-tree based RF regression model was built, describing the hourly concentrations of wood burning as predictor variable. The bagging algorithm was used, i.e., random samples of the original data set and their explanatory variables were taken to train the model. Multiple trees (300) were produced and combined to yield a single consensus prediction. A testing data set was then used to evaluate the performance of the model. Here, 70% of the original data set was used to build the model (bagging procedure) and 30% to test it.

A RF model was built for each site using both meteorological variables (wind speed and direction, air temperature, relative humidity, atmospheric pressure and ceilometer height) from the nearest site belonging to the Integrated Surface database and extracted using the R-worldmet package (Carslaw, 2019); and temporal variables to capture the timing of emissions: decimal date (indicated as trend) to represent a possible change over the time; Julian date, day of the week and hour to capture the seasonal, weekly and hourly variation, respectively. A categorical variable indicating when Covid-19 restrictions were in place was also included as an explanatory variable in the RF models. Four periods were included: pre-covid (concentrations as measured before March 2020); pre-lockdown (12–23 March 2020); lockdown (24 March–15 June 2020 for all sites; between 5 November and December 2, 2020; and from January 4, 2021 until April 12, 2021 for the English sites; and between January 4, 2021 until March 1, 2021 for the Scottish sites) and eased lockdown (periods between lockdown periods depending on the nation). Hourly concentrations of C_{wood} were predicted and averaged monthly for trend calculations. Between 12 and 14 random forest models (depending on the availability of meteorological parameters) were tested for each site using different combinations of the explanatory variables. This are summarised in Supplementary Table 2. The performance of each RF model was evaluated based on the root-mean-square error (RMSE) and the correlation coefficient (R^2). Different model formulations were tested for each monitoring site and the one with the highest correlation coefficient on the monthly means was then chosen.

The weather normalization technique predicts the concentration of wood burning at a specific time with randomly selected meteorological and temporal variables using the selected RF model. A new data set was built selecting explanatory variables randomly, except the trend variable. This was then fed to the RF model to predict the concentration of C_{wood} at a particular hour. A total of 1000 predicted concentrations for a specific hour were calculated and the average concentration was then referred as the weather normalized (or de-weathered) concentration. More details about the method can be found in Carslaw & Taylor (2009) and Grange et al. (2018).

One of the most attractive features of the RF method is that it allows an investigation of the relationship between the explanatory variables and the predicted variable through partial dependency plots. These show the median concentration of the variable to predict (C_{wood}) to the range of values of the explanatory variable while holding the value of the other covariates at their mean value. Also, RF ranks explanatory variables based on their importance as a predictor. Partial dependency plots were used to both confirm the plausibility of the model and to evaluate the relationship between meteorological parameters and the

C_{wood} concentration.

2.4. Linear trends

Linear trends of the de-weathered time series of C_{wood} were then calculated using again the TheilSen stimator (Sen, 1968; Theil, 1950) available from the R-*openair* package (Carslaw and Ropkins, 2012). Trends were calculated from monthly means calculated from hourly de-weathered concentrations. A statistically significant trend was assumed when $p < 0.001$ (marked by ***). Whenever $p > 0.1$ the trend was not statistically significant.

3. Results

3.1. Ångström coefficients

The calculation of C_{wood} relies on the input values for the Ångström coefficient for traffic and wood burning. As a first step in our analysis, we verified the applicability of the values used previously in London by Fuller et al. (2014). Mean Ångström coefficients (α) derived from the aethalometers at the rural and urban background sites ranged from 1.14 at Glasgow Townhead (reflecting its city centre location) to 1.36 at Chilbolton Observatory (rural location in southern England). These indicate that a mix of traffic and wood burning emissions were present at the sites. The mean α value at the kerbside site in central London (Marylebone Road) was 1.09, close to 1 associated with traffic aerosols from diesel engines as expected due to the proximity of the monitoring site to the road.

The distribution of hourly α values showed that the majority were within the range $\alpha = 0.96$ and $\alpha = 2$, corresponding to traffic and ambient wood burning values used previously by Fuller et al. (2014). Aside from increased measurement resolution, no substantial changes in the distribution of α values and annual mean values were observed through the study period (Supplementary Fig. 2) indicating the consistency of the Ångström coefficients for traffic and ambient wood burning over time.

The percentage of hours with $\alpha > 2$ was below 5% annually, indicating that $\alpha = 2$ was appropriate for wood burning aerosols. The percentage of hours with $\alpha < 0.96$ was generally below 10% annually with few exceptions: London North Kensington in 2009 and 2018; and Glasgow Townhead in 2013 and 2016 (Supplementary Fig. 3). Whenever $\alpha > 2$, C_{wood} concentrations were negative. These data were removed from RF modelling.

3.2. Overview and temporal dynamics of C_{wood} concentrations

C_{wood} was detected at all monitoring sites, with annual mean concentrations ranging from 0.19 μg m^{-3} at the rural site at Auchencorth Moss; to 0.92 μg m^{-3} at London North Kensington (Table 1). The largest concentrations were observed during the heating season from November to March (Fig. 1), with winter concentrations ranging from 0.26 μg m^{-3} (Auchencorth Moss) to 1.30 μg m^{-3} (North Kensington) (Table 1). Concentrations in November were, on average, greater than those observed in December, probably due to the Guy Fawkes and Diwali festivities with bonfires that take place each November (Bibi et al., 2021; Godri et al., 2010; Hamad et al., 2016; Singh et al., 2015). Concentrations were at a minimum in late spring and summer (May to August) (Fig. 2) suggesting relatively low emissions from the outdoor burning sources, such as bonfires, firepits and patio heaters that would be expected at these times of year.

The C_{wood} concentrations in the winter months (November to March) peaked in the evenings with the lowest concentrations at midday (Fig. 3). The greatest concentrations at the two urban sites were on Saturday evenings, with mean evening concentrations of ~2–2.5 μg m^{-3} at London North Kensington and at 0.7–0.9 μg m^{-3} at Glasgow Townhead. Evening concentrations at all rural sites were similar in magnitude on all days of the week (~2 μg m^{-3} at the English rural sites and ~0.5 μg m^{-3} at the Scottish rural site). Evening concentrations of C_{wood} in London were similar in magnitude to those at the rural sites during weekdays (~2 μg m^{-3}) although the mean concentration on Saturdays was greater at the urban site than at the rural sites of Chilbolton and Maidstone–Detling by 1 μg m^{-3}. The two urban sites observed a second enhancement around 8–9 am coinciding with the morning rush hour when traffic concentrations are expected to peak (Supplementary Fig. 4).

The aethalometer can also be used to apportion black carbon between traffic and wood burning sources. The concentration of black carbon from wood burning is directly proportional to the C_{wood} concentration but additional perspective can be obtained by considering the split between the two sources. As expected, black carbon from traffic was more dominant in urban areas compared with rural locations. For example, black carbon from traffic comprised 62% of black carbon at North Kensington in 2018 but only 27% at Maidstone-Detling.

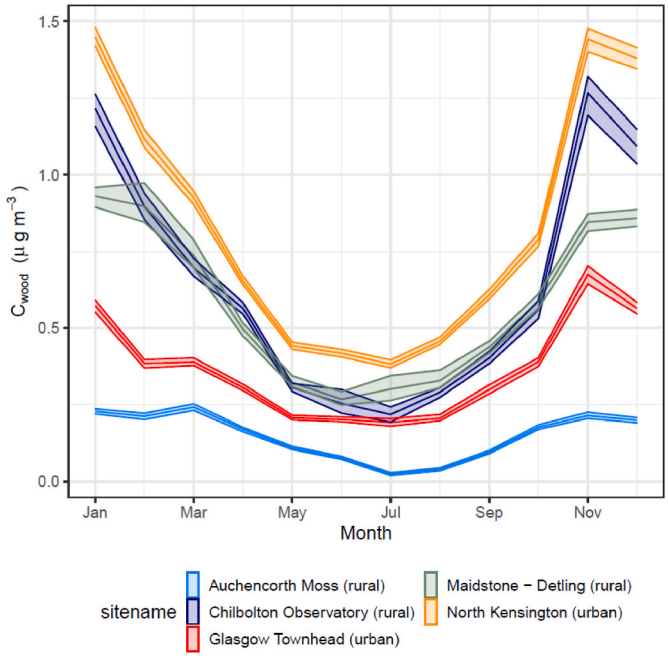

Fig. 2. Mean monthly variations and 95% confidence intervals of C_{wood}.

Table 1
Mean (95% confidence interval) and number of available hourly observations (N). Winter period refers to heating period: November to March.

Site	Period	Mean [95% CI]	N	Mean Winter [95% CI]	N
Auchencorth Moss	2012–2021	0.190 [0.188–0.193]	68,937	0.259 [0.254–0.264]	28,332
Chilbolton Observatory	2016–2021	0.667 [0.655–0.679]	39,771	1.068 [1.043–1.093]	15,199
Glasgow Townhead	2013–2021	0.424 [0.419–0.429]	58,526	0.558 [0.549–0.568]	25,849
Maidstone–Detling	2012–2021	0.655 [0.643–0.666]	69,473	0.895 [0.874–0.916]	31,151
North Kensington	2009–2021	0.917 [0.909–0.924]	97,695	1.315 [1.300–1.330]	42,434

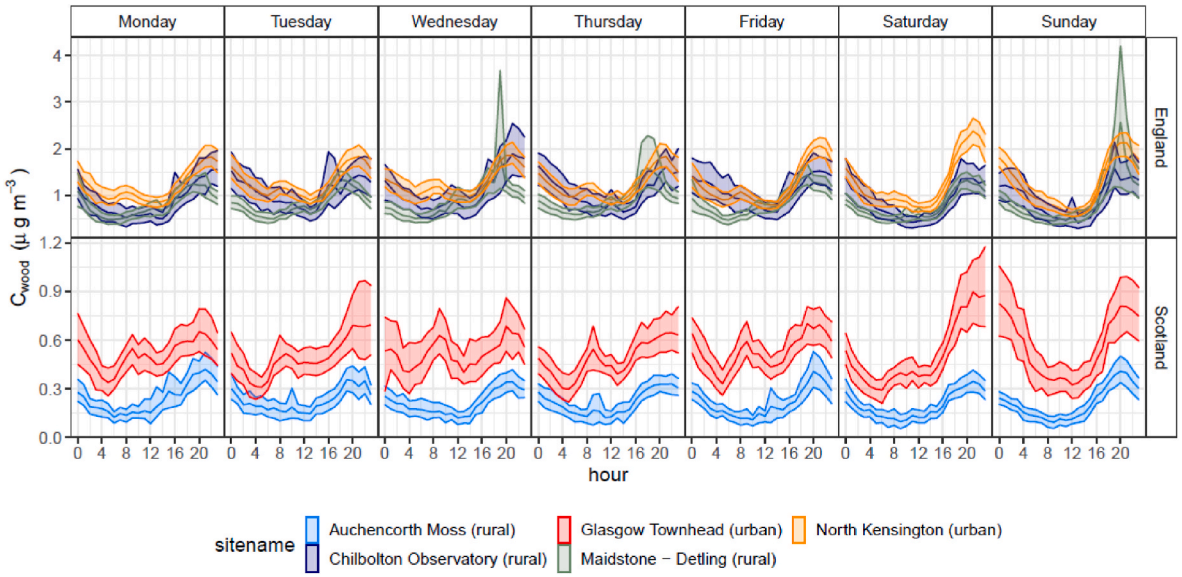

Fig. 3. Mean hourly variation per day of the week (and 95% confidence interval) of C_{wood} concentrations in μg m^{-3} for the heating season (November to March).

3.3. Proportion of C_{wood} in PM mass concentrations

On average, C_{wood} represented 5% (±6% as one standard deviation) of the annual mean PM_{10} concentrations; and 8 ± 11% of $PM_{2.5}$ in urban sites. At London North Kensington the C_{wood} to PM_{10} ratio was larger before 2013 (on average ~6%); falling to around ~3–4%. The contribution to $PM_{2.5}$ was between 7 and 8% in London (except 2009 and 2015 when contributions were >9%). The C_{wood} to PM_{10} ranged between 3 and 6% at Glasgow; and between 18% and 6% for the C_{wood} to $PM_{2.5}$ ratio, with 2013 being the year with the largest contribution to both fractions (Supplementary Table 1). The large ratio in 2013 at Glasgow was because only December data was available for all metrics, a winter month when C_{wood} emissions were expected to be large.

In rural sites, the contribution of C_{wood} to PM_{10} and $PM_{2.5}$ was 4 ± 10%; and 8 ± 20%, respectively. On average, wood burning concentrations were between 2% and 5% of PM_{10} at Auchencorth Moss; and between 3% and 19% of $PM_{2.5}$. The contribution to $PM_{2.5}$ was larger than 10% in 2015 and 2016. At Chilbolton Observatory, C_{wood} concentrations contributed ~3–6% to PM_{10} and 7–10% to $PM_{2.5}$. $PM_{2.5}$ measurements were limited at Maidstone–Detling and only available for 2012 and 2013. C_{wood} contributions to $PM_{2.5}$ were around 9% for those years. The contribution of C_{wood} to PM_{10} was 3–4%.

3.4. Random-forest modelling and influence factors

Twelve to fourteen RF models were tested for each measurement site depending on the availability of meteorological variables (Supplementary Table 2). The RF model formulations that only used weather variables (model#2) were the poorest at reproducing C_{wood} concentrations (R^2 = 0.395–0.744). The best RF models used both meteorological and temporal explanatory variables. The selected RF model for each site reproduced monthly means of C_{wood} with R^2 ranging from 0.823 (Maidstone–Detling) to 0.953 (London North Kensington) (Table 2; Supplementary Table 2 – Supplementary Table 6). Generally, monthly C_{wood} modelled concentrations were slightly underpredicted with slopes of 0.824 (Maidstone–Detling) to 0.991 (Chilbolton Observatory) (Table 2; Supplementary Fig. 5). The RF models were better predictors of C_{wood} concentrations in urban environments when compared with rural. This may be due to C_{wood} being more diffuse in space and concentrations being less in rural areas. The Covid restriction periods were an explanatory variable in the best performing model at the Chilbolton Observatory and the two urban sites (Table 2).

At the three rural sites, the trend over time explained the greatest part of the variability, with 61% of influence at Chilbolton Observatory; almost 50% at Maidstone–Detling; and 24% at Auchencorth Moss. Julian date, capturing the seasonal variability of C_{wood}, was the second most important variable at Maidstone–Detling (13%) and Auchencorth Moss (18%). At Glasgow Townhead, the air temperature was the most important variable, explaining 26% of the variability. At London North Kensington the wind speed and temperature were the two variables that explained the largest part of the C_{wood} variability, accounting for 31% and 26%, respectively. Overall, the Julian date captured more variability of C_{wood} (7–18%) than the hour (4–11%). Air temperature also captured a fair proportion of C_{wood} variability at both Auchencorth Moss and Chilbolton Observatory (12%). At the three sites where the Covid restrictions were an explanatory variable in the RF model (Chilbolton Observatory, Glasgow and London), it captured only 0.2–0.04% of the variability (Fig. 4).

Air temperature and wind speed were the meteorological variables with greatest influence on C_{wood} concentrations. This may indicate first, that wood burning takes place at cold conditions as a source of residential heating; and second, that the dispersion of local emissions is constrained at cold temperatures. For many sites, C_{wood} concentrations levelled off at temperatures >10 °C (Fig. 5A). C_{wood} concentrations were also greatest at low wind speeds when emissions accumulate in the boundary layer. At wind speeds >5 m s^{-1}, C_{wood} concentrations levelled off at all sites with mean concentrations ~0.5 μg m^{-3}.

The partial dependency plot of C_{wood} for Julian date shows the seasonal variation of C_{wood}, with lower concentrations in spring and summer and larger concentrations in late autumn and winter. The greatest mean C_{wood} concentrations were measured on Julian dates 1 and 365 (or 366 if leap year), suggesting a festive use of local domestic fireplaces and bonfires or perhaps long-range fire smoke from the Netherlands and Belgium where New Year is traditionally celebrated with large public bonfires (Cordell et al., 2016). Further evidence of long-range transport of C_{wood} can be seen at both Chilbolton and Detling sites (Fig. 5E) where greatest mean concentrations arose on broadly easterly winds, consistent with the relative locations of London and continental Europe. This was not evident at the Scottish sites, which are further from the European land mass. A second peak of C_{wood} was measured on the Julian date of 311 at both London and Glasgow sites, coinciding with the Guy Fawkes Night (Fig. 5C).

In contrast to the diurnal concentrations of C_{wood} shown in Fig. 2, the partial dependency plots did not exhibit a morning peak in C_{wood} for the

Table 2
Best RF selected to reproduce C_{wood} concentrations for each site. Slope refers to the RMA slope calculated relating monthly C_{wood} modelled vs monthly C_{wood} observed.

	Model selected	Variables	R^2 hourly	R^2 monthly	Slope
Auchencorth Moss (AUC - rural)	#3	Wind speed Wind direction Temperature Pressure Ceilometer height Trend Julian date Weekday Hour	0.354	0.870	0.859
Chilbolton Observatory (CHO - rural)	#13	Wind speed Wind direction Temperature Trend Julian date Weekday Hour Covid restrictions	0.536	0.941	0.991
Glasgow Townhead (GLA - urban)	#10	Wind speed Wind direction Temperature Relative humidity Trend Julian date Weekday Hour Covid restrictions	0.541	0.917	0.927
Maidstone – Detling (DET - rural)	#5	Wind speed Wind direction Temperature Trend Julian date Weekday Hour	0.287	0.823	0.824
London North Kensington (LNK - urban)	#13	Wind speed Wind direction Temperature Trend Julian date Weekday Hour Covid restrictions	0.627	0.953	0.929

hour variable at both Glasgow Townhead and London North Kensington.

3.5. Linear trends of C_{wood}

Linear trends of the de-weathered time series showed significant downward trends at all sites except Chilbolton Observatory (Table 3; Fig. 6). Downward trends ranged between −0.04 μg m^{-3} $year^{-1}$ (London North Kensington, 2010–2021) and −0.01 μg m^{-3} $year^{-1}$ (at both Glasgow Townhead and Maidstone – Detling; 2014–2021), statistically significant at $p < 0.001$. Expressed as a percentage, the trends ranged between −1.5% $year^{-1}$ (Maidstone – Detling, 2014–2021) and −3.8% $year^{-1}$ (Auchencorth Moss, 2012–2021). Chilbolton Observatory observed a positive trend of 0.006 μg m^{-3} $year^{-1}$ which represented an upward trend of 1% $year^{-1}$ (2015–2021). However, this was not statistically significant.

Trends for 2015–2021 showed faster downard rates at Auchencorth Moss (−5.5% $year^{-1}$), Glasgow Townhead (−2.4% $year^{-1}$) and London North Kensington (−3.8% $year^{-1}$) ($p < 0.001$), compared with the early part of the study, indicating that downward trends were faster from 2015 onwards. Maidstone–Detling showed an upward trend of 0.2% $year^{-1}$ ($p > 0.1$); which was probably due to the increased concentrations observed at the site in 2021 (Fig. 6). Monthly means of observed C_{wood} concentrations for 2015–2021 showed faster trends at the two urban sites (London and Glasgow), at rates of −4.1 and −3.5% $year^{-1}$ for 2015–2021 ($p < 0.05$), compared with trends from de-weathered concentrations (−3.8 and −2.5% $year^{-1}$, respectively). The contrary was observed at Auchencort Moss where downward trends from monthly concentrations were slower (−2.2% $year^{-1}$) than the de-weathered results (−5.5% $year^{-1}$). Both Chilbolton Observatory and Maidstone-Detling in SE England showed positive trends when calculated from monthly concentrations, with faster upward trends, and at the later site this was statistically significant.

4. Discussion

There is no reference method for estimating PM from wood burning in ambient air. Wood smoke tracers such as levoglucosan (Yttri et al., 2014; Yttri et al., 2020); and fine potassium in ambient air (Andreae, 1983; Hsu et al., 2009) have been used sporadically in the UK (Harrison et al., 2012). These methods are based on filter measurements with low time-resolution limiting any linkage to hourly variability and changing weather. Wood burning derived from daily 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. However, C_{wood} derived from the aethalometer were less than those from levoglucosan by 13%. High-time resolved measurements of organic mass fractions can also be used to derive C_{wood} concentrations using the positive matrix factorization (PMF) algorithm. C_{wood} concentrations from PMF using the aerosol mass spectrometer (AMS) at London North Kensington for January 2012–January 2013 (Reyes-Villegas et al., 2016) showed similar temporal dynamics to those observed by the aethalometer method, with greatest concentrations at nights and most noticeably during the evening at the weekends (Supplementary Fig. 6). However, the aethalometer method measured larger concentrations by 69% on average. The smaller C_{wood} concentrations from AMS-PMF may be partly explained by the misrepresentation of biomass burning in the solid fuel organic aerosol (SFOA) factor (Young et al., 2015).

Like other methods to estimate PM from wood burning, the aethalometer method is not free from uncertainties both in terms of specificity and quantification. One of the main uncertainties derives from the assumption only two varying sources of carbonaceous aerosols exist in ambient air. However, this is not the case in many environments where other sources of carbonaceous aerosols (e.g. cooking, secondary organics, etc.) exist (Chen et al., 2022; Young et al., 2015). These uncertainties are therefore transferred to the factors used in regression models of traffic and wood burning absorption against carbonaceous matter (see Eq. 8 in the Supplementary Information) (Herich et al., 2011). However, previous studies showed that C_{wood} calculated by the aethalometer method correlated directly with the apportionment in black carbon fraction. Zotter et al. (2017) showed good correlation between black carbon from wood burning estimated using the aethalometer method and that resolved from carbon isotopic analysis. Another source of uncertainty of the aethalometer method comes from the fixed Ängstrom coefficients assumed for both traffic and wood burning. For instance, the value of α for wood burning has been found to be variable, ranging from 1.4 to 2.2 and it is dependent on the fuel and atmospheric aging (Kirchstetter et al., 2004; Saleh et al., 2013; Zotter et al., 2017). However, our study looked to characterise sources in background locations where the influence of any individual wood smoke source is diluted. There was no evidence that α for wood burning changed over the time and that the majority of hourly α values in our sites were within the assumed range of $\alpha = 0.96$ and $\alpha = 2$ (Supplementary Fig. 2,

Table 3
Linear trends as calculated from monthly C_{wood} concentrations and de-weathered means of C_{wood}. *** significant at $p < 0.001$; * significant at $p < 0.05$; significant at $p < 0.1$; (blank) not statistically significant.

		C_{WOOD} CONCENTRATIONS		DE-WEATHERED C_{WOOD} CONCENTRATIONS			C_{WOOD} CONCENTRATIONS		DE-WEATHERED C_{WOOD} CONCENTRATIONS	
Site	Time span	Trend (μg m^{-3} $year^{-1}$)	Trend (% $year^{-1}$)	Trend (μg m^{-3} $year^{-1}$)	Trend (% $year^{-1}$)	Time span	Trend (μg m^{-3} $year^{-1}$)	Trend (% $year^{-1}$)	Trend (μg m^{-3} $year^{-1}$)	Trend (% $year^{-1}$)
Auchencorth Moss (AUC)	2012–21	−0.004 [-0.010, 0.001]	−2.7 [-5.1, 0.9]	−0.008 [-0.011, −0.005]***	−3.8 [-4.6, −2.7]***	2015–21	−0.003 [-0.014, 0.006]	−2.2 [-7.8, 5.1]	−0.011 [-0.015, −0.009]***	−5.5 [-6.9, −4.3]***
Glasgow Townhead (GLA)	2014–21	−0.009 [-0.018, 0.001]$^{+}$	−2.3 [-4.2, 0.3]$^{+}$	−0.010 [-0.014, −0.005]***	−2.1 [-2.8, −1.2]***	2015–21	−0.014 [-0.026, 0.000]*	−3.5 [-5.9, −0.1]*	−0.012 [-0.015, −0.007]***	−2.5 [-3.1, −1.5]***
Chilbolton Obsevatory (CHO)	2015–21	0.014 [-0.022, 0.052]	2.6 [-3.5, 12.8]	0.006 [-0.009, 0.015]	1.0 [-1.4, 2.5]	2015–21	0.014 [-0.022, 0.052]	2.6 [-3.5, 12.8]	0.006 [-0.009, 0.015]	1.1 [-1.4, 2.6]
Maidstone-Detling (DET)	2014–21	−0.003 [-0.017, 0.013]	−0.5 [-2.5, 2.5]	−0.010 [-0.013, −0.007]***	−1.5 [-1.9, −1.1]***	2015–21	0.034 [0.012, 0.053]***	7.5 [2.3, 13.1]***	0.001 [-0.003, 0.004]	0.2 [-0.5, 0.7]
London North Kensington (LNK)	2010–21	−0.038 [-0.048, −0.028]***	−3.5 [-4.4, −2.8]***	−0.038 [-0.044, −0.033]***	−3.2 [-3.6, −2.9]***	2015–21	−0.032 [-0.053, −0.012]*	−4.1 [-6.3, −1.6]*	−0.034 [-0.046, −0.025]***	−3.8 [-5.0, −3.0]***

Supplementary Fig. 3).

Given the good correlation between C_{wood} derived from the aethalometer and other methods in London we have good confidence in the capability of C_{wood} metric to measure changes in wood burning aerosol over time such as the long-term trend and hourly and seasonal variations. We also have good confidence in the assessment of daily changes and our attribution of wood burning as residential. Despite the uncertainties of the aethalometer method, in the absence of alternative metrics, the length and temporal resolution of the C_{wood} time series in the UK, offers a unique opportunity for policy makers to learn about long-term trends.

Some authors have highlighted that the aethalometer method may suffer from interferences from traffic black carbon (Harrison et al., 2012). This was based on the appearance of a morning peak in the diel profiles from C_{wood} estimates, co-including with the morning peak in traffic. This effect was observed at the urban sites studied here (London and Glasgow) where a peak at 8–9 am was observed, as in Fig. 3. The morning peak coincided with traffic-related pollutants (nitrogen oxides) at the same sites (Supplementary Fig. 4). However, some authors have attributed this peak to a re-lighting of the wood burners early in the mornings or low average dispersion conditions at that time of day. This has been found in studies in New Zealand, Switzerland and early 20th century in London (Grange et al., 2013; Grange et al., 2020; Whipple, 1929). Information about the timing of indoor burning in winter months is not available in the UK. However, the partial dependency plots from the random-forest modelling at both London North Kensington and Glasgow Townhead did not show that the morning peak in C_{wood} (Fig. 5D) suggesting that the morning peak in the diurnal concentrations of C_{wood} is probably related to morning meteorological conditions (i.e. lower temperatures and lower dispersion conditions) rather than a time

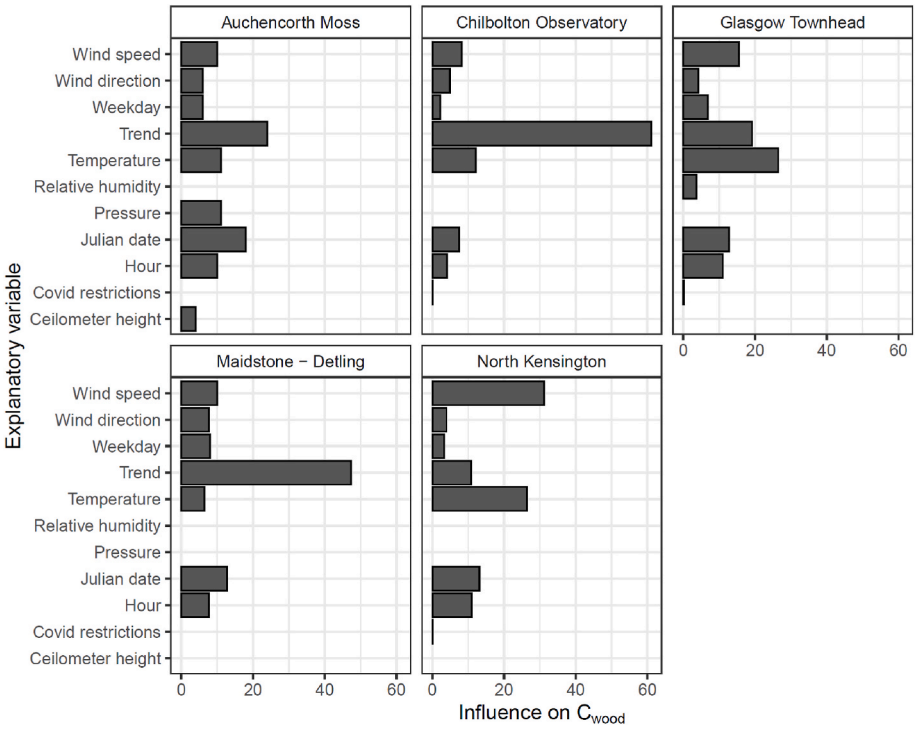

Fig. 4. Influence (in %) on C_{wood} concentrations as explained by each of the explanatory variables for the best performing random-forest model for each monitoring site.

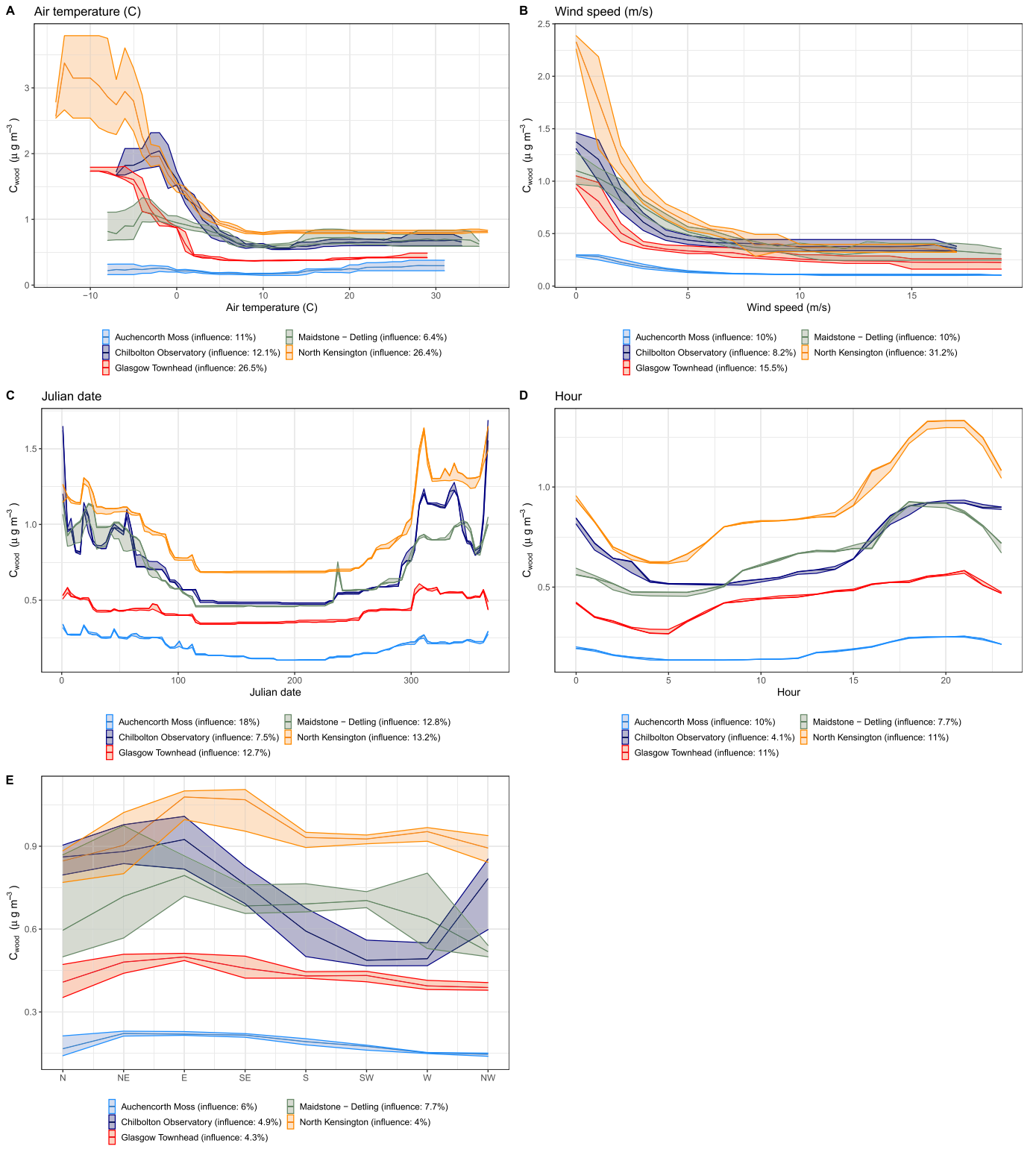

Fig. 5. Partial dependency plots (mean and 95% confidence interval) for each monitoring site for air temperature (A), wind speed (B), Julian date (C), hour (D) and wind direction (E). Influence from each explanatory variable is included and expressed in %.

dependent emission or interference from traffic aerosols. Also, the good correlation between C_{wood} from the aethalometer method in comparisons during 2010 and then again 2012/2013 with other metrics indicate that an interference effect is likely to be small.

Inorganic species such as fine potassium might also contribute to PM mass emissions from wood burning. These would not be quantified in the C_{wood} metric. However, in stove emission tests potassium and other inorganic salts make little contribution to mass emissions. For example, in stove-based tests (Fine et al., 2001), found that potassium and other inorganic salts contributed less than five percent to mass emissions.

Although the absolute concentrations of C_{wood} might differ depending on the methodology used, the C_{wood} metric can be used to determine

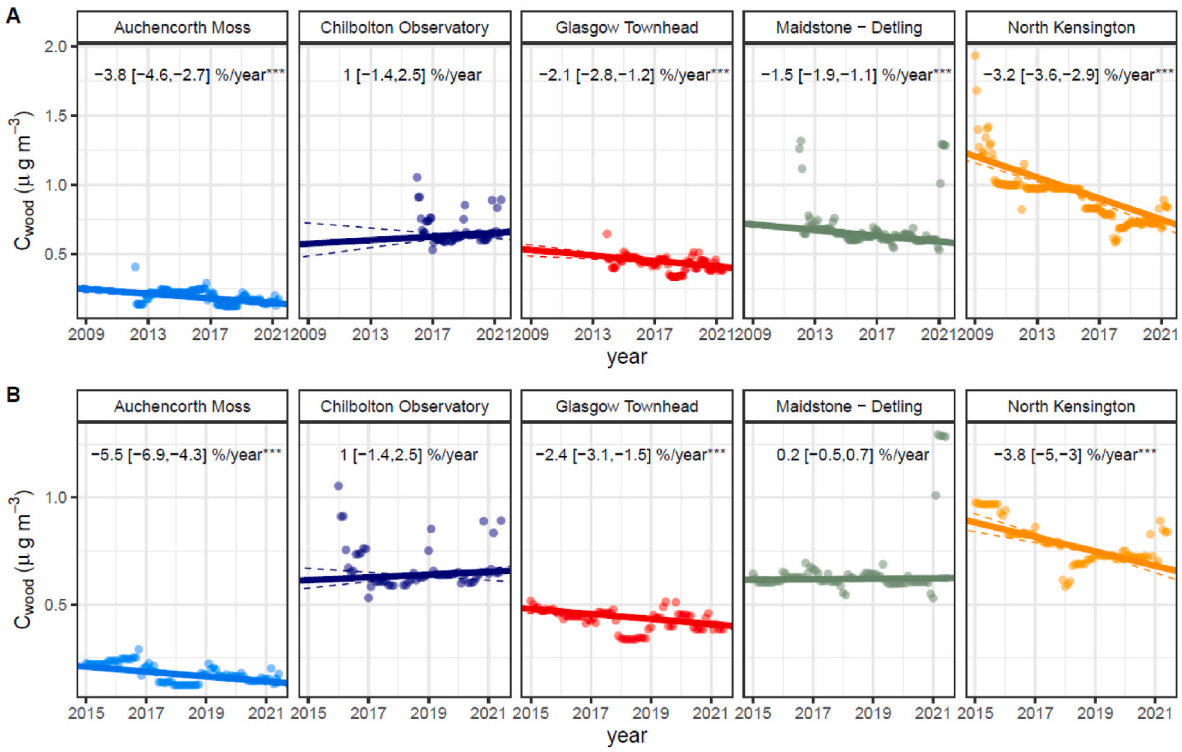

Fig. 6. Linear trends (and 95% confidence interval) in C_{wood} de-weathered concentrations (% year-1). *** significant at $p < 0.001$; (blank) not statistically significant. A. Trends calculated from all available data for each site. B. Trends for the 2015–June 2021 period.

changing temporal patterns in concentrations and emissions. Some authors have pointed out probable increases in biomass burning derived PM_{10} pollution in years to come (Cordell et al., 2016). The NAEI estimated that emissions of $PM_{2.5}$ from domestic wood burning increased by 35% between 2010 and 2020 (GOV.GOV.UK, 2022). However, downward trends in C_{wood} concentrations were observed in four of the five locations studied in the UK; three locations when looking at the common period between 2015 and 2021. Downward trends were also observed elsewhere in Europe. Organic mass fraction observations in a suburban site outside Paris showed downward trends in the wood burning component of organic matter for the period 2012–2018 (Zhang et al., 2019), with a trend of -0.065 µg m^{-3} year^{-1}, although not statistically significant ($p > 0.01$). The black carbon concentrations associated with wood burning (BC_{wb}) from the same site showed also a negative trend and that was although not statistically significant. Measurements of levoglucosan at the remote Birkenes Observatory in southern Norway showed downward trend for the period 2008–2018 at a rate of −2.8% year^{-1}. The lack of any local sources and the regime of air masses transport to the site suggests that the biomass burning tracer was largely explained by continental emissions. The authors concluded that wood burning emissions in continental Europe were therefore declining (Yttri et al., 2021). Grange et al. (2020) also observed significant downward trends of the equivalent black carbon from wood burning at three locations in Switzerland, comprising two urban locations (one background and one traffic) and one rural site. Downward trends ranged between −0.008 µg m^{-3} year^{-1} (2008–2018) and −0.05 µg m^{-3} year^{-1} (2014–2018). The rural Swiss mountain site observed significant upward trends in the period 2013–2018. In the UK, three out of the five sites investigated here showed significant downward trends for the period 2015–2021, at rates ranging between −2.5% year^{-1} and -5.5% year^{-1} (or −0.034 µg m^{-3} year^{-1} and -0.011 µg m^{-3} year^{-1}). These downward trends in the UK were slightly less than those observed in Switzerland and Paris; but in line with the trend found in Norway. However, trends reported here are based on de-weathered concentrations. In our study the raw observations showed faster rates of decline in urban areas than the de-weathered data and it is therefore possible that changes in meteorology may have contributed to the trends from the Swiss and Paris studies.

Local meteorological conditions were expected to have an impact on the ambient concentrations linked to wood burning emissions and therefore might also influence long-term trends. Liakakou et al. (2020) reported large concentrations of black carbon at calm wind conditions and low mixing layer in three monitoring sites in Athens (Greece). This was also observed for the wood burning component of black carbon, with statistically significant negative correlations between wood burning concentrations and boundary layer heights. In our study, we used the partial dependency plots from RF modelling to study the relationship between meteorological variables and C_{wood}. We found that air temperature and wind speed influenced C_{wood} concentrations, especially at the urban sites. Larger concentrations were observed at low and calm conditions, associated with poor dispersion conditions in cold weather.

The timing of emissions (larger domestic wood burning emissions in winter months) and the atmospheric dispersion conditions (lower temperatures and calmer conditions) favoured larger concentrations of C_{wood} in winter. At Auchencorth Moss, winter mean concentration was 10 times greater than the minimum summer concentration. This ratio was almost 6 and 3.5 in the rural sites of Chilbolton Observatory and Maidstone–Detling, respectively. The winter to summer C_{wood} ratios were 3.5 and 3.8 in Glasgow and London, respectively. According to interview-survey data, the incidence of indoor burning was estimated at 1% of homes or below in summer 2018, while the peak in winter in late December/early January was ~6.5% (Defra, 2020). However, outdoor burning was more frequent in summer. Around three quarters of people who did outdoor burning (73%) did so in the summer with a small proportion burning outdoors throughout the year (10%) (Kantar, 2020). Barbeques had a marked seasonality with 85% taking place in summer and more outdoor burners lived in urban areas (82%) compared with rural areas (18%) (Kantar, 2020). This could explain the smaller winter to summer ratio of C_{wood} concentrations in urban areas compared with rural ones in our data.

Downwards trend in C_{wood} were unexpected given the policy context incentivising the use of biomass as source of domestic heating and the estimated increase of 35% projected by the NAEI. This increase was projected from industry surveys and data (BEIS, 2016) which predicted a growth rate in stove installations of 50% each two to three years from 2003 onwards. Data from the Stove Industries Alliance in the UK indicates an increase in the number of sales of wood stoves, peaking in 2014, with an estimate 210,000 new appliances sold (Milligan, personal communication). However, the number of Defra exempt new appliances – those tested and passed by the UK Government's Defra criteria for emission and smoke levels and exempt in control smoke areas - also increased between 2009 and 2015 from 5% of the new appliances in 2009 to 30% in 2015. According to analysis of the English Housing Survey between 2003 and 2016 (Kantar, 2020), primary solid fuel heating systems fell below 1% in England whilst there was a rise in the ownership of secondary solid fuel appliances, driven largely by stove installation.

Although the prevalence of indoor burning was stable over time - 8% of UK homes burnt wood in 2018–19 (Kantar, 2020) compared with 7.5% in 2016 (Waters, 2016) - the proportion of open fires decreased by 16% in Scotland, 13% in England and 12% in London. Previous studies have highlighted the disparity of emissions between open fires and new residential woodstoves with higher combustion efficiencies. Emission tests undertaken in biomass combustion facilities in Portugal found that particle emissions from fireplaces were ~3 times greater than those from a traditional stove, ~12 times greater than an eco-labelled appliance and ~15 times greater than those from a pellet stove (Gianelle et al., 2010). In Libby (Montana, United States) a community where 32% of households used wood as a main heating fuel, a scheme that swapped out old appliances for newer stoves, observed an improvement of the ambient $PM_{2.5}$ levels by 30% the following winter (Noonan et al., 2011). Another conclusion from the Kantar (2020) survey is that households with indoor burners were not using them every week even in winter. The replacement of old fireplaces with new wood burners and a reduction of daily use of wood burners might therefore explain the observed downward trends in C_{wood} in the UK.

Lockdown and restrictions applied to reduce the spread of the Covid-19 had clear impact on the air pollution in many urban areas in the UK (Singh et al., 2022), China (Dai et al., 2021) and around the world (Venter et al., 2020), but the decrease was mainly in those pollutants from traffic emissions. It could be hypothesized that C_{wood} concentrations might increase with lockdown restrictions as citizens were required to spend more time at home; giving greater opportunity for wood burning. In our study, Covid restrictions were introduced and tested as a categorical variable in the RF prediction model. Chilbolton Observatory, Glasgow and London had this variable in the best performing model. The percentage of C_{wood} explained by this variable, over the whole study period, was very little (<0.2%) indicating that the effect of lockdown restrictions had a limited influence on long-term C_{wood} concentrations. However, this is the impact over a multi-year time frame. De-weathered, de-trended and de-seasonalized C_{wood} concentrations were calculated using the RF model at these three locations for lockdown and non-lockdown periods. The ratio of the mean predicted concentration during lockdown over pre-covid times was 1.0 at London North Kensington; and 1.1 at both Chilbolton Observatory and Glasgow Townhead. This indicates that Covid restrictions resulted in a slight increase, of up to 10%, in C_{wood} concentrations.

5. Conclusions

We presented long-term evaluation of airborne particles from wood burning in the UK. Five locations comprising two urban sites (London and Glasgow) and three rural (two in south England and one in Scotland) measured light attenuation properties of particles which were used to calculate C_{wood} concentrations. Time series of C_{wood} concentrations were between six and ten years.

As expected, air pollution from wood burning was greatest in winter and almost absent in summer when residential heating is minimum and atmospheric dispersion is enhanced. Mean PM from wood burning during the heating season (November to March) were greatest in South-East England (1.3 μg m^{-3} in London and 1.1–0.9 μg m^{-3} in the rural areas) compared with Scotland where C_{wood} was ~0.6 μg m^{-3} in Glasgow and ~0.3 μg m^{-3} in the rural environment. Wood burning PM concentrations were greater in evenings everywhere, indicating local residential combustion and low and calmer wind conditions leading to larger concentrations. C_{wood} concentrations were greater at weekends in London and Glasgow. This was probably due to more decorative or leisure wood burning at these times.

Compared with rural areas, there were greater concentrations in London and Glasgow during the summer months. This was consistent with survey data that found greater outdoor burning in urban areas than rural ones (Kantar, 2020).

Trends in C_{wood} concentration, once the influence of weather variables and the timing of emissions were removed, were downward at the two urban sites for the period 2015–2021; at a rate of −2.5% $year^{-1}$ (Glasgow) and −3.8% $year^{-1}$ (London). The rural site in Scotland also showed a downward trend at a rate of −5.5% $year^{-1}$. The two rural sites in south-east England showed upward but not statistically significant trends. However, one of these sites, Maidstone – Detling, observed significant downward trends for the period 2014–2021 but an upward trend for 2015–2021 indicating a possible change of trend direction towards the end of the time series. This contrasts with the UK NAEI that estimated an increase of 35% in $PM_{2.5}$ emissions from domestic wood burning between 2010 and 2020.

We based our C_{wood} concentrations on the aethalometer method. For greater confidence in trend analysis and quantification, on-going future assessments would benefit from other collocated measurements of tracers of wood burning emissions (e.g. levoglucosan, fine potassium, organic mass fraction). These supplementary measurements could take place as short-term winter campaigns at Defra black carbon network sites. Future research needs include methods to separate ambient PM from different types of solid fuels and wood; between smokeless fuels and wood; and also, from novel waste biomass fuels.

Trend analysis of C_{wood} concentrations could be repeated in future years to investigate the impacts of new policies to reduce air pollution from wood burning in the UK. These new polices include a new ban on sales of some types of solid fuel (house coal and wet wood bought in small quantities) in May 2021 (Defra, 2021) and the eco design requirements for new wood stoves from January 2022 (HETAS, 2022).

Author statement

A Font: Conceptualization; Methodology; Software; Formal analysis; Investigation; Data Curation; Writing - Original Draft, Visualization, **K Ciupek**: Validation; Investigation; Resources; Data Curation; Writing - Review & Editing, **D Butterfield**: Validation; Investigation; Resources; Data Curation; Writing - Review & Editing, **G W Fuller**: Conceptualization; Methodology; Software; Formal analysis; Investigation; Writing - Review & Editing; Supervision; Project administration; Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.120105.

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