The contribution of indoor domestic solid fuel burning to Scotland’s air pollution

Final Report to Scottish Government

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- Several methods exist to isolate and quantify the contribution of solid fuel burning to particulate matter, focussing on the carbonaceous emissions (black carbon and organic particulate mass), but there is no standardised approach. All approaches have uncertainties and none quantify the secondary inorganic (and organic) aerosol components that derive from the reaction of the gaseous emissions associated with solid fuel burning.
- The analysis of the long-term timeseries of aethalometer measurements at the two Scottish sites of Defra's Black Carbon Network (Glasgow Townhead and Auchencorth Moss) was extended to quantify solid fuel burning PM for the years 2022 to 2024.
- At Glasgow Townhead a weak downward trend in solid fuel burning related $PM_{2.5}$ concentrations continued, with annual average concentrations (2020-24) of 0.28 $\mu g\ m^{-3}$ and winter concentrations of 0.43 $\mu g\ m^{-3}$. At Auchencorth Moss concentrations were smaller at 0.12 $\mu g\ m^{-3}$ (annual) and 0.17 $\mu g\ m^{-3}$ (winter).
- In addition, two winter measurement campaigns were carried out to apply two independent state-of-the-art methods (aerosol mass spectrometer and aethalometer) to quantify the solid fuel burning PM, contrasting a smoke-control area (Edinburgh) and a village setting (Charlestown, Fife). Both periods were characterised by unusually high wind speeds, leading to reduced concentrations during the respective measurement periods. Averaged over these campaigns, Glasgow Townhead reported concentrations which were 26% lower than typical winter concentrations during the Edinburgh campaign and 16% lower during the Charlestown campaign.
- At both campaign sites PM_1 concentrations were dominated by secondary aerosol components. In Edinburgh secondary inorganic aerosol (SIA) components (ammonium, nitrate, sulfate) accounted for 42% of the PM_1 mass and secondary organic aerosol (SOA) for another 22%. At Charlestown, SIA accounted for 49% and SOA also for 22%. Gas-phase emission control of NO_x, NH_3, SO_2 and VOCs) at national and international scales remains key to reducing PM concentrations.
- It is estimated that in Edinburgh primary emissions from solid fuel burning accounted for 8% of PM_1 during the winter campaign, traffic for 19% and (restaurant) cooking for 8%. The cooking contribution is less seasonal than solid fuel burning and likely makes the larger contribution at the annual average. This highlights a largely unregulated and policed source in urban settings.
- At Charlestown solid fuel burning accounted for 17% and traffic for 12%. Here coal and biomass (wood) burning could be distinguished, with wood accounting for 2/3 of the solid fuel contribution.
- Based on the aethalometer approach applied at all four sites, during the first campaign concentrations of solid fuel burning primary $PM_{2.5}$ components were 0.19 $\mu g\ m^{-3}$ in Edinburgh, 0.32 $\mu g\ m^{-3}$ in Glasgow and 0.12 $\mu g\ m^{-3}$ at Auchencorth.

During the second campaign, concentrations averaged 0.24 µg m^{-3} in Charlestown, 0.36 µg m^{-3} in Glasgow and 0.18 µg m^{-3} at Auchencorth.

Emissions

- Based on new fuel usage statistics and new emission factors, revised Scottish estimates have been developed for the domestic indoor solid fuel burning sector, for $PM_{2.5}$, NOx, CO, VOCs, SO_2, BC and condensable PM.
- New fuel usage statistics suggest that solid fuel use in Scotland is larger than previously reported, by a factor of 2 for wood logs, 5.9 for other wood products, 2.8 for house coal and 7.7 for other coal products, including manufactured solid fuels.
- New emission factors show small differences between house coal and between manufactured low-sulfur and high-sulfur solid fuels (MSF). This would imply more limited potential for lowering emissions by replacing house coal with MSF including smokeless fuels. These new emission factors were measured with a unified state-of-the-art methodology and relevant for UK conditions, but necessarily cover only a small number of appliances and fuels. The representativeness of these is difficult to judge and the associated uncertainty is difficult to quantify.
- These emission factors also point to modern stoves (clearSkies level 2 or higher; Ecodesign) not having lower emissions than somewhat older stoves, but the sample size of stove models is small (one or two of each category) and uncertainties are likely significant. Emissions from pellet stoves and Blue-Angel ecolabelled stoves were much smaller.
- The same emissions database for wood burning shows large variations in the $PM_{2.5}$ emission factors as a function of moisture content, with seasoned wood showing lower emission factors than pre-dried wood. There is some uncertainty whether the moisture content used for seasoned wood is representative for the damp Scottish climate, which could lead to an underestimating of emissions.
- Overall, the revision of the emission factors from wood burning has resulted in a reduction in associated $PM_{2.5}$ emissions from this fuel type, despite an increase in fuel consumption allocated to Scotland.
- By contrast, the new $PM_{2.5}$ emission estimate of emissions from coal, coal products and MSF has increased by a factor of 7.3 compared to a previous study (Masey et al., 2023) and a factor of 4.4 compared to the NAEI2021, as Scottish fuel amounts have been revised upwards and the MSF emission factors also. This in particular allocates more emissions to smoke control areas. The uncertainty in the allocation of coal/coal-based fuels is larger than that of wood quantity.
- At 1567 t yr^{-1}, the new estimate of Scottish $PM_{2.5}$ emissions from domestic solid fuel burning is 52% larger than that of Masey et al. (2023) and 22% larger than NAEI2021.
- The new estimate of Scottish NOx emissions from domestic solid fuel burning of 600 t yr^{-1} exceeds the NAEI2021 by a factor of 2.8, and the SO_2 emissions estimate of 1990 t yr^{-1} exceeds the NAEI2021 by 67%.

- Despite improvements in the data situation, the large changes in fuel amounts attributed to Scotland and of the emission factors from one study to another is indicative of the large uncertainties that still persist for estimating emissions from this sector.
- In addition, there are some concerns around the generalisation of the data and its application for Scotland, including whether the laboratory burn-cycles are representative of real-world behaviour, whether beech is the dominant wood type for all of Scotland, and whether the moisture content assumed for seasoned wood is representative for Scottish conditions and whether "super emitters" burning e.g. treated and painted timber can be correctly represented by this approach.

Modelling of concentrations

- An atmospheric chemistry and transport model (ACTM) was applied with the new emissions to calculate hourly concentrations for Scotland at 1 km x 1 km resolution for the meteorological year of 2023 (and additionally Jan-Mar 2024 for comparison with the village measurement campaign). In contrast to previous studies, this model also simulates the formation of secondary $PM_{2.5}$ components from gaseous compounds emitted from solid fuel burning.
- Annual average $PM_{2.5}$ concentration enhancement due to Scottish solid fuel burning were estimated to be 0.086 $\mu g\ m^{-3}$ averaged over the Scottish landmass, with a population weighted mean concentration (PWMC) enhancement of 0.36 $\mu g\ m^{-3}$. Winter values are 0.14 and 0.61 $\mu g\ m^{-3}$, respectively. The largest local values were found in the Central Belt between Bathgate and Livingston dominated by MSF emissions, with the coal contribution peaking in Midlothian (Dalkeith/ Newtongrange) and wood contribution peaking in Fort William. These details are uncertain, however, due to uncertainties in the geographical allocation of fuel quantities.
- Non-Scottish UK domestic burning (dominated by import from England and lesser extent Northern Ireland) is estimated to make an additional PWMC contribution to $PM_{2.5}$ of 0.09 $\mu g\ m^{-3}$ at the population-weighted average mean, with 1 km^2 maximum contribution of 0.39 $\mu g\ m^{-3}$ (winter only: 0.60 $\mu g\ m^{-3}$). Hotspots were identified near Newcastle and Carlisle. More than half of this non-Scottish contribution is due to secondary components not considered in most modelling estimates.
- Model results suggest that almost a quarter (24%) of the $PM_{2.5}$ enhancement due to Scottish sources is due to secondary compounds (mainly ammonium sulfate and ammonium nitrate and associated particle-bound water). This component would be missed by simpler modelling approaches and also not attributed to solid fuel burning when interpreting measurements. This component relies on the presence of ammonia and most of it would also be controllable through agricultural ammonia control.
- For NO_2, the other compound of concern for human health, the PWMC contribution from all solid fuel burning sources is very small: 0.062 $\mu g\ m^{-3}$ (1.0%)

at the annual mean and 0.102 µg m^{-3} (1.2%) in winter, with a maximum contribution of 0.92 µg m^{-3}.

Lessons learnt from the comparison of measured and modelled estimates

- Overall, there was good agreement between modelled and measured winter concentrations of primary solid fuel $PM_{2.5}$ at Glasgow Townhead, Auchencorth Moss and Charlestown. The measurements detect additional contributions in summer, likely from outdoor burning, muirburn and (global) wildfires.
- At the Edinburgh site the modelled estimate significantly exceeds the measured primary PM concentration associated with solid fuel burning. The emissions of coal and MSF appear to be overestimated for Edinburgh, whilst they appear to be more appropriate for Glasgow. It is unclear at this stage whether this is due to the distribution algorithm within Scotland or possibly also indicates an overestimation of coal-based fuels to Scotland as a whole. In either case, shifting emissions away from Edinburgh would likely lower the population weighted mean concentration somewhat.
- The modelling suggests that secondary $PM_{2.5}$ formed from gases emitted from the burning of solid fuels accounts for another 1/3 of the $PM_{2.5}$ which is not accounted for in the measurements or in previous estimates of the impacts of domestic solid fuel burning.

1 Context and Scope

Domestic solid fuel burning is increasingly recognised as a key contributor to air pollution and is now estimated to be the single largest emission source of fine particulate matter ($PM_{2.5}$) in the UK National Atmospheric Emissions Inventory (NAEI), where it accounts for about 25% of the primary $PM_{2.5}$ emissions. $PM_{2.5}$ is the primary driver of ambient air pollution health impacts in the UK. It is associated with respiratory and cardiovascular disease and has recently also been linked to cognitive impairment. Whilst $PM_{2.5}$ affects the health of everyone, particularly susceptible groups include those with pre-existing conditions such as asthma, pregnant women and the young. There is evidence that $PM_{2.5}$ emitted directly from solid fuel burning is particularly toxic as it contains polyaromatic hydrocarbons (PAHs), heavy metals and black carbon (BC). In addition to emitting particulates directly (primary $PM_{2.5}$), solid fuel burning emits gaseous compounds such as carbon monoxide (CO), nitrogen oxides (NOx), sulphur dioxide (SO_2) and volatile organic compounds (VOCs; also often referred to as non-methane hydrocarbons, NMHCs). NOx leads to the production of nitrogen dioxide (NO_2) which also has direct health impacts. In addition, oxidation of NOx and VOCs in the atmosphere contributes to the chemical formation of additional compounds in the particle phase which further contribute to the mass concentration of $PM_{2.5}$ (secondary $PM_{2.5}$). Secondary aerosol components are formed at larger spatial scales than those that govern the distribution of primary $PM_{2.5}$. Finally, NOx and VOCs are also involved in the formation of tropospheric ozone, which has further impacts on human health. It also damages ecosystems and lowers crop yields.

Emissions and $PM_{2.5}$ contributions from domestic solid fuel burning are particularly uncertain and difficult to estimate. Emission factors (emitted amount per quantity of fuel or heat generated) can vary by two orders of magnitude between fuel types, fuel conditions (e.g. moisture content), appliances and burning conditions. There is inherent uncertainty due to the upscaling from a few controlled emission factor measurements to country-wide emissions, and fuel amounts and types are uncertain, together with their spatial distribution.

Measurement quantification of the contribution of domestic solid fuel burning to ambient $PM_{2.5}$ is also challenging, for a number of reasons: concentrations are highly variable in space and time, and measurement methods are expensive, still have some uncertainty and cannot capture the full impact on air quality. Solid fuel emissions include a particularly large fraction of low volatility VOCs (LVOCs) and intermediate volatility organic compounds (IVOCs) which readily partition into the aerosol phase very rapidly during dilution and cooling. Uniquely to solid fuel burning, national emission estimates should include this "condensable particulate matter" which makes measurement protocols more complicated and allows for additional variability between studies. There is active international debate on how to best deal with these for emission reporting requirements and how to treat the condensable fraction in the models. Some of the resulting aerosol components can re-evaporate into the gas phase.

Through the Cleaner Air for Scotland 2 (CAfS2) Strategy (Scottish Government, 2021), the Scottish Government is committed to actions to reduce this source. This study was commissioned through the Rural and Environment Science and Analytical Services (RESAS) programme to reduce uncertainty around the contribution of domestic solid fuel burning in Scotland and to provide scientific evidence on where to target interventions. The objectives of the study were:

O1. To improve the emission inventory for domestic and commercial biomass burning for Scotland.
O2. To quantify the contribution of biomass burning aerosol to $PM_{2.5}$ in understudied settings and analyse existing data to provide a measurement database to inform the emission inventory work and for model assessment.
O3. To assess the contribution of biomass burning to $PM_{2.5}$ concentrations across Scotland through high resolution (1 km^2) modelling
O4. To assess the effect of $PM_{2.5}$ concentrations due to biomass burning on health outcomes.

Scottish Government later decided to separate Objective 4 from this project and to consider options to take it forward as a separate research piece.

This work was not conducted in isolation. Another Scottish study was commissioned in parallel to look specifically at the contribution of coal and the most polluting manufactured solid fuels to Scotland's $PM_{2.5}$, and to inform the impact of regulatory action for health benefits and businesses (Masey et al., 2023). That study also quantified the contribution from wood burning and this estimate is referred to as Masey2023 throughout this report.

In addition, Defra also commissioned two significant projects on domestic biomass burning: a UK-wide fuel survey was undertaken in 2022/23 by Ipsos and Air Quality Consultants (AQC) to provide insights into domestic burning practices in the UK (Defra, 2024a). This was a rerun and extension to an earlier fuel survey conducted in 2018/19 (Kantar, 2020), providing new data for Scotland. In addition, Defra commissioned the Emission Factors for Domestic Solid Fuels (EFDSF) project to make standardised measurements of emission factors for key pollutants from a range of fuels and appliances relevant to the UK (Allan et al., 2024a, 2024b, 2025). Much of the emission inventory improvement work has therefore focused on making use of these new sources of information to develop a best-estimate emission inventory for Scotland. This has somewhat delayed the delivery of the project, but at the same time it has generated much added value to the study and ensured that these emissions will remain state-of-the-art beyond the next release of the NAEI which will also make use of the new statistics, but with different, likely simplified, assumptions on the spatial distribution for Scotland.

2 Measurements

2.1 Methodology

2.1.1 Overview of existing methodologies

The contribution of solid fuel burning to $PM_{2.5}$ consists of several primary and secondary chemical components. Primary components include black carbon (soot) and a wide range of organic compounds, with small contributions from trace metals and some inorganic components (chloride, sulfur, nitrogen compounds). The organic compounds may chemically evolve as the emissions age and some may evaporate. The gases emitted during combustion can also contribute to secondary aerosol production: NOx and SO_2 can be oxidised to HNO_3 and H_2SO_4, respectively, and then combine with ammonia (NH_3), mainly from agricultural sources, to form secondary ammonium aerosol components. Standard measurement approaches are incapable of separating these contributions from ammonium salts derived from other sources. Volatile organic compounds (VOCs) can be oxidised to less volatile components, some of which eventually partition into the particle phase, i.e. condense onto existing particles and thus contribute to $PM_{2.5}$. At the point of combustion, solid fuel emissions include a particularly large fraction of low volatility VOCs which rapidly form particulate matter upon cooling and dilution of the flue gas to form "condensable particulate matter" which should be included in the national reportable primary $PM_{2.5}$ estimates. Some of the resulting aerosol components could potentially later re-evaporate into the gas phase, however. Solid fuel burning also emits more volatile VOCs which are included in the inventories as VOCs and some of which may also contribute to $PM_{2.5}$ in the form of secondary organic aerosol (SOA) by being converted to ever less volatile compounds through atmospheric oxidation.

Measurement approaches aiming to quantify $PM_{2.5}$ from biomass burning emissions should ideally attempt to estimate the total amount of primary and secondary organic $PM_{2.5}$ associated with the emissions, as well as the black carbon component. Current approaches include aerosol mass spectrometry, multi-wavelength aethalometry, molecular trace methods and isotope measurements. Of these, the first two were applied during this project to quantify the biomass burning contribution at contrasting Scottish locations, and these are described in more detail in the following sections, whilst the others are briefly covered for completeness.

2.1.2 Aerosol mass spectrometry and chemical speciation by positive matrix factorisation

The High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS, Aerodyne Research Inc.) provides real-time measurements of the chemical composition (nitrate, sulphate, ammonium, chloride and organic aerosol) of the submicron non-refractory species ($NR\text{-}PM_1$) and has been described in detail elsewhere (DeCarlo et al., 2006; Drewnick et al., 2005). Briefly, the HR-ToF-AMS samples particles through an aerodynamic lens into a vacuum and then impacts the focused particle beam on a heated tungsten surface (~600 °C). The obtained vapours are ionized by standard 70 eV electron impact ionization and the ions are analysed using time-of-flight mass spectrometry. The HR-ToF-AMS data were recorded at a time resolution of 5 min for the two field campaigns during which it was deployed. The instrument was calibrated with 350 nm mono-disperse ammonium nitrate particles during each campaign. Organic aerosol components are measured as total organic mass rather than OC, i.e. including the associated contributions from oxygen, hydrogen and nitrogen. Whilst the inorganic components can be quantified by compound, the organic aerosol is made up of hundreds or even thousands of different chemical compounds which the AMS does not quantify individually. Instead, it provides mass spectra of the chemical composition of the total organic aerosol as a function of time. Positive Matrix Factorization (PMF) (Paatero and Tapper, 1994) is a commonly used method to investigate the organic aerosol (OA) sources from this mass spectral information (Crippa et al., 2014). Briefly, time series of organic mass spectra, arranged as a matrix **X,** are represented as a linear combination of several factors and expressed by the matrix product of factor contributions **G** and factor profiles **F.** The fraction that the model cannot explain is represented by the residual matrix **E**.

$$\boldsymbol{X} = \boldsymbol{GF} + \boldsymbol{E} \tag{1}$$

Several contributors to OA can be identified through PMF analysis. Depending on the dataset these may include hydrocarbon-like (HOA) associated mainly with tailpipe emissions, biomass burning (BBOA), cooking (COA), coal combustion (CCOA) among the primary OA, and oxygenated organic aerosol (OOA) which represents secondary OA. The analysis here was carried out using the PMF Evaluation Tool (PET, v3.08) using the PMF2 algorithm in robust mode (Ulbrich et al., 2009). PMF relies on differences in the temporal pattern in the contribution from different sources and cannot distinguish two sources which have the same time signature; it would lump them into a combined source factor. For the quantification provide here, a collection efficiency (CE) value between 0.5 and 1, generally composition dependent (Middlebrook et al., 2012), was applied to the AMS output, following current best practice, together with a relative ionization efficiency (RIE), which varies according to how efficiently compounds are ionized inside the instrument. For organics a RIE value of 1.4 is generally used. However, as (fresh) COA has been found to be collected and ionised very efficiently inside the AMS, and for this component an RIE of 2 and a CE of 1 were used, effectively downscaling this component.

The AMS used here is limited to sub-micron particles and only detects non-refractory chemical components (i.e. those that flash-evaporate at 600 °C), therefore referred to as NR-PM_1. In reality, there is little difference between PM_1 and $PM_{2.5}$ when it comes to solid fuel burning emissions and the components measured by the AMS. The AMS does not detect black carbon.

2.1.3 Use of aethalometers for source apportionment of black carbon and total carbonaceous aerosol

2.1.3.1 Black carbon source apportionment

Another approach is to quantify the biomass burning related aerosol from the light absorbing qualities of the PM. The Magee Scientific aethalometer model AE33 measures the light attenuation by particles collected onto a filter tape at seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm). An absorption coefficient (*b*) can be obtained from the light attenuation using a multiple scattering coefficient appropriate for the tape (Drinovec et al., 2015). According to the Beer-Lambert’s law, the absorption of light b_λ is dependent on the wavelength (λ):

$$b_\lambda \propto \lambda^{-\alpha} \tag{2}$$

where α is the absorption Ångström exponent (AAE). The Ångström exponent describes the wavelength dependency of the absorption coefficient and it is used to obtain information about the predominant aerosol source (Favez et al., 2009; Harrison et al., 2012a). Particles from different combustion sources show different wavelength dependencies of light absorption, especially in the near ultraviolet (N-UV) and lower visible range. This can be used for source apportionment studies (Sandradewi et al., 2008). The Ångström coefficient obtained from the absorption coefficient measured by the aethalometer in the N-UV (470 nm) and N-IR (950 nm) channels, can provide information on fossil fuel traffic and wood burning sources:

$$\alpha = \frac{-\log\left(\frac{b_{abs470}}{b_{abs950}}\right)}{\log\left(\frac{470}{950}\right)} \tag{3}$$

Black carbon aerosols derived from diesel traffic sources are characterised by α ~ 1, whilst particles derived from solid fuel burning have a large content of organic material and absorb more radiation in the UV part of the spectrum. This is reflected in higher Ångström coefficients (Helin et al., 2021). However, the value of α depends on the efficiency of combustion. For wood smoke a value of 2 is often assumed (Favez et al., 2010; Sandradewi et al., 2008; Zotter et al., 2017), for coal combustion values can range from 1 to 3 (Blanco-Alegre et al., 2022). Different values for wood burning (α_{wb}) and traffic (α_{tr}) are used in literature. In reality, they depend on the fuel burned and if more than two tightly correlated BC sources are present, the values can vary widely and would ideally need to be evaluated with a reference method. In fact, as domestic burning involves coal combustion, the term “solid fuel” is often used to describe this type of source. Furthermore, Zotter et al. (2017) showed that the AAE values can also vary depending on the aethalometer wavelengths used in the calculation and recommended using $\alpha_{wb} = 1.68$ and $\alpha_{tr} = 0.9$ for 470-950 nm wavelengths. Savadkoohi et al. (2025) proposed a practical method to determine site-specific AAE values for solid fuel and traffic sources in the absence of ancillary data. In their study, these values represented the 1st and the 99th percentiles, respectively, of the AAE, obtained using all the aethalometer wavelengths. Following their approach, in the present study we calculated AAE in two different ways: first, it was calculated as the ratio of the logarithms of the absorption coefficient as in Eq. (3), and secondly it was obtained from the log-log fit of the absorption coefficients as a function of the wavelengths (370 nm to 950 nm) (Savadkoohi et al., 2025; Tobler et al., 2021). A stringent filter ($r^2 > 0.99$) was applied to the AAE obtained from the fit, and values smaller than 0.7 were discarded as not representative of any ambient source. Although site-specific AAE values are discussed in the next sections, for consistency with a previous UK-wide study on the quantification of wood burning by Font et al. (2022), here we adopted the same values they used: $\alpha_{wb} = 2$ and $\alpha_{tr} = 0.96$ unless stated otherwise.

The source apportionment model developed by Sandradewi et al. (2008) assumes that only two sources contribute to the aerosol and the total absorption at a given wavelength $b_{abs}(\lambda)$ can be expressed as the sum of a component due to wood burning $b_{abs}(\lambda)_{wb}$ and one due to fossil fuel $b_{abs}(\lambda)_{tr}$:

$$b_{abs}(\lambda) = b_{abs}(\lambda)_{wb} + b_{abs}(\lambda)_{tr} \quad (4)$$

From the Beer-Lambert's law two equations relating the absorption coefficient (b_{abs}), the wavelengths, and the α for conditions of pure traffic and pure wood burning can be derived:

$$b_{abs}(470)_{wb}/b_{abs}(950)_{wb} = (470/950)^{-\alpha wb} \quad (5)$$

$$b_{abs}(470)_{tr}/b_{abs}(950)_{tr} = (470/950)^{-\alpha tr} \quad (6)$$

The aethalometer model can be used to quantify the contribution of wood burning and traffic to total mass equivalent black carbon (eBC_{tot}), which can be expressed as the sum of eBC_{wb} and eBC_{tr}. These two components can be derived using the Mass Absorption Cross-section (MAC = 7.77 $m^2 g^{-1}$ at 880 nm) as recommended by Zotter et al. (2017):

$$eBC_{tot} = eBC_{wb} + eBC_{tr}, \quad (7)$$

where:

$$eBC_{tot} = \frac{b_{abs}(880)}{MAC_{880}} \quad (8)$$

$$eBC_{wb} = \frac{b_{abs}(950)_{wb}}{b_{abs}(950)} * eBC_{tot} \quad (9)$$

$$eBC_{tr} = (1 - \frac{b_{abs}(950)_{wb}}{b_{abs}(950)}) * eBC_{tot} \quad (10)$$

Here $b_{abs}(\lambda)_{tr}$ and $b_{abs}(\lambda)_{wb}$ can be derived from Eqs. (4) - (6). Although it is a practical tool to obtain the solid fuel combustion contribution to PM, the aethalometer model carries large uncertainties introduced by the choice of the α values for fossil fuel and solid fuels, which depend on the wavelengths used for their determination (Fuller et al., 2014; Harrison et al., 2013), and by the type of sources and their changes during measurements (Tobler et al., 2021).

2.1.3.2 Wood-burning contribution to PM using aethalometer data

The aethalometer model can also be used to quantify the contribution of wood burning and traffic to the total carbonaceous material (CM), defined as the sum of organic matter (OM) and black carbon (BC). CM can be expressed as a multi-linear regression of $b_{abs}(880)_{tr}$ and $b_{abs}(370)_{wb}$ and coefficients C_1,C_2,C_3 (Sandradewi et al., 2008):

$$CM = OM + BC = C_{tr} + C_{wb} \tag{11}$$

$$CM = C_1 * b_{abs}(880)_{tr} + C_2 * b_{abs}(370)_{wb} + C_3 \tag{12}$$

The intercept C_3 represents a constant background concentration of non-combustion carbonaceous material (Favez et al., 2010; Küpper et al., 2018). In the present study we focused on the calculation of C_{wb}, later referred to as c_{wood}. As most UK cities and rural areas have several sources of organic aerosol rather than just traffic and wood-burning, here we adopted a value for C_2 (41800 µg m^{-2}) as used in Font et al. (2022), and obtained as the average of values from studies where traffic and wood-burning were the only OM sources (Fuller et al., 2014; Sciare et al., 2011). For continuity with the aforementioned study, MAC values typically implemented for the 2-wavelenghts aethalometers were used here (16.6 m^2 g^{-1} for 880 nm and 39.5 m^2 g^{-1} for 370 nm) for the calculation of c_{wood}.

It is important to notice that in addition to the uncertainty associated with the aethalometer model, the c_{wood} calculation can depend on the type of measurements used to determine the C_1, C_2 coefficients.

2.1.4 Molecular tracer method: levoglucosan and potassium

Rather than deriving the total mass from solid fuel burning sources from the bulk properties (mass spectrum or light-absorbing characteristics) of that contribution, the molecular tracer method quantifies individual chemical compounds that are characteristic for a source and scales these up to provide the total source contribution to $PM_{2.5}$ according to the "typical" prevalence of that tracer in the aerosol. It relies on this contribution to be consistent across datasets and stable over time and the tracer to be unique for that source. Levoglucosan ($C_6H_{10}O_5$) is an organic compound formed from the pyrolysis of carbohydrates and cellulose, and it is often considered a suitable chemical tracer for wood smoke. The most common method for the detection and quantification of levoglucosan involves the use of gas chromatography-mass spectrometry (GC-MS) after the extraction via solvent of samples collected on quartz filters (Crilley et al., 2015; Harrison et al., 2012b). Other techniques used include thermal desorption gas chromatography (TD-GC-MS), high-performance liquid chromatography (HPLC) and ion chromatography coupled with pulsed amperometric detection (HPAEC-PAD) (Bhattarai et al., 2019).

Because levoglucosan is relatively volatile concentrations can decrease during long-range transport. Information on the type of wood smoke source can be derived from the ratio of levoglucosan and OC or potassium (K^+), distinguishing between regional and long-range transport sources but also between traditional domestic fires (fireplaces, woodstoves, etc.) and more efficient modern appliances (Crilley et al., 2015).

Several scaling factors have been derived to quantify the wood smoke contribution to PM directly using the measured levoglucosan mass concentration (Harrison et al., 2012a; Küpper et al., 2018; Puxbaum et al., 2004; Schmidl et al., 2008). Although widely used, these scaling factors might be strongly related to the local setting and might not be applicable to all environments. Due to the volatility of levoglucosan the scaling factor changes as wood smoke ages. Similarly, K^+ can be used as a wood smoke tracer after its concentration is corrected for the minor contributions of sea salt and soil (Harrison et al., 2012b). Because the scaling factors are uncertain and measurements of K^+ and levoglucosan are usually of low time resolution, this quantification approach for wood smoke was not used in the current study.

2.1.5 Isotope methods

The most common isotope method to quantify the relative contribution of biogenic and fossil fuels to carbonaceous aerosols uses the radioisotope of carbon (^{14}C). While in fossil fuels ^{14}C has completely decayed during long geological processes, living material is in equilibrium with CO_2 in the atmosphere containing a known abundance of ^{14}C. By analysing the ratio of $^{14}C/^{12}C$ in PM filter samples it is thus possible to distinguish between fossil and contemporary carbon via accelerator mass spectrometry technique (Heal, 2014). Data from the ^{14}C analyses in combination with measured concentrations of OC and EC from the same samples provide information for source apportionment of carbonaceous PM, with the separation of anthropogenic and biogenic sources (Crilley et al., 2015; Gelencsér et al., 2007; Szidat et al., 2004). The younger (biogenic) carbon reflects the sum of biomass burning and other biogenic carbon, such as biogenic secondary organic aerosol (BSOA) that derives from the oxidation of biogenic volatile organic compounds emitted by plants, such as monoterpenes and isoprene. It does not include fossil carbon stored in coal products. As such this approach is applied mainly in situations where wood burning dominates solid fuel burning, e.g. in winter in areas where coal burning is uncommon. For this reason, isotopic measurements were not considered suitable for this study. As with molecular tracer methods, this approach only quantifies a component of the burning derived particulates, in this case the carbon element, and a scaling factor has to be applied to approximately scale this up to the full organic mass.

2.2 Description of the measurement sites and meteorology

Measurements were made at two contrasting sites for one burning season each, comparing a smoke-control area (Edinburgh) and a village setting (Charlestown, Fife).

For the first campaign, measurements were made at an urban site at the University of Edinburgh campus at Infirmary Street, Edinburgh, (3°11'4.01"W, 55°56'54.81"N) between 15th December 2022 and 6th March 2023. The site is located in the city centre, 150 m away from a busy road with many commercial activities and restaurants, and about 400 m northwest of the urban background AURN measurement site at St Leonard's.

The second field campaign was carried out at Charlestown (3°29'58.02"W, 56° 2'14.74"N), a small residential village about 20 km northwest of Edinburgh, across the Firth of Forth. Measurements at this village site were made between 5th January 2024 and 26th March 2024. The Grangemouth refinery is situated 12 km west of the site and the towns of Dunfermline and Rosyth 5 km to the NE and E, respectively. The instrumentation setup was the same as the one used in Edinburgh. Power cuts at the site led to gaps in the measured data (~9% of the data). In both cases the measurement height was 3 m.

2.2.1 Ancillary datasets

Hourly PM_{10}, $PM_{2.5}$ and NO_x concentrations were extracted from UK-Air for one of the Defra Automatic Urban and Rural Network (AURN) stations. As part of this national network NO_X ($NO + NO_2$) is measured by chemiluminescence and fortnightly calibrations enable the traceability of measurements to national metrological standards. All instruments are subject to twice yearly audit tests by the National Physical Laboratory or Ricardo AEA.

Meteorological parameters were measured with a metstation (Vaisala Weather Transmitter WXT530). Additional measurements of total mass of PM_1, $PM_{2.5}$ and PM_{10} and other trace gases (NO_2, CH_4, CO) were made using an optical particle counter (Dustdecoder 11-D, GRIMM Aerosol Technik), similar in principle to the Fidas® instrument now deployed in the UK networks, and a Multi-Gas-Analyser (MGA, MIRO analytical AG), respectively, at the same height as the main measurements.

2.3 Measurement Results - Winter 2022-2023 campaign: Edinburgh (smoke control area)

2.3.1 Overview

An overview of the hourly PM chemical composition measured by the AMS at Infirmary Street alongside some meteorological variables are shown in Figure 2.1.

The average wind speed at measurement height (3 m; sheltered by surrounding buildings) was 7 m/s and the predominant wind direction was from the southwest. The windspeed during the campaign was unusually high, with storm Otto impacting the UK on 17 Feb. An average temperature of 5 ºC was recorded with a minimum of -1.7 ºC and a maximum of 11 ºC. The hourly total NR-PM_1 concentration measured by the AMS was mostly lower than 5 µg m^{-3}, except for some sporadic events that coincided with a change in wind direction, decrease in wind speed, or drop in temperature. The most evident pollution event was on 14/02/2023 with concentrations of up to 17.8 µg m^{-3}, associated with a change in wind direction and a drop in the wind speed.

The diurnal cycles of three inorganic components (NO_3^-, SO_4^{2-}, NH_4^+) measured by the AMS showed similar diurnal patterns with an increase in concentration at night and a decrease as the morning progressed (Figure 2.1). Such behaviour is generally associated with the daytime increase in boundary layer height that provides more dilution in the middle of the day, with a consequent reduction of concentrations. In contrast, the organic aerosol and the chloride showed two distinct peaks, one in the morning and the other in the afternoon/early evening, which suggests a strong influence of local emission sources on the concentration of these components (Figure 2.2).

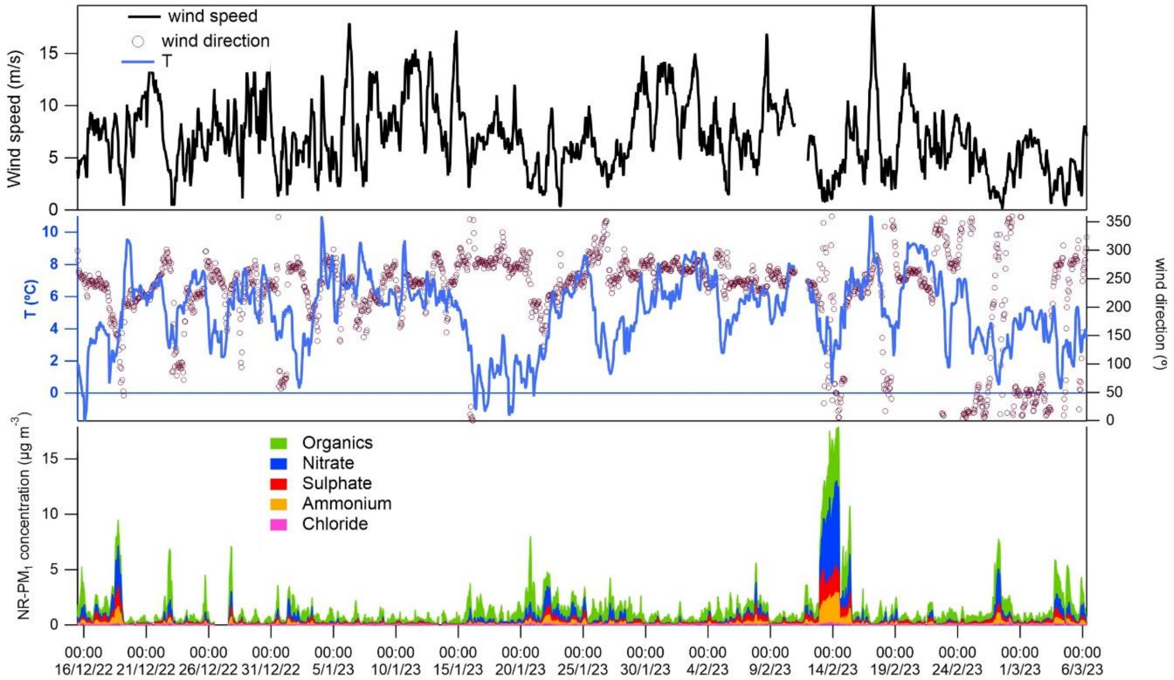


Figure 2.1. Hourly wind speed, wind direction, temperature and NR-PM1 chemical composition as measured at Edinburgh Infirmary Street by the met station and AMS.

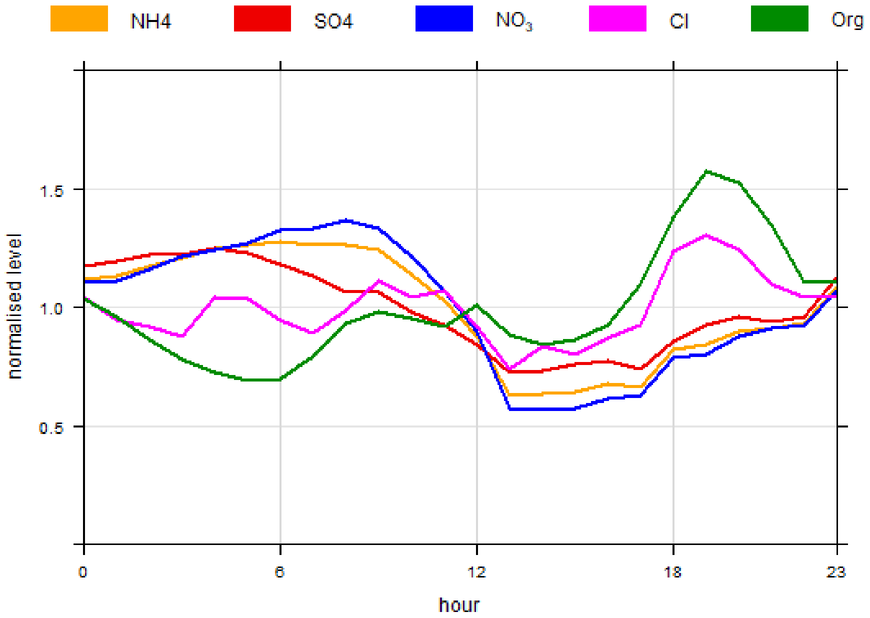


Figure 2.2. Diurnal patterns for the inorganic (NO_3^-, SO_4^-, NH_4^+, Cl^-) and organic aerosol measured with the AMS at Edinburgh Infirmary Street.

2.3.2 PMF source apportionment

The PMF analysis on the organic aerosol measured by AMS provided a 5-factor solution, with three primary sources identified: traffic (hydrocarbon-like organic aerosol, HOA), cooking (COA) (mostly from commercial activities), and domestic biomass burning (BBOA). The remaining two factors showed mass spectra characteristic of secondary OA: a relatively freshly formed, less oxidised OA (LO-OOA) and a more oxidised OA (MO-OOA), generally associated with long-range transport (Figure 2.3). The traffic factor showed a morning peak between 8 and 9 AM, related to the morning rush hour, but did not show a second peak that would be indicative of an evening rush hour. The cooking factor showed two peaks at mealtimes, as expected, and the biomass burning factor showed an evening peak, consistent with the use of fireplaces and stoves for domestic heating. The MO-OOA concentration evening increase and morning decrease followed the same pattern as the inorganic components, whereas LO-OOA did not show a clear diurnal pattern (Figure 2.4).

The use of correlation between the OA components identified by PMF and other known tracers associated with specific sources is a widely used method to support the PMF results. Figure 2.5 shows the correlation matrix of the measured compounds indicating their grouping into clusters. Each cluster contains variables that have similarities in their pattern of temporal variation. Chloride and BBOA appeared in the same cluster, and they correlated well with each other, with chloride showing the highest correlation coefficient with BBOA. The highest correlation coefficient for BBOA was with BC, suggesting that wood burning contributed to BC.

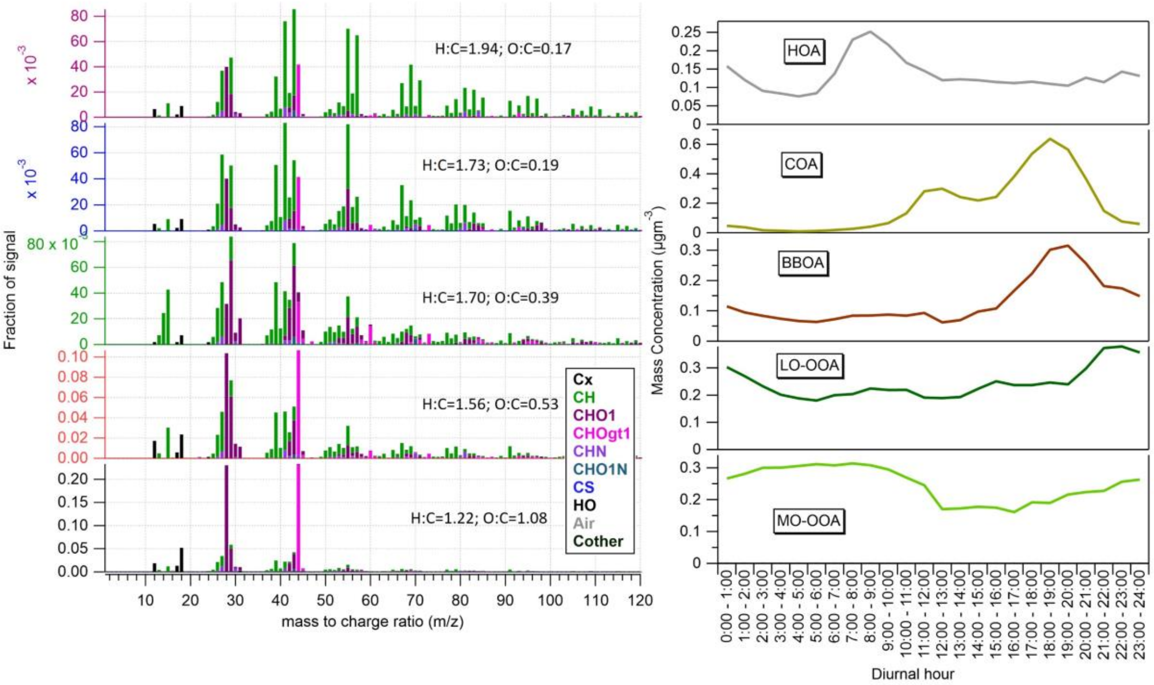


Figure 2.3 PMF results for the measurement campaign at Edinburgh Infirmary Street. Left: spectra of the 5-factor solution found for the composition of the organic PM. Right: diurnal cycles of the 5-factor concentrations.

Particularly high correlation was found between the secondary aerosol components NH_4^+, NO_3^-, SO_4^{2-} and MO-OOA indicating a shared source or control. They all tend to peak when polluted air is advected, e.g. from Continental Europe and/or the rest of the UK, and are low in clean Atlantic air masses.

The PMF analysis did not isolate a coal factor in Edinburgh. At this urban background site the measurement averaged over many burners and it is possible that the BBOA signal contains a contribution from the burning of smokeless manufactured solid fuel and other (unauthorised) coal products which showed the same temporal pattern as wood smoke and therefore could not be distinguished. The mass spectral signature suggests that the BBOA factor was dominated by the contribution from wood burning, with a large peak at m/z 60 associated with levoglucosan, but it also includes some peaks that are characteristic for coal burning such as m/z 77. The high correlation of BBOA with chloride (Cl^-) could also indicate a contribution of coal burning to this PMF factor, with coal being a key source of Cl^- emissions depending on the Cl^- content of the coal. Alternatively, it could be explained by some household waste being burnt at the same time, which contains chlorinated compounds such as PVC. Overall, Cl^- concentrations are very low, however, accounting for 2% of measured PM_1 mass. HOA best correlated with traffic signature compounds like BC and NO_x, as expected, but was also related to CO and CH_4. Wind direction did not correlate with anything overall, while wind speed was slightly anticorrelated with some of the compounds associated with combustion (BC, NO_x, BBOA, CH_4) and local sources. This is expected, as decreasing wind speeds lead to slower dispersion of local near-ground sources. MO-OOA was strongly correlated with inorganic OA suggesting a long-transport nature by association. The LO-OOA factor did not show a correlation with any other measured compound, perhaps indicating that it could represent a slightly aged aerosol coming from a variety of sources in the city.

The diurnal pattern of traffic tracers (CO and NO_x) confirmed that the evening peak was less prominent than the morning one, in agreement with the HOA factor, except for Fridays when all the compounds showed an evening peak (Figure 2.4).

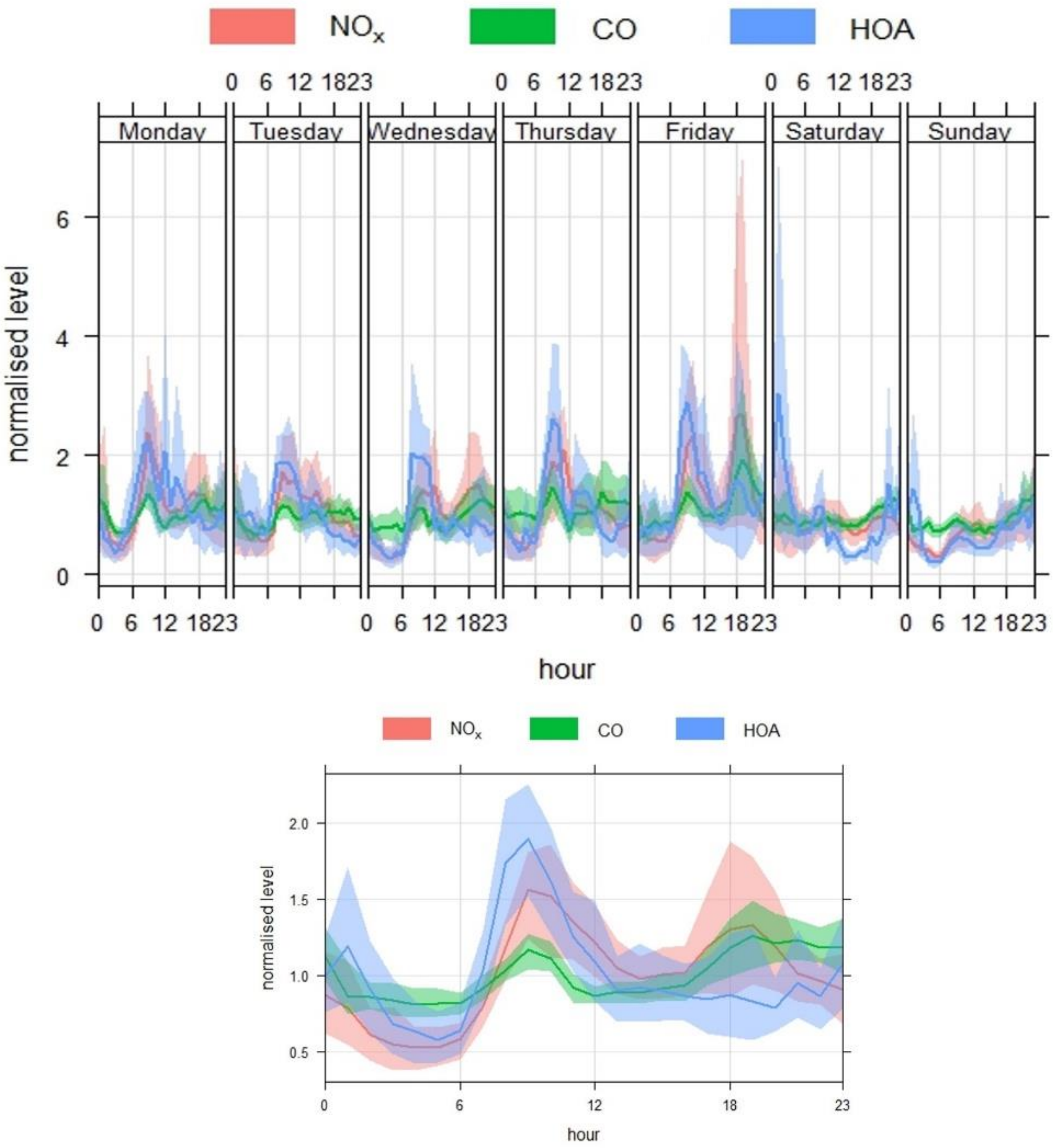
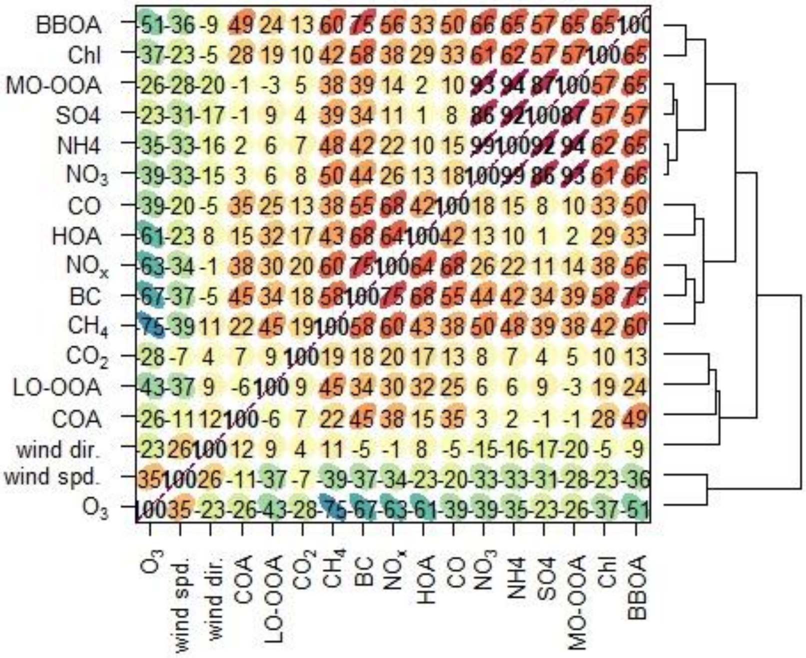


Figure 2.4. Diurnal cycle of NO_x, CO, HOA calculated for each day of the week (top) and over the entire period (bottom) measured at Edinburgh Infirmary Street. Concentrations are normalised for comparison**.**



	O_3	wind spd.	wind dir.	COA	LO-OOA	CO_2	CH_4	BC	NO_x	HOA	CO	NO_3	NH4	SO4	MO-OOA	Chl	BBOA
BBOA	-51	-36	-9	49	24	13	60	75	56	33	50	66	65	57	65	65	100
Chl	-37	-23	-5	28	19	10	42	58	38	29	33	61	62	57	57	100	65
MO-OOA	-26	-28	-20	-1	-3	5	38	39	14	2	10	93	94	87	100	57	65
SO4	-23	-31	-17	-1	9	4	39	34	11	1	8	86	92	100	87	57	57
NH4	-35	-33	-16	2	6	7	48	42	22	10	15	99	100	92	94	62	65
NO_3	-39	-33	-15	3	6	8	50	44	26	13	18	100	99	86	93	61	66
CO	-39	-20	-5	35	25	13	38	55	68	42	100	18	15	8	10	33	50
HOA	-61	-23	8	15	32	17	43	68	64	100	42	13	10	1	2	29	33
NO_x	-63	-34	-1	38	30	20	60	75	100	64	68	26	22	11	14	38	56
BC	-67	-37	-5	45	34	18	58	100	75	68	55	44	42	34	39	58	75
CH_4	-75	-39	11	22	45	19	100	58	60	43	38	50	48	39	38	42	60
CO_2	-28	-7	4	7	9	100	19	18	20	17	13	8	7	4	5	10	13
LO-OOA	-43	-37	9	-6	100	9	45	34	30	32	25	6	6	9	-3	19	24
COA	-26	-11	12	100	-6	7	22	45	38	15	35	3	2	-1	-1	28	49
wind dir.	-23	26	100	12	9	4	11	-5	-1	8	-5	-15	-16	-17	-20	-5	-9
wind spd.	35	100	26	-11	-37	-7	-39	-37	-34	-23	-20	-33	-33	-31	-28	-23	-36
O_3	100	35	-23	-26	-43	-28	-75	-67	-63	-61	-39	-39	-35	-23	-26	-37	-51

Figure 2.5. Correlation matrix of all the measurements at Infirmary Street. CO and NO_x were obtained from the AURN site. O_3, CO_2, CH_4 were measured by MIRO. NO_3^-, SO_4^{2-}, NH_4^+, Cl^- and OA components were measured by AMS and BC by AE33. Wind speed and direction were measured with a Vaisala met station. The correlation is coded in three ways: the number (perfect correlation is 100), the elliptic shape (perfect positive correlation is a line with a 45º slope, zero correlation is a circle), and the colour (positive correlation is red, zero correlation is yellow and negative correlation is blue).

2.3.3 Black carbon source apportionment

The aethalometer model assumes that only two sources contribute to BC (wood burning and traffic). However, this is likely not completely true in Edinburgh, where the solid fuel burned for domestic purposes is expected to include coal or coal products and where some BC may also be associated with cooking activities. The AAE was calculated here in the two different ways described in Section 2.1.3: the distribution obtained by the fitting approach is slightly narrower than the AAE derived from the two channels (Figure 2.6). The two source-specific values from Font et al. (2022) are shown in red in Figure 2.6 for comparison. As the range between 0.96 and 2 includes most of the AAE values in the distribution, this could indicate that the values were a reasonable choice for this site. In contrast, applying the percentile approach to the Edinburgh site provided lower values, at 0.74 and 1.51 for traffic and solid fuel, respectively. Although smaller than AAE figures commonly used for these sources, they agree with results from other European urban sites (Savadkoohi et al., 2025). In their study, Savadkoohi et al. (2025) suggested obtaining the traffic (or ”liquid fuel”) AAE coefficient from summer data to avoid interference with other combustion sources, however, this was not possible for the field campaigns in this project.

Applying the aethalometer model to apportion eBC emission sources (traffic and wood burning) using the "Font" AAE values, the results indicated that, on average, 16% of the eBC was attributed to wood burning (eBC_{wb}), while the remainder was traffic-related (eBC_{tr}). If using the site-specific AAE values, eBC_{wb} and eBC_{tr} would contribute equally to total eBC, similar to the ratios observed for other urban cities (Savadkoohi et al., 2025). The values recommended by Zotter et al. (2017) would split the contribution 30% to solid fuels and 70% to traffic. Similarly, using 0.74 for traffic and 2 for solid fuel, that correspond to a "mid-point", in line with Charlestown (see Section 2.4.3), the split would be 25% to solid fuels and 75% to traffic. Average concentrations for eBC_{wb} and eBC_{tr} using the "Font", the site- specific and the "mid-point" values resulted 0.05 µg m^{-3} and 0.29 µg m^{-3}, 0.17 µg m^{-3} and 0.17 µg m^{-3}, 0.08 µg m^{-3} and 0.26 µg m^{-3}, respectively).The c_{wood} concentration calculated as in Section 2.1.3.2 in Edinburgh was 0.19 µg m^{-3} (representing 4% of PM$_{2.5}$ and 2% of PM$_{10}$). Applying the AAE site-specific values c_{wood} was 0.41 µg m^{-3} and with $\alpha_{wb} = 2$ and $\alpha_{tr} = 0.74$ the calculation was 0.3 µg m^{-3}.

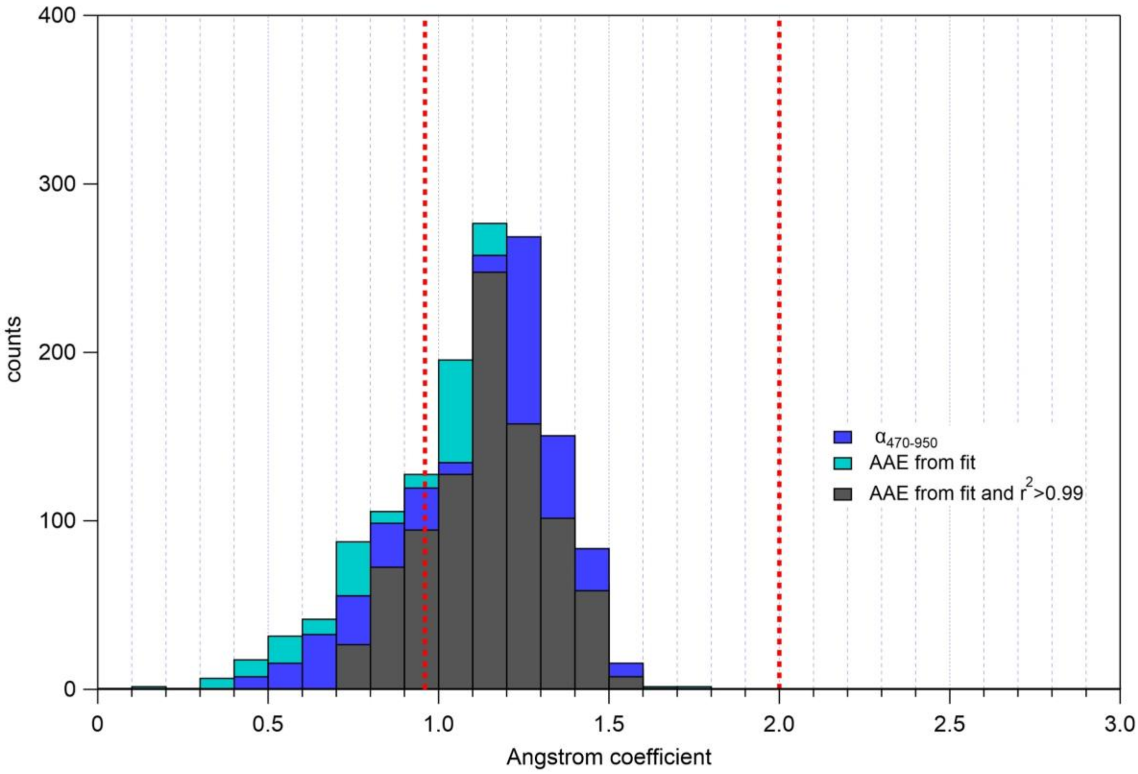


Figure 2.6. The absorption Ångström exponent (AAE) distribution derived for the Edinburgh Infirmary Street site: AAE calculated from the ratio of the 470 nm and 950 nm channels (blue), from the log-log fit of the absorption coefficient as a function of wavelength (green), and with the additional application of a filter (r^2>0.99) (grey). The red lines represent the α values used for traffic and wood smoke.

While eBC_{wb}, peaked in the evening, between 18:00 and 20:00, eBC_{tr} showed its maximum increase in the morning, between 07:00 and 09:00, aligning with the PMF factors associated with the same sources (Figure 2.7). A correlation plot between BBOA and the non-brown carbon part of c_{wood} (i.e. c_{wood} - eBC_{wb}) showed good agreement between the two variables, with the best R^2 obtained when calculations were done using the "mid-point" AAE values (Figure 2.9). The data also reflected a behavioural pattern of the population, which tends to use domestic burning appliances predominantly during the weekend (Fri-Mon) rather than throughout the rest of the week, consistent with published measurements for London (Font et al., 2022). This is consistent with solid fuel predominantly being used as a secondary, supplementary heating source in Edinburgh, or for ambience. In contrast, the traffic pattern exhibited smaller peaks for HOA and eBC_{tr} on Saturdays and Sundays compared to weekdays (Figure 2.7).

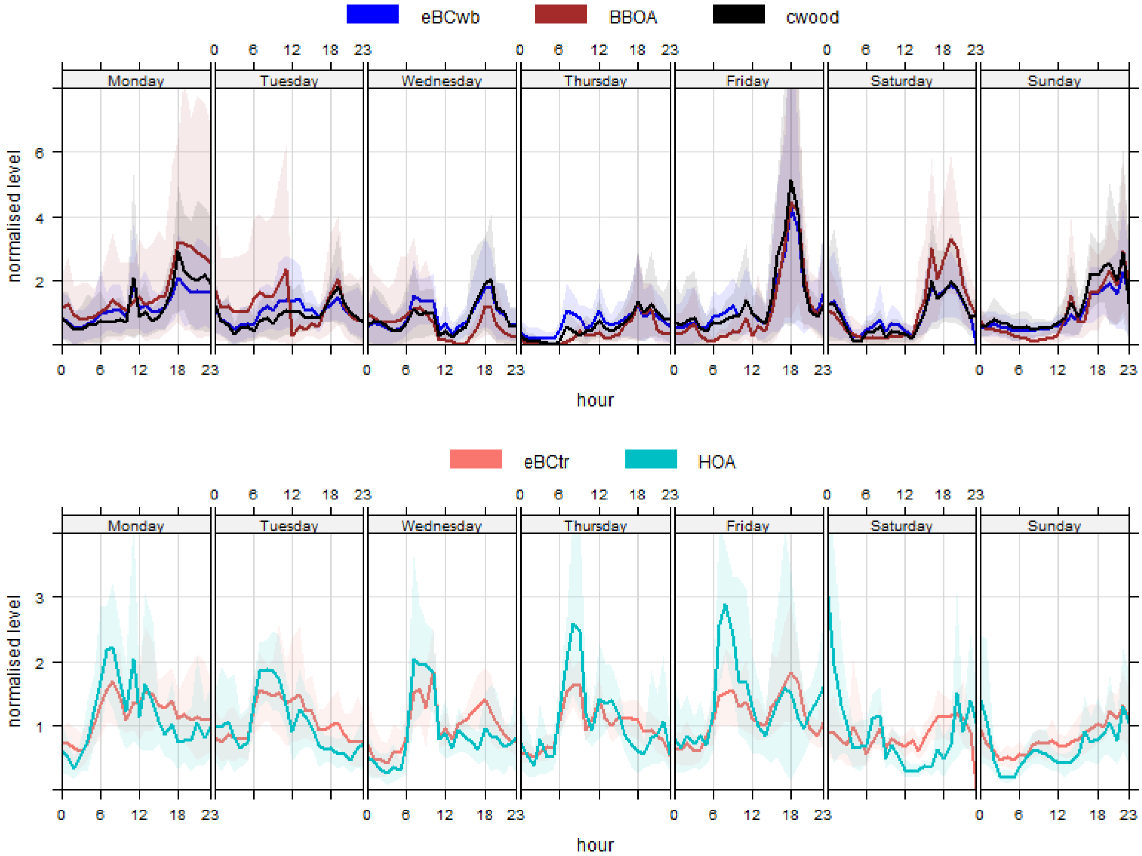


Figure 2.7. Diurnal patterns for each day of the week of concentration for the domestic burning sources (top) and the traffic component (bottom).

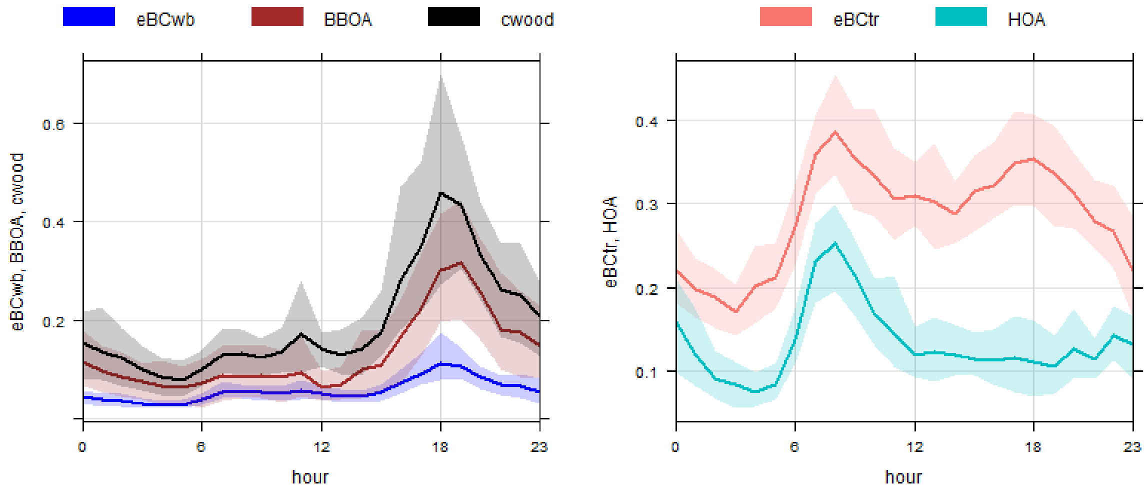


Figure 2.8. Diurnal cycles for the domestic burning (left) and traffic contributions (right) estimated with the aethalometer model and PMF between 13/01/23 and 06/03/2023.

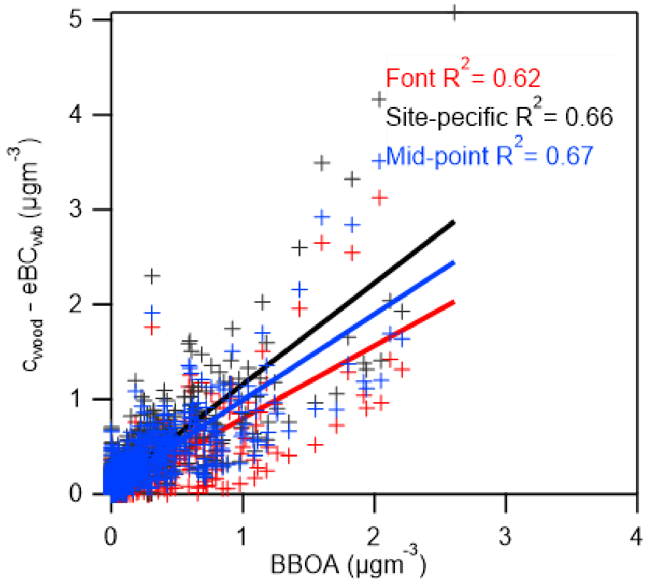


Figure 2.9 Correlation plot between c_{wood} - eBC_{wb} and BBOA when three different sets of values are used for α_{tr} and α_{wb} (Font, site-specific and mid-point).

2.3.4 Spatial analysis of sources

The polar plot of mean concentrations showed that domestic burning sources made the largest contribution at low wind speeds, whereas the traffic source contributed at all conditions and was spread through the city centre (Figure 2.10). This may in part reflect that solid fuel burning tends to coincide with times of low wind speed, e.g. during the evening / night and calm, cold conditions. The conditional probability function (CPF) was used in polar plots to investigate whether specific sources could be located. Here, CPF was defined as the ratio of samples with concentrations larger than the 90th percentile and associated with a specific wind direction and wind speed (Figure 2.11). With this approach, again the largest traffic sources seemed to be in the city centre, whereas the domestic burning sources had some probability to be scattered around the city, with the largest source of BBOA occurring at low wind speed, or coming from the more residential area southwest of the measurement at higher wind speed (Figure 2.11).

The same approach to investigating the spatial variation of cooking aerosol emissions (Figure 2.12) confirmed that the main sources were located near the measurement location, where many restaurants and takeaways can be found. COA is commonly thought to originate from restaurant kitchens with high occurrence of deep frying, rather than residential cooking.

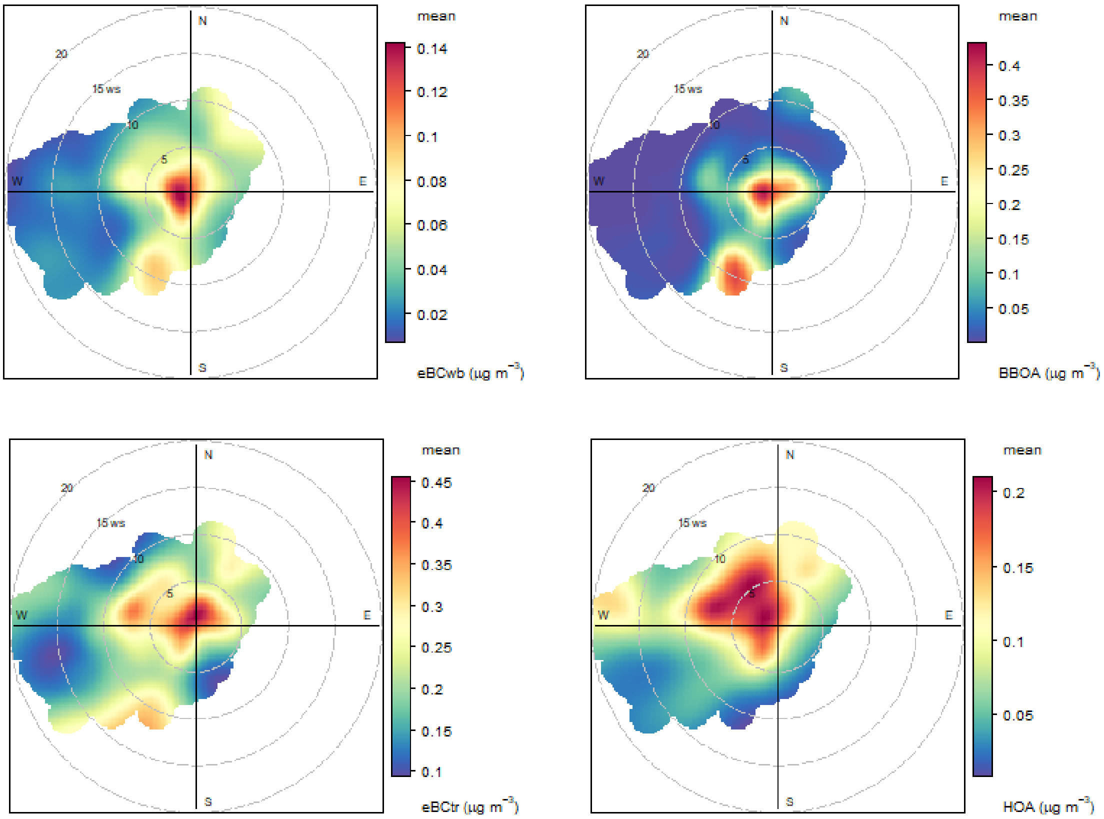
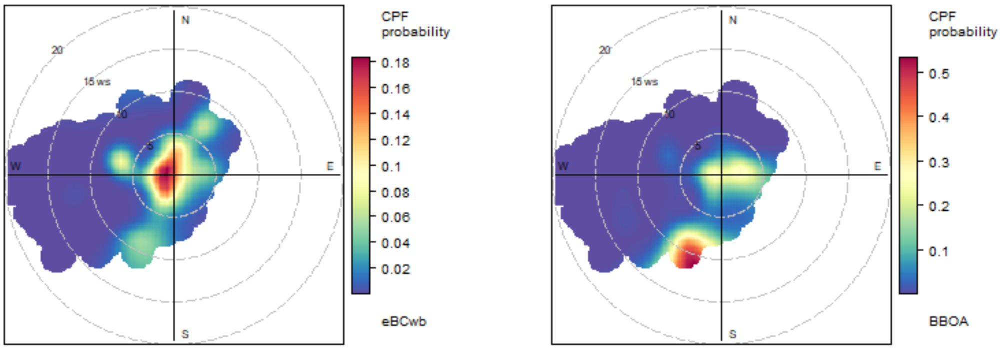
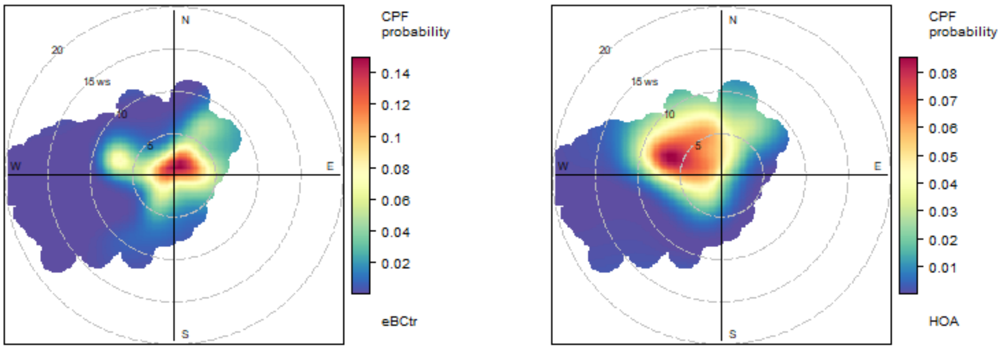


Figure 2.10. Mean concentration variation with wind speed and wind direction for the domestic burning (top) and traffic sources (bottom). Wind speed is in m s^{-1} along the radial axis.



CPF at the 90th percentile (=0.15)

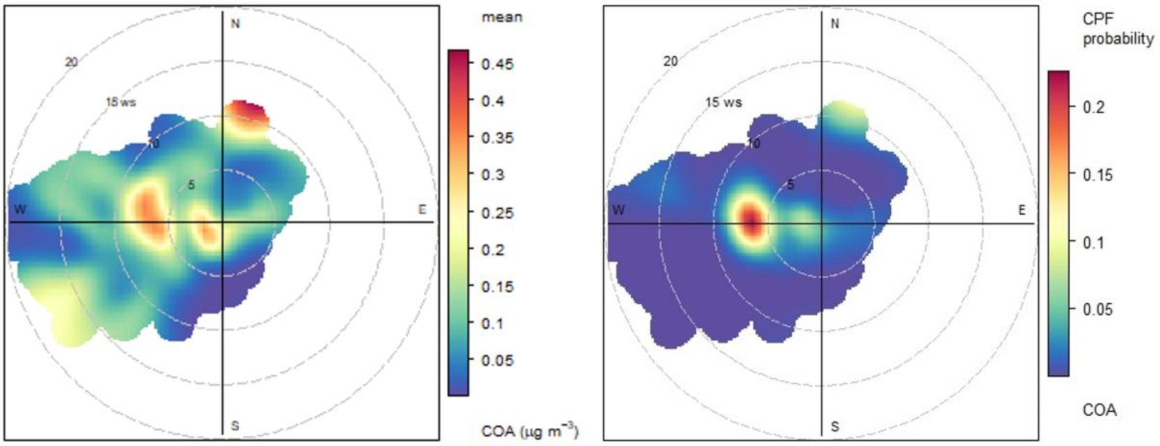
CPF at the 90th percentile (=0.39)



CPF at the 90th percentile (=0.51)

CPF at the 90th percentile (=0.3)

Figure 2.11. CPF variation with wind speed and wind direction for the domestic burning (top) and traffic sources (bottom). Wind speed is in m s^{-1}.



CPF at the 90th percentile (=0.48)

Figure 2.12. Mean concentration (left) and CPF (right) variation with wind speed and direction for the cooking factor COA. Wind speed is in m s^{-1}.

2.3.5 PM mass composition

The average PM_1 mass concentration measured at Edinburgh Infirmary Street by the GRIMM optical instrument was 2.16 µg m^{-3}, in good agreement with the sum of the non-refractory components measured by the AMS plus the BC from the aethalometer (2.18 µg m^{-3}) (Table 2.1). Mass loadings of $PM_{2.5}$ and PM_{10} were significantly larger than the PM_1 mass, at 5.18 and 10.96 µg m^{-3}, respectively, with a large contribution from sea salt associated with the high wind speeds.

Figure 2.13 shows the diurnal pattern of the PM_1 mass concentration and the chemical components determined with the AMS and the aethalometer. Overall, the primary sources (emitting directly to the atmosphere) contributed 36% of the total PM_1 mass, with traffic and domestic burning contributing 19% and 9%, respectively, and cooking contributing 8%. The inorganic component was the largest part of secondary aerosols, amounting to 42% of the PM_1 mass, and the secondary organic aerosols (SOA) added up to 22% of the PM_1 mass (Figure 2.13). It is possible that a fraction of the SOA components is due to solid fuel burning, either from the oxidation of the VOCs emitted or via volatilisation of some primary particulates into gas-phase compounds which could partition back into the particle phase through chemical processing but then have the mass spectral signature of SOA rather than BBOA. Like other measurement approaches, AMS cannot identify the origin of secondary compounds.

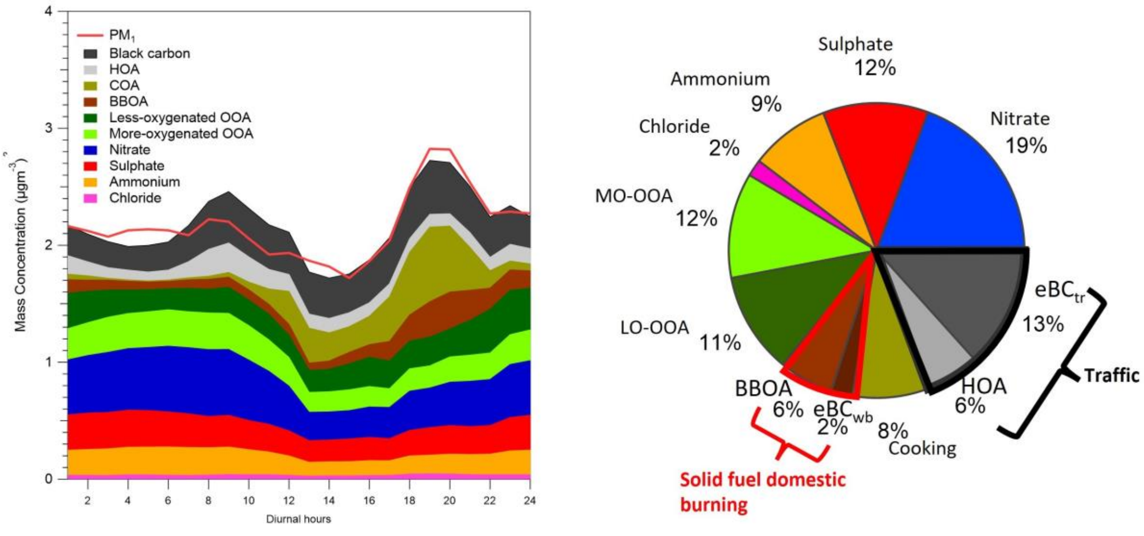


Figure 2.13. PM_1 mass composition: diurnal cycle with stacked components (left); pie chart with quantification (%) of each component contribution (right).

Table 2.1 Summary of the mean concentrations measured during the campaign in Edinburgh between 13/01/2023 and 06/03/2023.

PM_1	Mean ± stdev ($\mu g\ m^{-3}$)	N points
NO_3^-	0.42 ± 1.05	1241
SO_4^{2-}	0.25 ± 0.37	1241
NH_4^+	0.19 ± 0.40	1241
Cl^-	0.04 ± 0.04	1241
eBC_{wb}	0.05 ± 0.09	1241
eBC_{tr}	0.29 ± 0.20	1242
MO-OOA	0.25 ± 0.51	1228
LO-OOA	0.24 ± 0.17	1228
BBOA	0.13 ± 0.28	1228
COA	0.18 ± 0.28	1228
HOA	0.13 ± 0.15	1228
Total PM_1	**2.18 ± 1.39**	
PM_1 GRIMM	*2.16 ± 2.52*	1233
$PM_{2.5}$ GRIMM	*5.18± 3.77*	1233
PM_{10} GRIMM	*10.96± 8.07*	1233
$PM_{2.5}$ St Leonard's	*4.41± 4.11*	1246

NB: The high standard deviation values compared to mean values are due to the concentrations following a log-normal distribution. Whilst the median and geometric mean are more appropriate metrics for describing log-normal distribution than the arithmetic mean shown here, exposure response functions for human health impacts are based on the latter.

2.4 Measurement Results - Winter 2023-2024 campaign: Charlestown, Fife

2.4.1 Overview

The average wind speed during the village winter campaign was 6.5 m s^{-1} at 3 m height and the predominant wind direction was from the west (Figure 2.14), but the measurement was obstructed by surrounding buildings. An average temperature of 6 ºC was recorded with a minimum of -3 ºC and a maximum of 13 ºC. The period included significant storms including the named storms Isha (21-22 Jan) and Jocelyn (23-24 Jan), and other very windy periods. The hourly NR-PM_1 concentration measured by the AMS was again mostly lower than 5 µg m^{-3}, except for some polluted episodes, of which the largest was observed between 06/03/2024 and 12/03/2024 with a maximum of 25.7 µg m^{-3}.

Most inorganic compounds (NH_4^+, SO_4^{2-}, NO_3^-) showed very little change during the day, and chloride showing a small peak in the morning and the early evening. On the contrary, the organic aerosol had a well-defined evening peak, most likely related to residential activities (Figure 2.15).

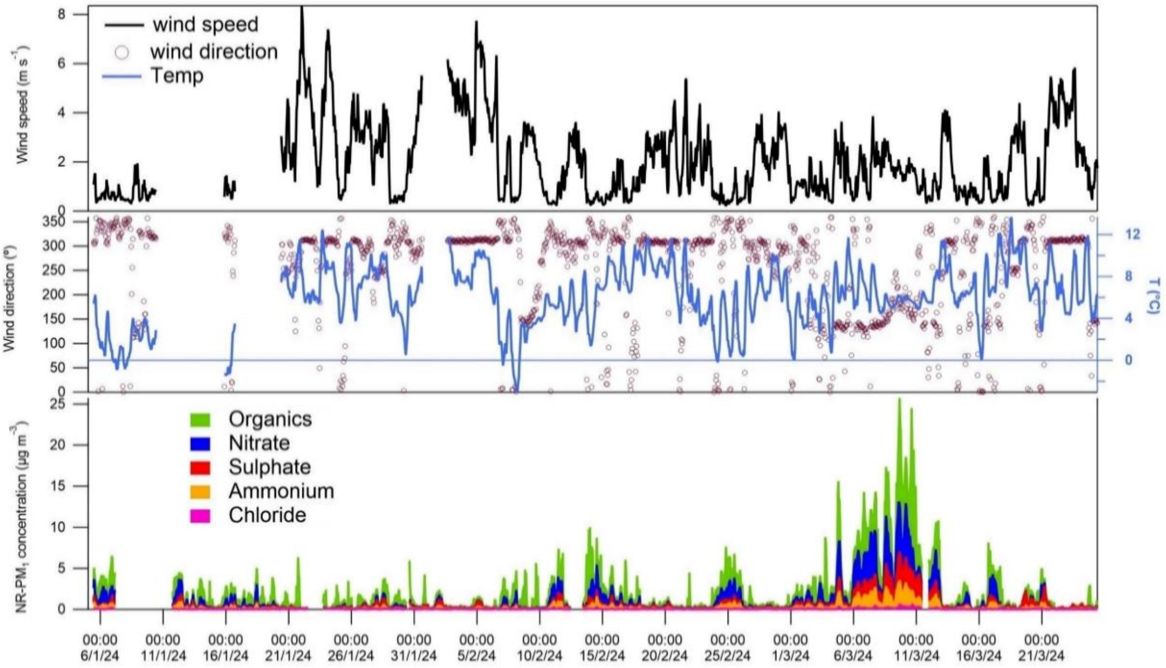


Figure 2.14. Hourly wind speed, wind direction, temperature and NR-PM_1 chemical composition as measured at Charlestown by the met station and AMS.

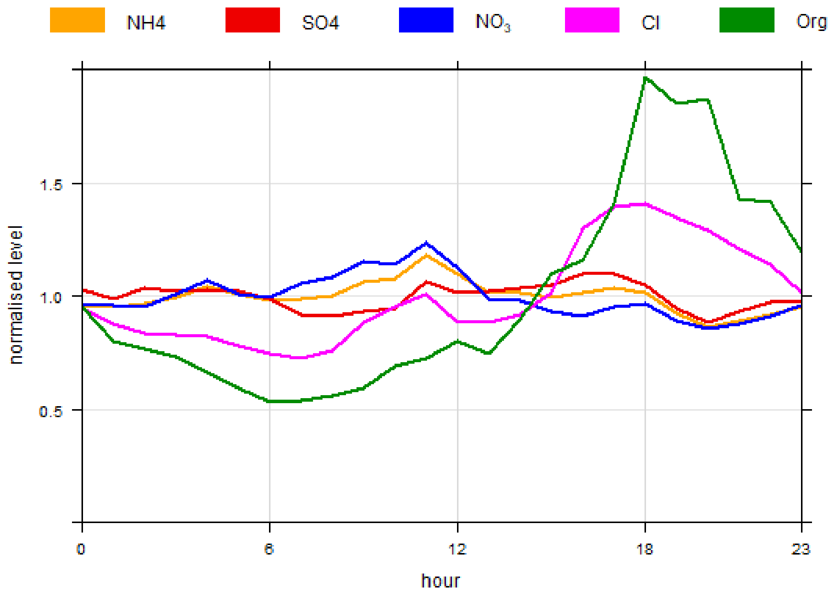


Figure 2.15. Diurnal patterns for the inorganic (NO_3^-, SO_4^{2-}, NH_4^+, Cl^-) and organic aerosol measured with the AMS in Charlestown, Fife.

2.4.2 PMF source apportionment

The PMF analysis on the organic aerosol produced a 5-factor solution, with three primary sources identified: traffic (HOA), which at Charlestown could also include other fossil fuel burning emissions from the Grangemouth refinery, domestic wood burning (BBOA) and domestic coal burning (CCOA). Here, the PMF was able to distinguish a coal factor. In this village setting, a small number of houses are likely to have made a large contribution to the concentration and the measurement therefore does not average over such a large number of burners as in a city setting. If different houses in different directions burn different fuels this would have caused BBOA and CCOA to have different temporal patterns making them distinguishable by PMF. Also, more coal is predicted to be burnt in rural vs urban smoke control areas (see Table 3.3 below). The BBOA factor was characterised by a mass spectrum with a recognisable signal at mass to charge ratios (m/z) 57, 69, 77, 91 and 115 (Figure 2.16) as also found by Lin et al. (2017a) and Tobler et al. (2021). No cooking factor (typically associated with commercial rather than residential cooking) was found during this field campaign, as the location was mainly residential and there were no restaurants nearby.

Unlike with the traffic in Edinburgh, here HOA did not show a clear diurnal pattern, but only a slight increase in the evening, perhaps due to lack of proximity to busy roads. BBOA and CCOA had very similar patterns due to the nature of their sources (domestic heating), with a high peak between 18:00 and 20:00 hrs and concentrations up to 0.8 µg m^{-3} and 0.4 µg m^{-3}, respectively. The two secondary factors LO-OOA and MO-OOA did not show a well-defined diurnal pattern, but just a very small variation in concentration during the day (Figure 2.16).

The correlation between the OA components identified by PMF and other known tracers is shown in Figure 2.17. CCOA and BBOA strongly correlated with each other, as expected, and moderately well with CO and Cl^-. The inorganic compounds strongly correlated with MO-OOA as they did in Edinburgh, consistent with common control through long-range transport of this SOA component. LO-OOA and HOA were only moderately correlated ($R^2 = 0.4$) with combustion tracers (NO_2 and CO). The wind speed seemed to affect LO-OOA, indicating the possibility that this secondary aerosol could be freshly formed locally.

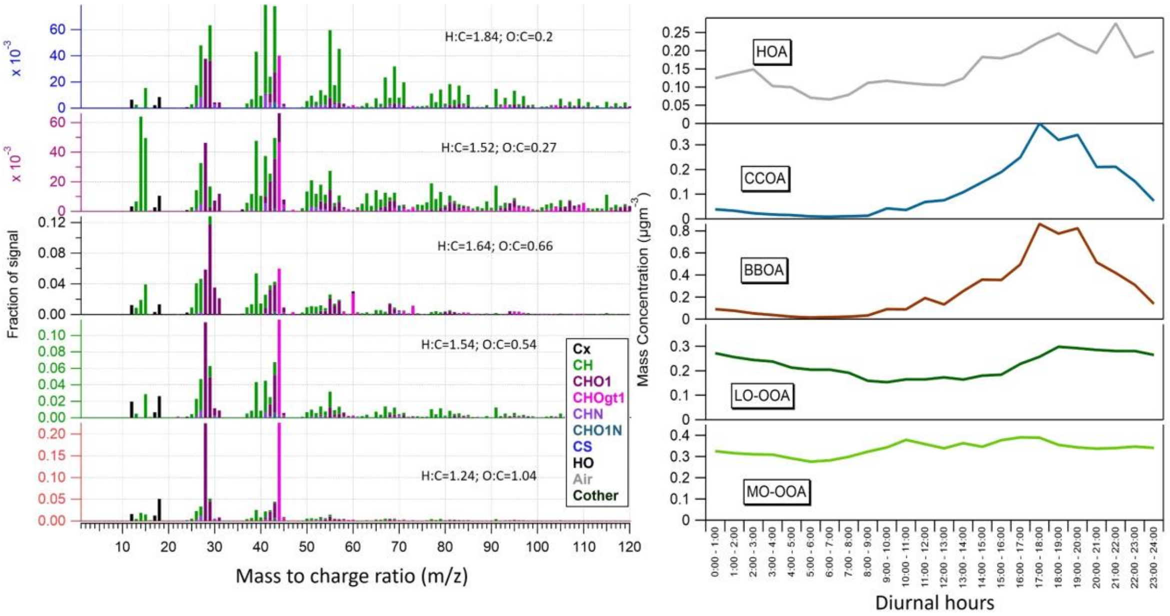
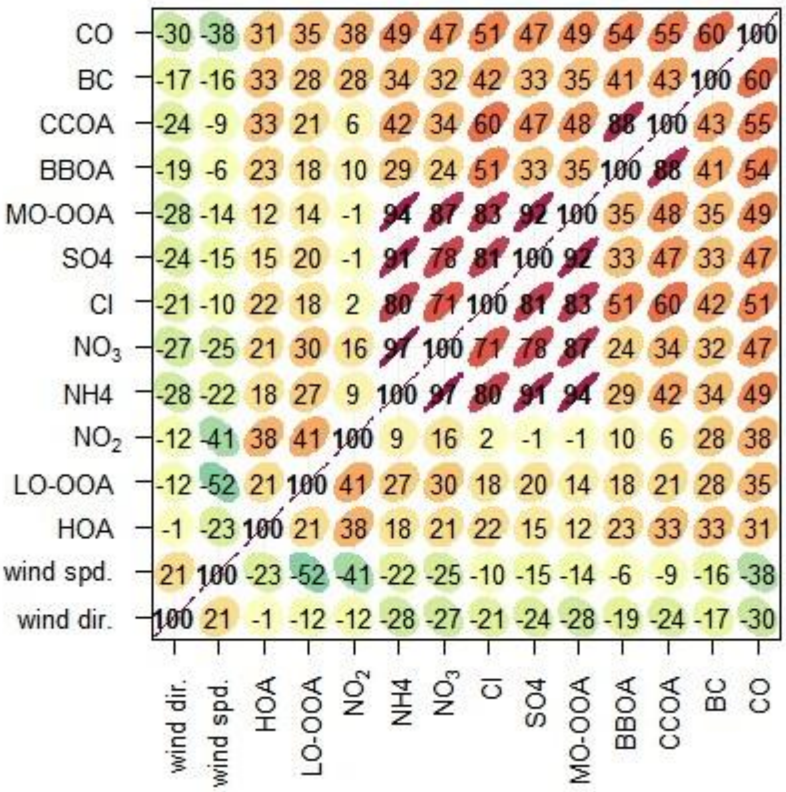


Figure 2.16. PMF results for the measurement campaign in Charlestown, Fife. Left: spectra of the 5-factor solution found for the composition of the organic PM. Right: diurnal cycles of the 5-actor concentrations



	wind dir.	wind spd.	HOA	LO-OOA	NO_2	NH4	NO_3	Cl	SO4	MO-OOA	BBOA	CCOA	BC	CO
CO	-30	-38	31	35	38	49	47	51	47	49	54	55	60	100
BC	-17	-16	33	28	28	34	32	42	33	35	41	43	100	60
CCOA	-24	-9	33	21	6	42	34	60	47	48	88	100	43	55
BBOA	-19	-6	23	18	10	29	24	51	33	35	100	88	41	54
MO-OOA	-28	-14	12	14	-1	94	87	83	92	100	35	48	35	49
SO4	-24	-15	15	20	-1	91	78	81	100	92	33	47	33	47
Cl	-21	-10	22	18	2	80	71	100	81	83	51	60	42	51
NO_3	-27	-25	21	30	16	97	100	71	78	87	24	34	32	47
NH4	-28	-22	18	27	9	100	97	80	91	94	29	42	34	49
NO_2	-12	-41	38	41	100	9	16	2	-1	-1	10	6	28	38
LO-OOA	-12	-52	21	100	41	27	30	18	20	14	18	21	28	35
HOA	-1	-23	100	21	38	18	21	22	15	12	23	33	33	31
wind spd.	21	100	-23	-52	-41	-22	-25	-10	-15	-14	-6	-9	-16	-38
wind dir.	100	21	-1	-12	-12	-28	-27	-21	-24	-28	-19	-24	-17	-30

Figure 2.17. Correlation matrix of all the measurements in Charlestown, Fife. CO and NO_2 were measured by the MIRO MGA. NO_3^-, SO_4^{2-}, NH_4^+, Cl^- and OA components were measured by AMS and BC by AE33. Wind speed and direction were measured with a Vaisala met station. The correlation is coded in three ways: the number (perfect correlation is 100), the elliptic shape (perfect positive correlation is a line with a 45º slope, zero correlation is a circle), and the colour (positive correlation is red, zero correlation is yellow and negative correlation is blue).

2.4.3 Black carbon source apportionment

The Ångström coefficient distributions obtained using the approaches described in Section 2.1.3 are shown in Figure 2.18. In Charlestown, the AAE distributions had a wider range compared with Edinburgh, with the higher values indicating periods of larger dominance from the domestic burning sources. The two site-specific values obtained with the method in (Savadkoohi et al., 2025) were 0.74 and 2.1 for traffic and solid fuels, respectively, not too dissimilar from the source-specific values used by Font et al. (2022) (0.96 and 2), shown in red in Figure 2.18. This indicates that the 0.96 and 2 are suitable coefficients at Charlestown.

The aethalometer model apportionment of the eBC emission sources (traffic and wood burning) showed that on average 27% of the eBC was due to wood burning (eBC_{wb}) and the rest was traffic related (eBC_{tr}). The change in AAE coefficients to site specific values did not make a difference to this ratio. The C_{wood} average concentration in Charlestown was 0.24 µg m^{-3} (representing 4% of $PM_{2.5}$ and 2% of PM_{10}). Applying the AAE site-specific values α_{wb} = 2 and α_{tr} = 0.74 increased average c_{wood} to 0.30 µg m^{-3}.

While eBC_{wb} peaked in the evening, between 18:00 and 20:00, eBC_{tr} showed a small peak around 12:00 and a larger increase around 16:00, in agreement with the PMF factors (Figure 2.20). In this case the measurements suggested a different temporal pattern in solid fuel burning compared to Edinburgh: the evening peak for all four of the solid fuel burning components (eBC_{wb}, BBOA, CCOA) was very prominent throughout the week although it still increased at the weekend, whereas the traffic components did not show any visible feature and only a slight evening increase at the weekend (Figure 2.20). This would be consistent with at least partial use of solid fuel as a primary heat source rather than for pleasure fires.

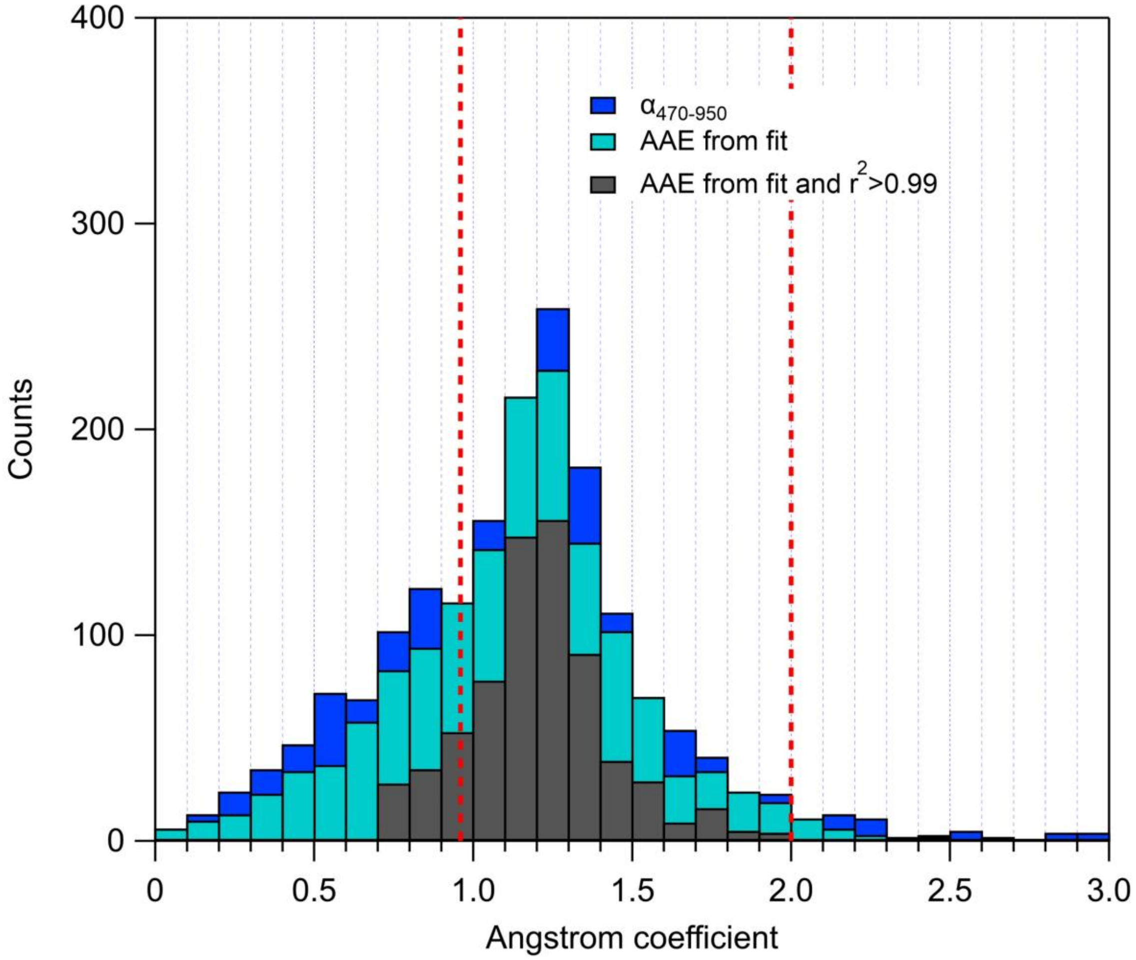


Figure 2.18. The absorption Ångström exponent (AAE) distribution in Charlestown, Fife: AAE calculated from the ratio of the 470 nm and 950 nm channels (blue**),** from the log-log fit of the absorption coefficient as a function of wavelength (green), and with the application of a filter (r^2>0.99) (grey). The red lines represent the α values used for traffic and wood smoke.

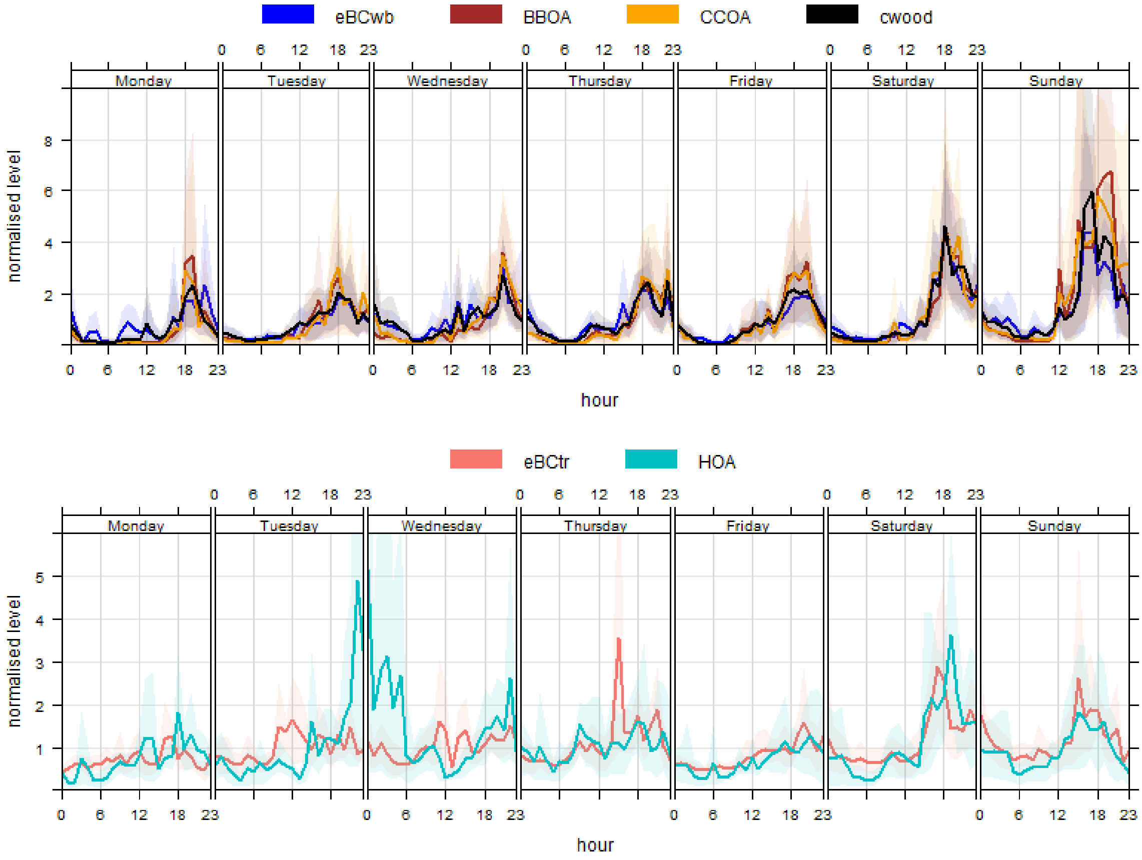


Figure 2.19. Diurnal patterns for each day of the week of concentration for the domestic burning sources (top) and the traffic component (bottom) observed in Charlestown, Fife.

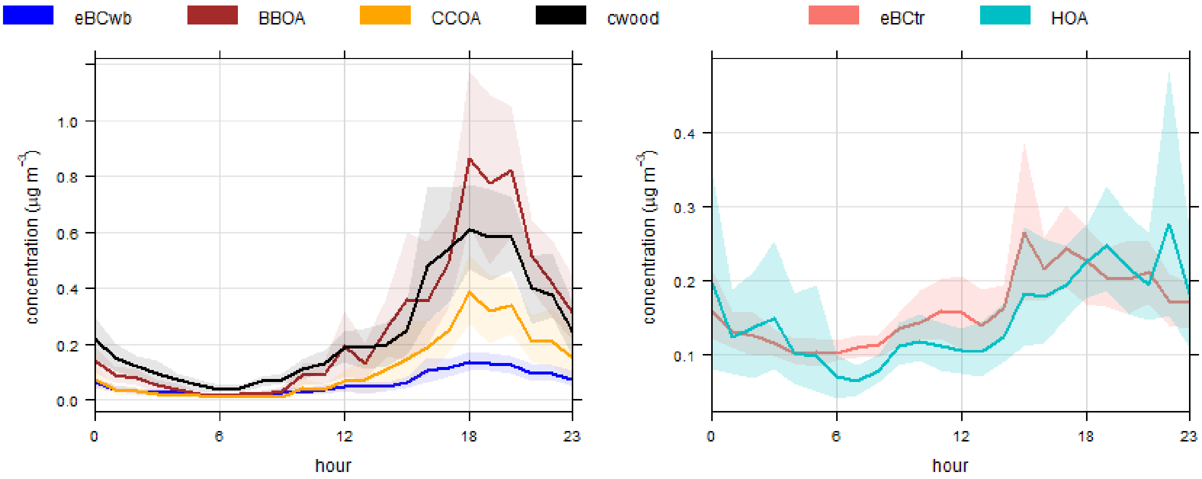


Figure 2.20. Diurnal cycles for the domestic burning (left) and traffic components (right) measured between 05/01/2024 and 25/03/2024 in Charlestown, Fife.

2.4.4 Spatial analysis of sources

The polar plots of the mean concentrations and CPF showed that domestic burning and traffic sources were localised towards the south of the measurement point and made the largest contributions at low wind speed, with some influence from sources WSW in the direction of Grangemouth and / or Falkirk (Figures 2.21 & 2.22).

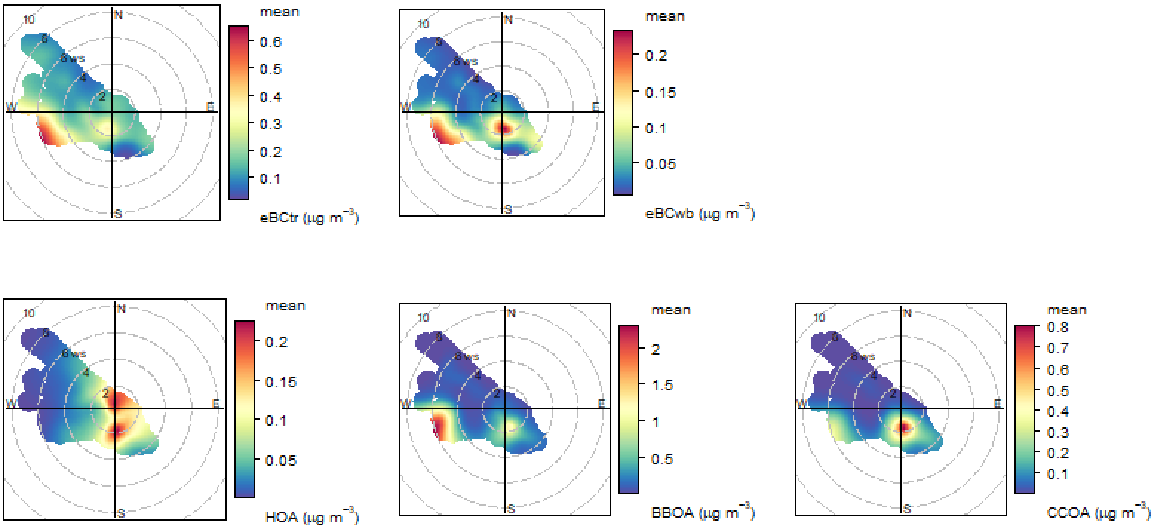
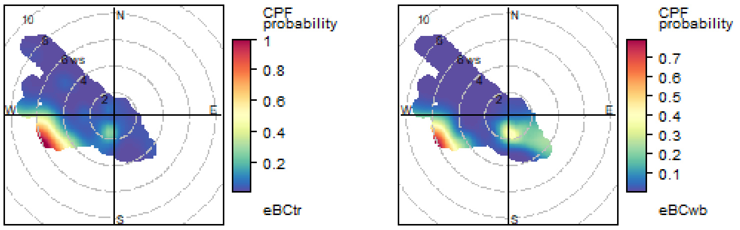
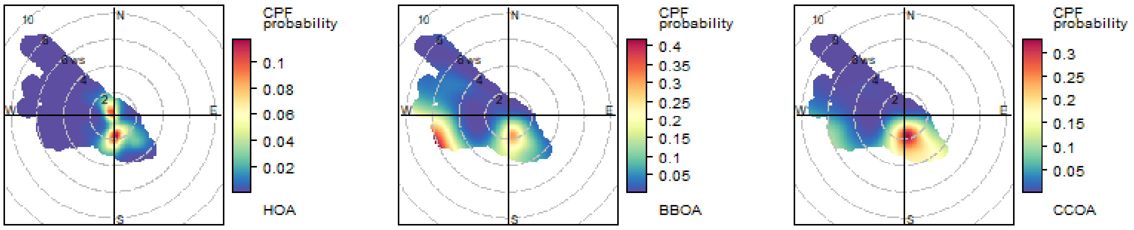


Figure 2.21. Mean concentration variation with wind speed and wind direction for the traffic sources (eBC_{tr} and HOA plots on the left) and domestic burning (eBC_{wb}, BBOA, CCOA plots on the right) in Charlestown, Fife. Wind speed is in m s^{-1} along the radial axis.



CPF at the 90th percentile (=0.35)

CPF at the 90th percentile (=0.17)



CPF at the 90th percentile (=0.35)

CPF at the 90th percentile (=0.69)

CPF at the 90th percentile (=0.32)

Figure 2.22. CPF variation with wind speed and wind direction for the traffic sources (eBC_{tr} and HOA plots on the left) and domestic burning (eBC_{wb}, BBOA, CCOA plots on the right) in Charlestown, Fife. Wind speed is in m s^{-1}.

2.4.5 PM mass composition

The average PM_1 mass concentration measured at Charlestown was 2.21 µg m^{-3}, while the sum of the components measured by the AMS and the aethalometer was 2.53 µg m^{-3}. As with the previous campaign, concentrations of $PM_{2.5}$ and in particular of PM_{10} were much (by a factor 2 and 5) larger at 5.77 and 12.53 µg m^{-3}, respectively, and this was due to a high contribution of coarse seasalt (Na^+ and Cl^-) associated with the high windspeeds, as corroborated with measurements of these components in $PM_{2.5}$ and PM_{10} at the Auchencorth Moss supersite, 20 km S of Edinburgh. The mean values and standard deviations for all the measured PM_1 components are shown in Table 2.2: the high standard deviations reflect the large variation in the measured hourly concentrations.

The evening peak in the PM_1 mass concentration was driven by the domestic burning components, representing 17% of the total mass. Overall, the primary sources in Charlestown contributed 29% of the total PM_1 mass, of which 12% was traffic. Like in Edinburgh, the inorganic component accounted for the largest part of the secondary aerosols (49% of the PM_1 mass), and the secondary organic aerosols added up to 22% of the PM_1 mass (Figure 2.23).

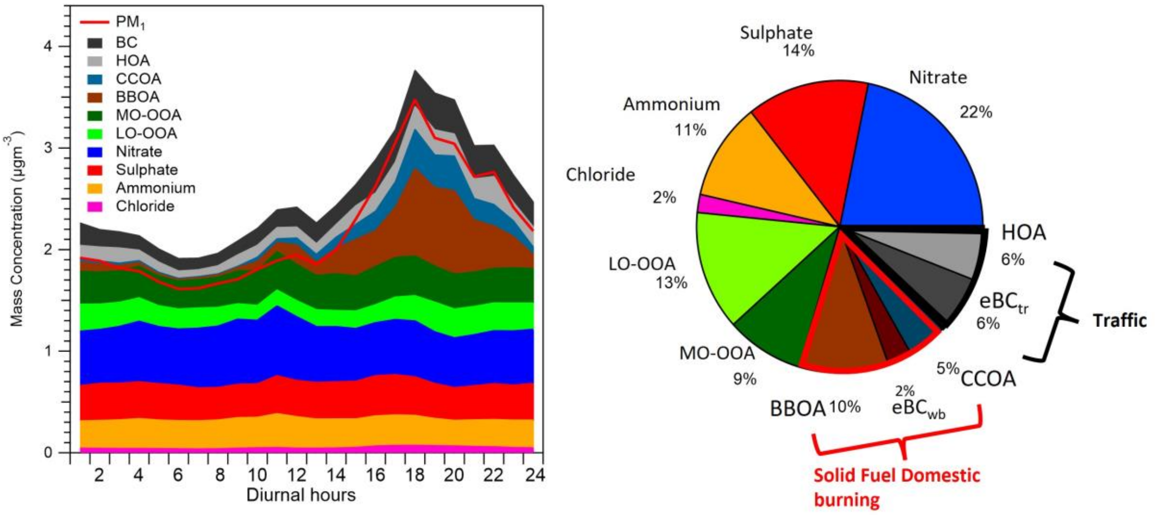


Figure 2.23. PM_1 mass composition: diurnal cycle with stacked components (left); pie chart with quantification (%) of each component contribution (right).

Table 2.2 Summary of the mean concentrations measured during the campaign in Charlestown between 05/01/2024 and 25/03/2024

PM_1	Mean ± stdev ($\mu g\ m^{-3}$)	N points
NO_3^-	0.56 ± 1.01	1744
SO_4^{2-}	0.35 ± 0.53	1744
NH_4^+	0.28 ± 0.45	1744
Cl^-	0.05 ± 0.07	1744
eBC_{wb}	0.06 ± 0.19	1820
eBC_{tr}	0.16 ± 0.19	1810
MO-OOA	0.34 ± 0.73	1744
LO-OOA	0.22 ± 0.21	1744
BBOA	0.26 ± 0.65	1744
CCOA	0.12 ± 0.34	1744
HOA	0.15 ± 0.34	1744
Total PM_1	2.55 ± 1.68	
PM_1 GRIMM	2.21 ± 2.67	1828
$PM_{2.5}$ GRIMM	5.77 ± 5.05	1828
PM_{10} GRIMM	12.53 ± 10.72	1828

NB. The high standard deviation values compared to the mean are due to the concentrations following a log-normal distribution.

2.5 Solid fuel burning PM derived from the Scottish sites of the UK black carbon network

Two Scottish sites, Auchencorth Moss (rural) and Glasgow Townhead (urban background), are part of Defra's black carbon (BC) monitoring network and have a long dataset of BC concentration measured by aethalometer since 2009. In their study on PM from domestic burning in the UK, Font et al. (2022) presented long time trends of c_{wood} concentration, intended to reflect the portion of PM due to wood burning, derived from the aethalometer data from 2009 until 2021 for the entire BC network. c_{wood} concentrations were calculated as described in Section 2.1.4. In the present study, the c_{wood} time series for Auchencorth Moss and Glasgow Townhead was extended until the end of 2024 (Figure 2.24), following the same methodology.

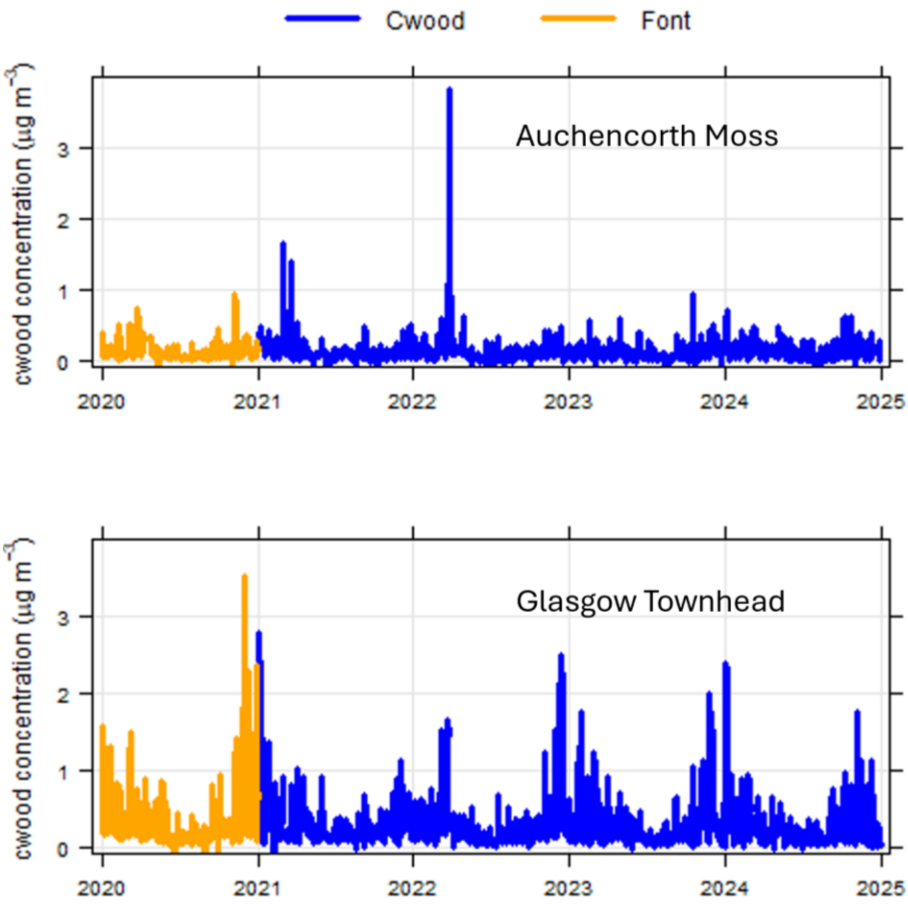


Figure 2.24. Time series of c_{wood} (daily averages) for the Scottish sites (Auchencorth Moss and Glasgow Townhead) from January 2020 until December 2024. Values from Font et al. (2022) are shown in yellow.

The annual mean c_{wood} concentration was 0.12 µg m^{-3} at Auchencorth Moss and 0.28 µg m^{-3} at Glasgow Townhead. The largest values were observed during the winter period from November to March, consistent with the domestic heating season (Figure 2.25), with mean concentrations of 0.17 µg m^{-3} for the rural site and 0.43 µg m^{-3} at the urban site (Table 2.3). The largest monthly values measured in November and December at Glasgow Townhead were in agreement with the observations in the previous study. The high concentration reported at Auchencorth Moss in March was driven by a pollution episode in 2022. Minimum concentrations were observed during the summer months between May and August. In Glasgow Townhead, on average, c_{wood} represented 3% of the annual mean PM_{10} concentration and 5 to 6% of $PM_{2.5}$. At Auchencorth Moss c_{wood} contributed 2 to 3% to PM_{10} and 3 to 4% to $PM_{2.5}$, with very little variation from year to year. However, the c_{wood} annual averages suggested a small but consistent decreasing trend, already reported by Font et al. (2022) which continued into 2022 to 2024 and was more evident in the urban environment (Table 2.3).

The same methodology was applied to the Edinburgh and Charlestown datasets. c_{wood} concentration in Edinburgh was 0.19 µg m^{-3} (representing 4% of $PM_{2.5}$ and 2% of PM_{10}) and 0.24 µg m^{-3} in Charlestown, representing 4% of $PM_{2.5}$ and 2% of PM_{10} (Table 2.3). The values were comparable with the ones obtained for the other two Scottish sites in winter, albeit a bit smaller. The slightly lower values might be because November and December, some of the most polluted months for domestic wood burning, were not covered by the campaigns, and both campaigns were characterised by unusually high wind speeds. When using the site-specific AAE values the average c_{wood} in Edinburgh increased to 0.41 µg m^{-3} and in Charlestown to 0.28 µg m^{-3}.

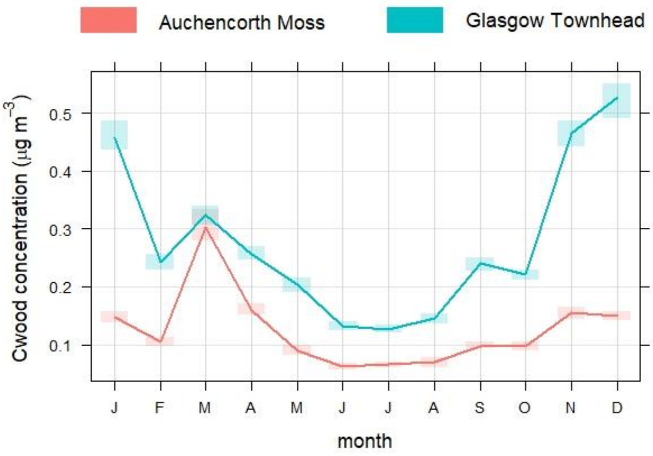


Figure 2.25 Mean monthly variations and 95% confidence interval of c_{wood} for Auchencorth Moss and Glasgow Townhead over the period (2020-2024).

Table 2.3 Summary of c_{wood} average calculation for the Scottish sites of the Black Carbon network.

Site	Year	c_{wood} Annual Mean ± st. dev (µg m^{-3})	N points	c_{wood} Winter mean ± st. dev (µg m^{-3})	N points
Auchencorth Moss	2012-2021	0.19 (0.19-0.19)*	68,937	0.26 (0.25-0.26)*	28,332
	2020	0.15 ± 0.25	6,298		
	2021	0.12 ± 0.30	8,557	0.19 ± 0.44	3,570
	2022	0.14 ± 0.43	7,466	0.19 ± 0.59	3,394
	2023	0.10 ± 0.24	8,091	0.12 ± 0.26 (0.12)***	3,481
	2024	0.15 ± 0.25	4,420	0.16 ± 0.25 (0.18)***	2,770
	2020-2024	0.12 ± 0.30	34,832	0.17± 0.39	13,215
Glasgow Townhead	2013-2021	0.42 (0.42-0.43)*	58,526	0.56 (0.55-0.57)*	25,849
	2020	0.30 ± 0.58	8,568		
	2021	0.29 ± 0.50	8,251	0.53 ± 0.93	3,395
	2022	0.29 ± 0.51	8,130	0.34 ± 0.46	3,334
	2023	0.26 ± 0.43	8,318	0.43 ± 0.69 (0.32)***	3,619
	2024	0.25 ± 0.44	6,362	0.40 ± 0.64 (0.36)***	3,240
	2020-2024	0.28 ± 0.50	39,629	0.43 ± 0.68	13,588
Edinburgh	2023			0.19 ± 0.36**	1,243
Charlestown	2024			0.24 ± 0.50**	1,808

NB. The high standard deviation values compared to the mean are due to the concentrations following a log-normal distribution.

**Mean (95% confidence interval) for the years indicated in the table as seen in Font et al. (2022).*

***Winter averages do not include November and December.*

****Matched to the campaign duration and coverage.*

3 Calculation and mapping of emissions

3.1 Methodology

3.1.1 Fuel usage statistics

As mentioned in the introduction, new statistics on fuel usage across the UK have become available from the 2022/23 fuel survey (Defra, 2024a). This survey was composed of two parts: a large survey of 50 weeks from July 2022 to June 2023 with around 1000 interviews per week across the UK targeting adults aged 16+, here referred to as the Core Activity Survey 2022 (CAS2022/23), which also included information from 3294 interviewees based in Scotland, and a more comprehensive Point in Time Survey on practices. This was a rerun of a similar exercise conducted in 2018/19 (CAS2018/19) (Kantar, 2020). Both surveys also covered outdoor burning, but those data are not used in the present study which focusses on indoor burning. The results from both surveys were scaled up by Ricardo to provide total fuel volumes for the four nations (Defra, 2024b) (Table 3.1).

Table 3.1. Fuel quantities burnt inside in residential properties based on the CAS2022/23 and CAS2018/19 (Defra, 2024b) in kt yr^{-1}.

	CAS2022/23				CAS2018/19			
Government Office region	Wood logs	Other wood	House coal	Other coal products	Wood logs	Other wood	House coal	Other coal products
Scotland	**193**	**113**	**34**	**69**	**98**	**19**	**12**	**9**
England	1,125	771	210	456	1,133	238	136	418
Wales	84	74	14	42	106	43	47	52
N Ireland	68	43	37	30	93	7	155	29
UK	1,471	1,000	296	597	1,429	307	350	509

The comparison of the UK totals from the two surveys overall shows good consistency not only between surveys, but also with the Digest of United Kingdom Energy Statistics (DUKES[1]). The exception is an increase in the estimated amount

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[1] https://www.gov.uk/government/collections/digest-of-uk-energy-statistics-dukes

of other wood products burnt, which would be consistent with an increasing stove population in the UK, but it could also in part reflect a larger use of waste wood triggered by the fuel crisis. The attribution of fuel to Scotland, however, increased significantly between the two surveys, by a factor of 2 for wood logs, 5.9 for other wood products, 2.8 for house coal and 7.7 for other coal products. It is worth noting that the previous Scottish study to estimate emissions from the most polluting solid fuels (Masey et al., 2023) was based on the CAS2018/19 statistics and that the modelling results presented here are therefore based on significantly larger fuel quantities being used in Scotland.

The fuel quantification based on the CAS2022/23 (Defra, 2024b) includes in an associated spreadsheet[2] a split of the Scottish wood log fuel and total wood fuel into (a) the three moisture categories into (b) rural vs urban and into (c) SCA vs non-SCA and (d) by appliance (open fires vs other appliances). The equivalent data for total "other wood-based fuels" can be derived by difference between total wood and wood log data. This data can directly be used to attribute fuel amounts, by moisture category in the case of logs, to the urban SCA, urban non-SCA and rural categories.

The spreadsheet does not provide the same breakdown for coal and coal products; the quantification report (Defra, 2024b) mentions that some CAS2022/23 respondents were confused about whether they were burning coal, coal products or charcoal. The totals in Table 3.1 include adjustments for reporting of inconsistent combinations of burning appliances and coal type, but overall the uncertainty associated with this misunderstanding was deemed to be too large to derive the same granularity of coal/coal product use by appliance type and rural/urban category.

For the purpose of this study we used the breakdown of the other wood-based fuel and other coal product categories available at aggregated UK level and we have assumed that this split is also representative for Scotland. Using this approach we estimate that the other 113 kt of wood-based fuels were composed of 9.9% wood briquettes, 8.7% pellets, 5.6% woodchips, 15.7% kindling, 7.5% green or garden waste and 52.6% waste wood. In addition, we estimate a consumption of 9.9 kt of wood pellets and 6.3 kt of wood chip. We assume each of these fuel types to be distributed across urban / rural categories as the total wood-based fuel.

Similarly, the 69 kt of coal products break down into 76.5% manufactured solid fuels (MSF), 13.6% anthracite, 6.0% petroleum coke and 3.8% lignite. Unlike for wood fuels no quantitative split into urban vs non-urban or SCA vs non-SCA is provided for house coal and coal products, and the total MSF amount is not broken down into smokeless and non-smokeless MSFs.

We allocate house coal and coals products to the three rural/urban/SCA categories as follows: we combined the fraction of CAS2018/19 UK respondents being coal (product) burners (6.1% of rural population, 1.4% of urban SCA and 4.3% of urban non-SCA) with the total number of Scottish households in each category, and respondents' data on the average burn length, to estimate that 32.8% of coal/coal products are burnt in rural households, 14.4% in urban SCA and 52.8% in

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[2] https://sciencesearch.defra.gov.uk/ProjectDetails?ProjectID=20159&FromSearch=Y&Publisher=1&SearchText=AQ1017&SortString=ProjectCode&SortOrder=Asc&Paging=10

urban non-SCA. We then used responses on whether UK respondents burnt house coal or "smokeless coal" or both to derive the distribution pattern of total coal-based fuel use shown in Table 3.2. This process has the potential to scale up small errors to have large impacts in the disaggregation. For example, the value of coal (product) burners in urban SCAs of 1.4% from the CAS might be unrepresentative due to the limited sample size of positive respondents. Because this value is multiplied by a large number of households in urban areas, too much or too little coal (product) fuel may be attributed to SCAs.

Table 3.2. Distribution of coal and coal-product consumption across residential classes and appliance categories

	Appliance category	Rural	Urban SCA	Urban non-SCA
House coal, lignite, peat	Open fire	12.4%	4.8%	15.1%
	Stove	26.9%	8.2%	32.6%
MSF	Open fire	8.5%	5.8%	18.1%
	Stove	18.4%	9.9%	39.3%

We use UK-wide data for the fraction of the population within each residential group who burn any type of coal from the CAS2018/19, together with average burn length by residential group from the CAS2022/23 and population statistics to allocate the total amount of coal (products) burned by each residential group.

Amounts of "other fuels" are only available at total UK level. We assume that 8.4% of all coffee logs are burnt in Scotland (2.4 kt) and 8.4% of all household waste (3.9 kt), following the ratio of the Scottish to the UK population (8.4%). Peat burning is dominated by the Western Isles and Northern Ireland. We make the somewhat arbitrary assumption that 1/3 of all the peat estimated to be burnt in the UK is burnt in Scotland (i.e. 19.4 kt).

Table 3.3. Scottish solid fuel consumption by type, together with allocation to burning area types and appliance type (open fire / stove) [in kt yr^{-1}]

Fuel		Moisture class	Total Scottish Consumption	Urban SCA (fire/stove)	Urban non-SCA (fire/stove)	Rural (fire/stove)
Wood logs	Wood logs	Pre-dried (10% MC)	124.6	1.3 / 5.1	11.6 / 35.3	12.2 / 59.1
	Wood logs	Seasoned (20% MC)	42.2	0.4 / 1.7	3.9 / 12.0	4.1 / 20.0
	Wood logs	Wet (30% MC)	27.1	0.3 / 1.1	2.5 / 7.7	2.7 / 12.9
Wood products	Wood briquettes	Pre-dried (10% MC)	13.0	0.7 / 1.3	2.2 / 5.3	1.0 / 2.5
	Wood pellets	Pre-dried (10% MC)	9.9	- / 7.4	- / 1.8	- / 0.7
	Wood chips	Seasoned (20% MC)	6.3	- / 4.6	- / 1.2	- / 0.5
	Waste wood	Pre-dried	69.4	1.1 / 2.5	7.4 / 22.4	12.4 / 23.6
	Kindling	Seasoned	20.8	0.3 / 0.7	2.2 / 6.7	3.7 / 7.1
	Green / garden waste	Wet (30% MC)	9.8	0.2 / 0.4	1.1 / 3.2	1.8 / 3.4
Coal / Coal products	House coal		34.0	1.6 / 3.4	5.1 / 10.7	4.2 / 8.8
	Anthracite		9.4	0.5 / 0.9	1.7 / 3.7	0.8 / 1.7
	Petroleum coke		4.2	0.2 / 0.4	0.6 / 1.3	0.5 / 1.1
	Lignite		2.6	- / -	0.5 / 1.0	0.4 / 0.8
	MSF		52.8	3.1 / 5.2	9.6 / 20.7	4.5 / 9.7
Other	Peat		19.4	- / -	10.6 / 7.3	8.8 / 6.0
	MSW*		3.9*	N/A	N/A	N/A
	Coffee logs		2.4	0.1 / 0.2	0.4 / 0.9	0.2 / 0.4

* Municipal solid waste (MSW) was not included in the emission estimates; emission factors are currently too uncertain.

3.1.2 Emission factors

Emission factors were taken from Defra's recent ESDFS study (Allan et al., 2025) which investigated the effects of both fuel and appliance types. Appliance tested included an open fire, two pre-2000 closed stoves ("Hunter Oakwood" and "Coalbrookdale (aka Aga) Little Wenlock" referred to as "old stoves"), two 2000-2009 closed stoves ("Stovax / Dovre Model 500MRF" and "Redfyre (Gazco) Kensal 20 RFD-KEN20M", referred to as "middle stoves") and very efficient modern stoves (clarSkies level 2 or above) ("Charnwood Model C4 wood and multifuel versions", "Hunter Aspect 5" and "Hunter Aspect 8 (Large Stove)", referred to as "modern stoves". Also included were a Blue-Angel-ecolabelled "Hase Sila IQ+" and a pellet stove ("Island Stoves, Ramsey"). Fuels included beech logs of three moisture classes, house coal (CPL Premium House coal trebles, not authorised smokeless), anthracite, coffee logs, wood briquettes and wood pellets. Two manufactured solid fuels were included: a high smoke/sulphur fuel (Briteflame ovoids, only sold in Scotland, not England, similar to Superheat) and a low smoke/sulphur fuel (HETAS approved, authorised smokeless fuel). This study has thrown up some surprising results. In particular:

- $PM_{2.5}$ emission factors from seasoned wood (defined as having dried for >12 months, but not kiln dried, or with MC of 20%) were greatly reduced compared with earlier emission factors, and seasoned wood showed significantly smaller emissions than pre-dried wood (MC 10%). As expected, largest emissions were measured for wet wood (MC 30%).
- "Modern stoves" (defined as those that have an Ecodesign certification or comply with clearSkies level 2 or higher) showed higher $PM_{2.5}$ emissions for some fuels than "middle stoves".
- There was surprisingly little difference in the emissions from approved and non-approved MSF and the difference did not always go the expected way. For the same stove, the approved fuel emitted 17% more $PM_{2.5}$ and 69% more SO_2.
- For several devices $PM_{2.5}$ emissions of low sulphur MSF (i.e. "smokeless fuel" permitted to be burnt in smoke control areas in approved appliances) showed higher emissions than house coal (which actually had lower sulphur content) and this included modern stoves.

Overall the ESDFS study derived lower emission factors than previously used in the NAEI for anthracite, coal and coffee logs, and higher emission factors for MSF.

The ESDFS study has advantages and disadvantages which were discussed by the Air Quality Inventory Steering Group (AQISG). Unlike previous emission factors these were derived from measurements with a unified and approved methodology with high quality control. However, they reflect a relatively small number of appliances and fuel types (beech wood was the chosen log species and the coals and MSF types may not be fully representative), potentially with idealised burning cycles. We have some concerns in terms of generalisation and application for Scotland:

- Burning practices vary widely and include many situations of non-optimum burning conditions. For example, appliances are often kept well below their optimum power output rating, increasing emissions. Banking of fires overnight will make relighting more efficient but, by definition, leads to poor combustion conditions during banking.

- A moisture content of wood of 20% for 12 months of seasoning seems optimistic for the Scottish climate. A lot of seasoned wood used in Scotland is likely to have a moisture content >20%. For example, the average relative humidity at the West Coast typically ranges from 75 to 85%. At this RH the equilibrium moisture content of wood is in the range 15% to 18.5% (Figure 3.1), depending on wood species, and the drying time towards <20% would be very long.
- The single wood species chosen (beech) may not dominate across the whole of Scotland, with some firewood suppliers specialising in local softwood.
- There is a possibility that a large fraction of emissions might originate from a small number of 'super polluters' e.g. burning high emission waste wood such as treated and painted timber.

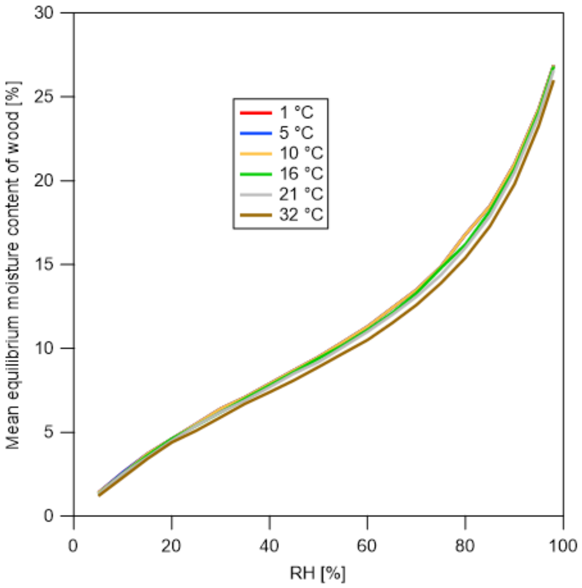

Figure 3.1. Mean equilibrium moisture content (%) of wood as a function of air relative humidity and temperature[3].

Emission factors are expressed either in terms of amount emitted per fuel mass burnt or, as from the EFDSF study, in terms of emission per energy amount contained in the fuel. Conversion between these units is by the Calorific Value (CV), i.e. the assumed energy content of the fuel per unit mass (kg). Emission factors were primarily taken from the EFDSF study, but there are gaps with respect to combinations between fuels and appliance types. These gaps were filled by ratioing

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[3] from: https://www.bowens.com.au/blog/understanding-humidity-and-timber-performance/

emissions to a measured emission based on a similar fuel that was tested on both appliance types.

Emissions of anthracite were not measured for fireplaces and here the MSF emission factor ratios were used to extrapolate the results on the basis of Old Stoves. Because the EFDSF study prioritised the burning practices that were deemed most common, this affects a minor fuel amount and should not introduce too much uncertainty into the end result.

Peat was not included as a fuel in the EFDSF study. Some emission factors are available from other studies, but they do not cover all pollutants. The decision was therefore made to include peat in the coal estimate and to use the house coal emission factors, on a per kt basis. Since the calorific value of peat (12.8 GJ/t) differs significantly to that of coal (26.56 GJ/t) we converted the 19.4 kt of peat fuel into a 9.4 kt equivalent of coal fuel. Distributing this together with the 34 kt of house coal also implicitly deals with the fact that in geographical regions of Scotland where peat fuel is used, it most likely substitutes for house coal use. Comparison with the data from the Future Fuels Report (Price-Allison et al., 2022) provides a sense-check of this approach. That report provided a total PM emission factor for peat briquettes burnt in a medium old stove of 2.48 g/kg, equivalent to 194 g/GJ. The EFDSF study's ratio of $PM_{2.5}$ to total PM for coal is of the order of 88%, suggesting that the Future Fuels Report implies a $PM_{2.5}$ EF of about 171 g/GJ. This is very close to the house coal emission factor of 178 g/GJ applied to peat in the methodology used here. Admittedly, this is likely at least partly fortuitous given the difference in the measurement methodology and burn cycles used between the two studies, and the variability between different fuels of a single category.

The fuels survey suggests about 3.9 kt of municipal solid waste (MSW, also referred to as household waste) are burnt in Scotland every year. We are not aware of any reliable indoor emission factors for MSW burning, and this fuel type was not covered in the EFDSF study. This waste presumably contains anything from newspaper and cardboard packaging to plastics, and it covers materials with vastly different emission factors. More fuel statistics would need to be collected to specify what exactly is burnt across Scotland to develop robust emission estimates. An emission estimate is not further included in this study, although it could be a significant local source similar to the burning of treated and painted waste wood mentioned above. This fuel is also not covered by the NAEI.

Inclusion of condensable PM into the emission factors is subject to active debate in the scientific community and at international level (e.g. Simpson et al., 2020). It was included in the $PM_{2.5}$ emission factor measurement approach of the EFDSF study, which also measured the filterable $PM_{2.5}$ fraction and derived the condensable fraction by difference between total and filterable. However, they estimates of condensable PM were not included in the numerical summary tables for all fuel types (Allan et al., 2025, Table A-7-1). Nevertheless, Tables 5-5, 5-6 and 5-7 include EFs for the condensable PM and plotting this EF against the EF for $PM_{2.5}$ shows a reasonably tight correlation (Figure 3.2), with a linear relationship of slope 0.56 (and of 0.59 for a linear fit with zero intercept). We here estimate the emissions for the Condensable PM fraction using EFs for this tables where available and estimating unavailable EFs as 0.59 times the EF for $PM_{2.5}$. Both Condensable PM and BC emission estimates provided here are sub-categories of the primary $PM_{2.5}$ emissions and are given for information.

Table 3.4 summarises the emission factors and CV values used in this study.

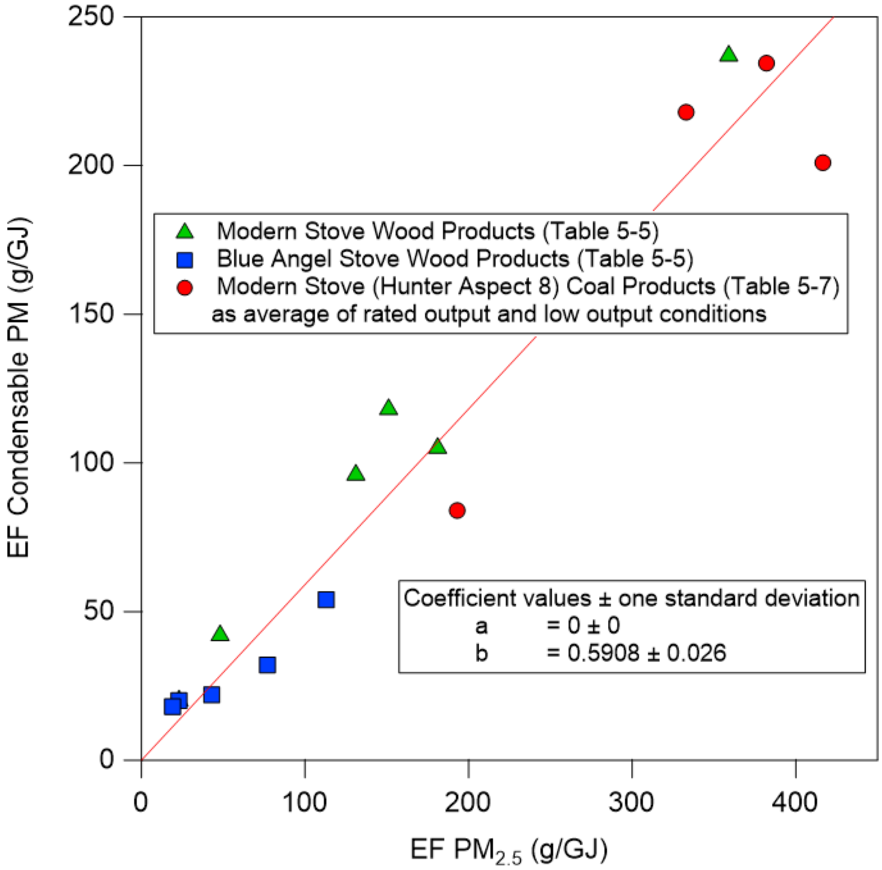

Figure 3.2. Relationship between the emission factors for Condensable PM and total $PM_{2.5}$ (including Condensable PM) from the EFDSF study (Allan et al., 2025).

Table 3.4. Emission factors for wood and wood-based fuels applied during this study and their literature source

		NCV	Emission factors [g/GJ]							References	
Fuel	Appliance	[GJ/t]	CO	NOx	VOCs	$PM_{2.5}$	SO_2	BC	Con PM^b	EFs	NCV
Logs pre-dried, waste wood	Open fire	17.25	1164	58	183	88	5	47	52	a	a
	Stove old		1486	52	179	79	6	39	47	a	
	Stove mid		2474	29	562	75	7	49	44	a	
	Stove modern		2386	41	373	131	8	58	96	a	
Logs seasoned, kindling, wood chip	Open fire	14.87	2159	57	391	306	4	288	181	a	a
	Stove old		1938	46	165	104	5	5	61	a	
	Stove mid		1959	59	328	161	7	63	95	a	
	Stove modern		1503	55	96	48	5	17	42	a	
Logs wet, green/garden waste	Open fire	13.04	2426	50	584	510	3	510	301	a	a
	Stove old		3135	61	293	258	5	36	152	a	
	Stove mid		3438	60	603	679	5	659	401	a	
	Stove modern		3315	55	753	359	5	226	237	a	
Wood briquettes	Open fire	17.79	2123	117	414	428	135	86	253	a	a
	Stove old		1974	129	127	100	96	18	59	a	
	Stove mid		4265	112	2117	431	338	250	254	a	
	Stove modern		1728	46	112	181	7	60	105	a	
Pellets	Pellet stove	16.94	502	66	3	17	90	4	36	a	a

a) EFDSF (Allan et al., 2025); b) From EFDSF Tables 5-5 to 5-7 or estimated as 0.59 x EF for $PM_{2.5}$; see text.

Table 3.5. Emission factors for coal and coal-based fuels applied during this study and their literature source

		NCV	Emission factors [g/GJ]							References	
Fuel	Appliance	[GJ/t]	CO	NOx	VOCs	$PM_{2.5}$	SO_2	BC	ConPM	EFs	NCV
House coal, lignite, petroleum coke, peat[d]	Open fire	26.46	2834	101	501	271	145	38	160	a	a
	Stove old		5184	91	262	148	188	21	87	a	
	Stove mid		4265	112	2117	431	338	310	254	a	
	Stove modern		3603	121	288	234	229	173	235	a	
Anthracite	Open fire	32.44	2496	91	789	171	316	9	101	c	a
	Stove old		3368	113	543	126	439	48	74	a	
	Stove mid		3371	120	181	34	359	12	20	a	
	Stove modern		5603	76	459	73	530	53	84	a	
MSF / SMSF	Open fire	26.14	3529	95	826	294	822	3	173	b	a
	Stove old		4763	118	569	217	1141	15	128	b	
	Stove mid		4110	120	531	194	1174	128	114	b	
	Stove modern		5123	122	993	397	1235	214	210	b	
Coffee logs	Open fire	17.57	921	49	91	142	5	128	84	a	a
	Stove old		2296	31	242	187	8	194	77	a	
	Stove mid		1572	123	73	117	81	120	23	a	
	Stove modern		1992	155	184	151	128	153	54	a	

a) from EFDSF (Allan et al., 2025); b) from EFDSF (Allan et al., 2025), calculated as average of EFs for low sulfur and high sulfur MSF; c) calculated as EF for anthracite for stove old x EF for MSF for open fire / EF for MSF for stove old; d) EFs (in g/GJ) for peat were approximated by the EFs for coal, and the quantity of peat was converted into the equivalent amount of coal with the same energy, see text.

3.1.3 Spatial distribution and allocation to appliance type

In the absence of Scotland specific numbers, fuel amounts used in stoves were distributed over the three different age categories (old, middle and modern stoves) based on the UK appliance age profiles from the CAS2022/23: 32% of appliances were estimated to be "old" (pre 2010), 51% of appliances were "mid" (2010-2020) and 17% of appliances were "modern" (2020 onwards). The same distribution was applied to all fuels, assuming no relationship between appliance age and its usage.

Spatial distribution maps of domestic solid fuel (SF) usage across Scotland were generated using Energy Performance Certificate (EPC) data geolocated by postcode and UPRN. EPC data provided information on the fuel type (wood, coal or smokeless fuel) for primary and secondary heating for individual households. Households using solid fuels were identified based on heating system descriptions and categorised by fuel type and whether the system was primary or secondary. This level of detail was not available in the Home Analytics database, which only indicated the presence or absence of solid fuel heating. Comparisons were made with the Home Analytics datasets to assess whether EPC data contained a representative picture of solid fuel use across different urban/rural and spatial classifications.

Spatially disaggregated emission maps by fuel type (wood, coal, and smokeless solid fuels) were generated using the EPC data (Figure 3.3). Like Masey et al. (2023), who based their disaggregation on the Home Analytics data, we assumed the fuel consumption of properties using solid fuel as a primary heat source to be ten times the consumption of using it as a secondary heat source. EPC records covered only ~55% of households relative to Home Analytics totals. To account for data gaps, household counts of SF usage were aggregated at the data zone level. These counts were then spatially distributed using a 1 km-resolution UK population raster (Carnell et al., 2025), creating a population-weighted proxy for domestic solid fuel use.

Table 3.6. Breakdown of number of properties using solid fuel burning in the rural / urban non-SCA / urban SCA categories in the EPC and Home Analytics datasets.

	Rural	Urban non-SCA	Urban SCA	EPC TOTAL	Home analytics
EPC primary burners	11,730	1,005	207	12,942	21,137
EPC secondary burners	75,741	19,216	8,287	103,244	156,525
Total households (primary or secondary)	82,910	19,997	8,457	103,244	-

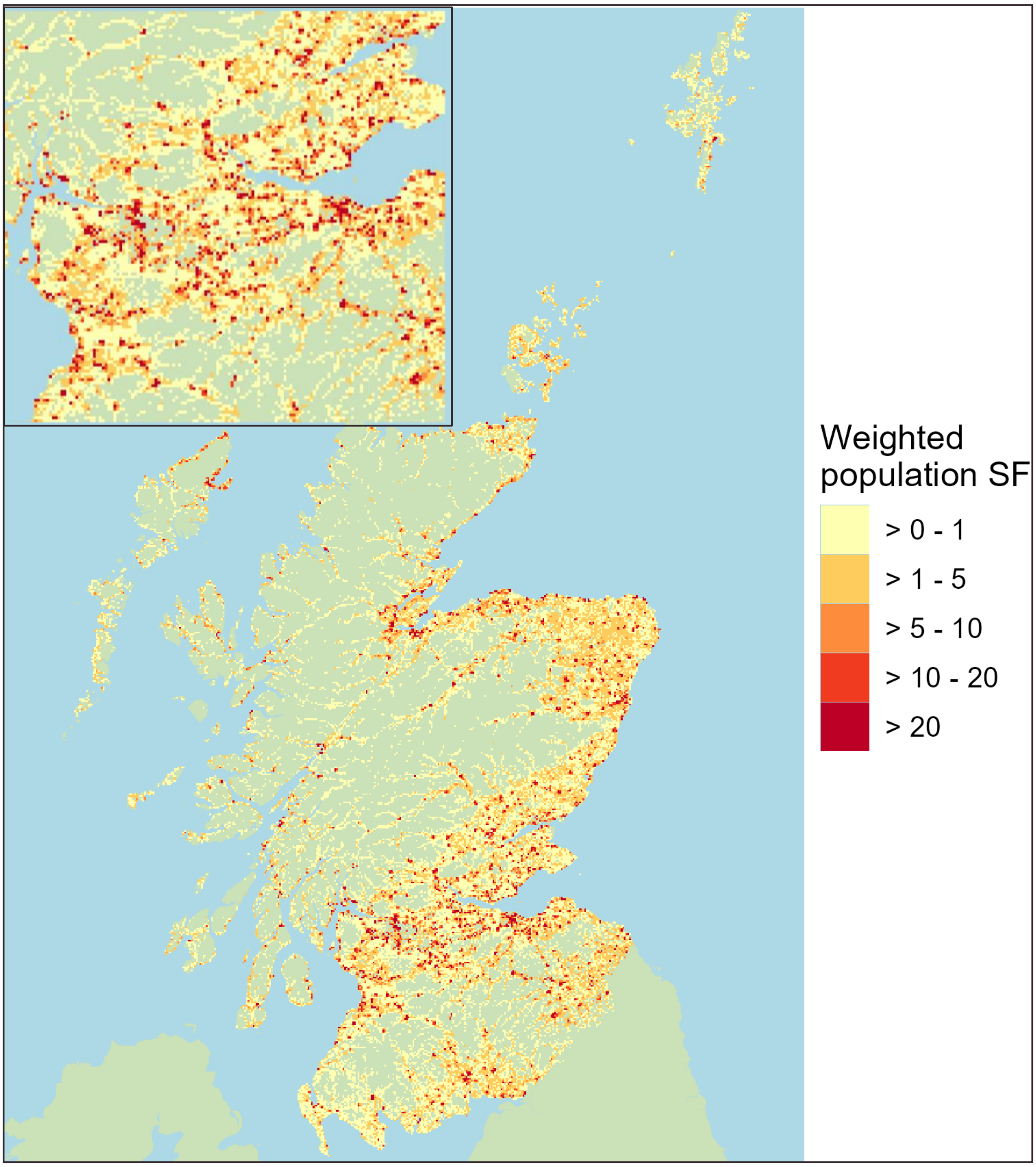

Figure 3.3. Distribution field for all burnt solid fuel (primary and secondary burners) at 1 km x 1 km resolution. The inset shows the distribution across the Central Belt in more detail.

To generate emissions maps for each fuel/species combination, the 1 km distribution maps were then scaled to match total emission values in each area (rural, urban-SCA and urban non-SCA, Table 3.8). Total emissions from domestic SF burning in Scotland were estimated by summing the contributions from wood, coal, and smokeless fuel sources.

3.2 Estimates of the domestic indoor biomass burning emissions in Scotland

Table 3.7 summarises the estimated Scottish $PM_{2.5}$ emissions from the different solid fuels once the fuel amounts have been attributed to the different urban classes and emission factors have been weighted by the distribution across the different appliance types and ages. A less detailed breakdown, lumped into three broader fuel categories, is shown in Table 3.8 for the other pollutants considered here.

Two other sources of emission totals and spatial emission fields are available for Scotland, from Masey2023 and from the UK National Atmospheric Emissions Inventory (NAEI). A comparison with Masey2023 for $PM_{2.5}$ is included in Tables 3.7 and 3.8, the latter of which also compares emissions for all pollutants with the NAEI estimates for 2021 according to the 2023 release. Total Scottish emissions from domestic solid fuel burning estimated here exceeded the Masey2023 estimate by 52% and the NAEI2021 by 22%. Emissions from wood were revised downwards reflecting the lower emission factors from the EDSFS study more than compensating for the larger fuel volume attributed to Scotland by the CAS2022/23 survey. Based on the new statistics we estimate much larger emissions from Coal and MSF, by a factor of 7.3 compared to Masey2023 and a factor of 4.4 compared to the NAEI. This reflects the increase in fuel allocation to Scotland coupled to an increase in some of the emission factors. Our NOx emissions exceed those of the NAEI2021 by a factor of 2.8 and SO_2 emissions by 67%. The domestic combustion model used for the NAEI is being updated for the 2025 release (Richmond et al., 2025).

Regarding the spatial attribution, the report of Masey et al. (2023) does not include any emission fields, only activity patterns, but they based their disaggregation on the Home Analytics statistics which should be fairly consistent with the EPC data and the underlying activity pattern should be similar. Here we additional additionally distinguish burning preferences and appliance use between smoke control and non-smoke control areas based on the CAS2022/23 survey which will somewhat change the pattern. We also include additional fuels with specific regional variations.

The NAEI provides the official reporting of UK emissions under national and international obligations. Its primary function is to estimate UK national total emissions, which are in a second step distributed to provide spatial emission fields and emissions for the four administrative regions (Mitchell et al., 2024). The NAEI methodology for the spatial disaggregation of the domestic solid fuel sector has changed greatly over the past few years. The methodology was updated when the statistics from the CAS2017/18 fuel survey were ingested. As a UK inventory, decisions were made to reflect average UK conditions which may not always be representative of Scottish conditions. For example, the current NAEI algorithm assumes that no burning occurs in flats and maisonettes across the UK, an assumption which does not reflect the possibility of solid fuel burning in Scottish tenement flats, a view that is supported by the Home Analytics dataset. It also assumes full compliance with smoke control legislation. This has led to a relocation of emissions away from urban centres and smoke control areas as exemplified by the comparison of the emission fields of the 2017 and 2023 releases (Figure 3.4).

The emission fields will continue to change when the CAS2022/23 statistics and EDSFS emission factors are implemented into the 2025 release.

Table 3.7. Total Scottish $PM_{2.5}$ emissions from different solid fuels [t]

		This study				Masey et al. (2023)		
Fuel		Total	Urban SCA	Urban nonSCA	Rural	Total	Urban	Rural
Wood logs	Wood logs, pre-dried	185.4	9.6	69.8	106.0			
	Wood logs, seasoned	100.6	5.2	39.8	55.6	511.8	225.2	286.6
	Wood logs, wet	174.8	9.0	65.9	99.9	411.0	222.2	188.8
Wood products	Wood briquettes	75.4	12.0	43.2	20.2			
	Wood pellets	2.9	2.1	0.5	0.2			
	Wood chips	4.5	3.3	0.9	0.3			
	Waste wood	103.5	5.3	44.4	53.8			
	Kindling	54.8	2.8	22.3	29.7			
	Green / garden waste	64.0	3.3	27.4	33.3			
	Total wood	**765.9**	**52.6**	**314.2**	**399.1**	**922.8**	**447.4**	**475.4**
Coal / Coal products	House coal	282.8	42.4	131.9	108.6	94.2	2.0	92.2
	Anthracite	31.4	5.1	17.9	8.4			
	Petroleum coke	37.0	5.5	17.2	14.2			
	Lignite	18.9	2.0	9.3	7.6			
	(S)MFS	351.3	55.7	201.3	94.3	15.4	3.2	12.2
	Total coal	**721.4**	**110.8**	**377.5**	**233.1**	**109.6**	**5.2**	**104.4**
Other	Peat	73.4	7.8	36.0	29.6			
	Household waste	-	-	-	-			
	Coffee logs	6.1	1.0	3.5	1.6			
	Total other	**79.5**	**8.8**	**39.5**	**31.3**			
	Total all fuels	**1566.7**	**172.1**	**731.2**	**663.4**	**1032.4**	**452.6**	**579.8**

Table 3.8. Summary of total emissions and split into distribution grids [t yr^{-1}].

Pollutant	Sector	Total	Urban SCA	Urban non-SCA	Rural
CO	Wood (prod)	10,655.7	775.4	4,270.0	5,610.2
	Coals[a]	6,501.0	936.3	3,166.0	2,398.6
	MSF	5,767.0	894.7	3,317.6	1,554.8
	Total	22,923.7	2,606.4	10,753.7	9,563.6
NOx	Wood (prod)	268.0	28.0	107.7	132.2
	Coals[a]	178.2	25.4	87.4	65.5
	MSF	153.9	23.9	88.5	41.5
	Total	600.1	77.3	283.6	239.2
VOCs	Wood (prod)	1,941.8	128.1	801.7	1,012.0
	Coals[a]	1,521.6	222.6	728.9	570.1
	MSF	949.3	151.0	543.6	254.8
	Total	4,412.7	501.6	2,074.2	1,836.9
$PM_{2.5}$	Wood (prod)	772.0	53.5	317.7	400.7
	Coals[a]	443.5	62.9	212.2	168.4
	MSF	351.3	55.7	201.3	94.3
	Total	1,566.7	172.1	731.2	663.4
SO_2	Wood (prod)	88.8	20.0	39.7	29.1
	Coals[a]	438.5	64.2	218.4	156.0
	MSF	1,462.3	225.8	842.0	394.6
	Total	1,989.7	309.9	1,100.1	579.7
BC[b]	Wood (prod)	480.8	29.6	196.0	255.2
	Coals[a]	208.4	30.7	98.3	79.4
	MSF	100.5	14.7	58.4	27.4
	Total	789.7	75.0	352.7	362.0
Cond PM[b]	Wood (prod)	471.3	35.5	192.7	243.1
	Coals[a]	278.6	39.6	133.3	105.7
	MSF	203.4	32.3	116.5	54.6
	Total	953.2	107.4	442.5	403.4

a) Including house coal, petroleum coke, anthracite, lignite and peat; b) both BC and Cond PM are subfractions of $PM_{2.5}$ and included in the total $PM_{2.5}$ emission estimate.

Table 3.9. Comparison with the emission estimates for Scotland with other inventories and estimated emission totals [t yr^{-1}].

Pollutant	Inventory	Total	Wood	Coal	MSF
$PM_{2.5}$	This study	1,566.7	772.0	443.5	351.3
	Masey et al. (2023)	1,032.4	922.8	109.6	
	NAEI 2021*	1,281.0	1100.8	171.9	8.3
NOx	This study	600.1	268.0	178.2	153.9
	NAEI 2021*	211.0	116.2	82.3	12.5
SOx	This study	1990.0	88.8	438.5	1462.3
	NAEI 2021*	1189.0	11.0	1089.4	88.6

* 2023 release.

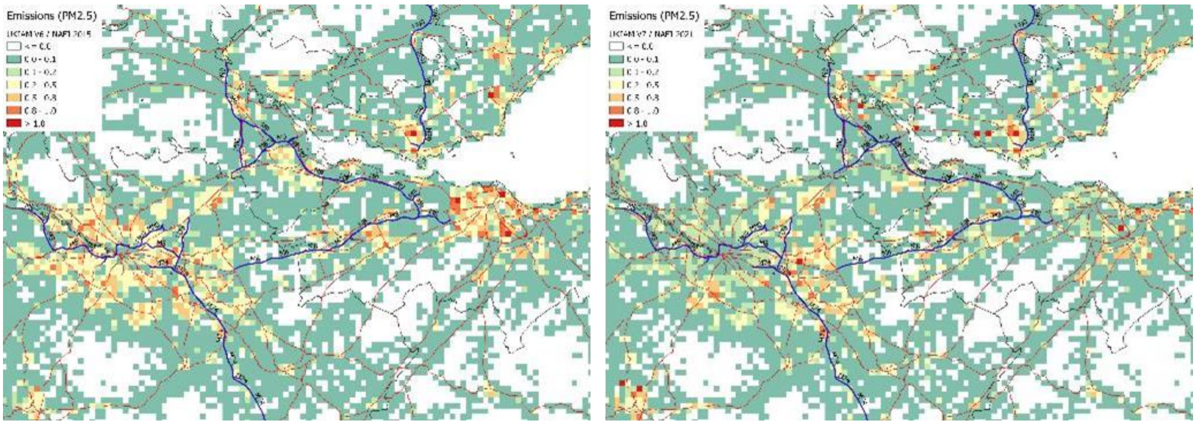

Figure 3.4. Comparison of the NAEI $PM_{2.5}$ emission fields from solid fuel burning for 2015 (2017 release, left) and 2021 (2023 release, right). The UK emission totals for the sector have been scaled to the same value for this comparison (Tim Oxley, Imperial College, pers. comm.).

4 Modelling of the contribution of domestic indoor burning to Scotland's air pollution

4.1 Description of the modelling approach

This study uses an atmospheric chemistry and transport model (ACTM) which simulates the emissions, transport, chemical transformations and deposition of a wide range of chemical components for a specific meteorological year. The advantage of a model which incorporates chemistry processes is that the approach is capable of accounting for secondary pollutants, including secondary organic aerosol (SOA) and secondary inorganic aerosol (SIA) that derive from gaseous emissions. For this calculation the model also takes into consideration the chemical interactions with other pollutants from Scottish as well as non-Scottish sources. The model generates outputs of concentrations at hourly resolution for a full model year, which enables the assessment of seasonal and diurnal variations and comparisons with long-term measurements. Primary $PM_{2.5}$ (including Condensable PM) is treated as being non-volatile in the model. It undergoes dispersion and deposition, but no chemical conversions.

The model used here is the EMEP4UK model (Gouldsbrough et al., 2024; Lin et al., 2017b; Ots et al., 2016; Vieno et al., 2016b, 2016a). EMEP4UK is a UK / Scottish application of the ACTM which is developed and used by the Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP), Meteorological Synthesizing Centre – West (MSC-W), hosted by the Norwegian Meteorological Institute (EMEP MSC-W model, (Simpson et al., 2012, 2023). EMEP is a co-operative programme under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) to solve transboundary air pollution problems across Europe and beyond. The EMEP model is applied at the European scale by MSC-W to underpin UNECE and EU negotiations on emission ceilings and the management of ecosystem impacts.

We here apply the EMEP model in a nested approach: a Scottish domain at 1 km x 1 km spatial resolution is nested within a European domain at 27 km x 27 km resolution. The WRF meteorological model which is used as input to EMEP also uses intermediate UK domains at 9 km x 9 km and 3 km x 3 km spatial resolution, as shown in Figure 4.1. Boundary concentrations for the European domain are prescribed. The model runs use UK emissions from the NAEI National Atmospheric Emissions Inventory for 2021 (2023 release) at 1 km x 1 km, where the solid fuel burning emissions for Scotland have been substituted with the emissions derived here. The emissions in the other UK nations have not been changed. As mentioned above, the NAEI for 2021 was based on the CAS2018/19 and compiled before the full results of the EFDSF study were available.

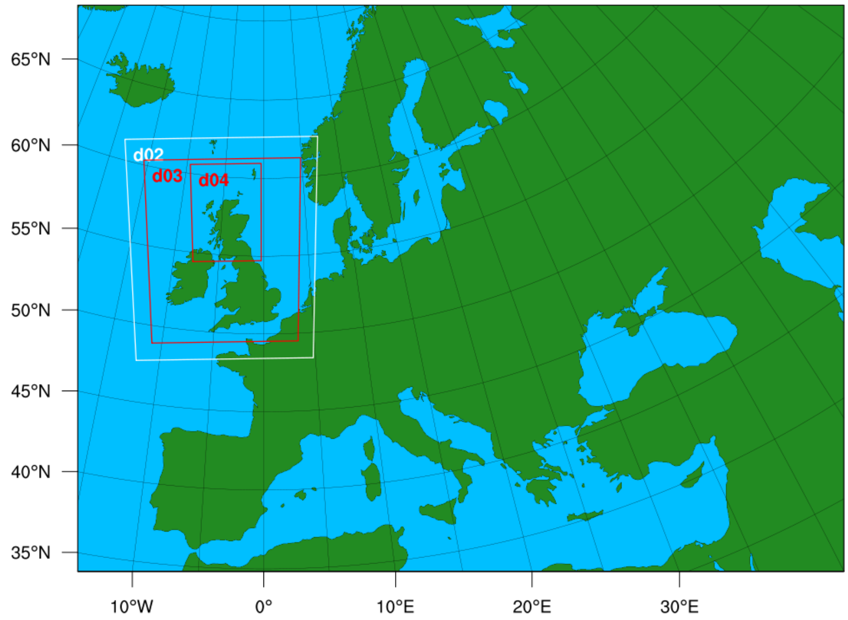

Figure 4.1. Nested model domains used for WRF and EMEP model runs: the full extent of the plot is the outermost domain (d01) with 27 km x 27 km resolution, used for both WRF and EMEP; d02 with 9 km x 9 km resolution and d03 with 3 km x 3 km are only used for WRF; d04 at 1 km x 1 km resolution is used for WRF and EMEP.

The 2022 inventory (2024 release) took on board some first gas-phase emission factors whilst the 2023 inventory (2025 release) is expected to be based on CAS2022/23 and the EDSFS and will therefore likely change significantly. Therefore, the non-Scottish emission estimates currently lag the Scottish estimates used here and this will result in some inconsistencies when it comes to the contribution of non-Scottish contributions to Scottish air pollution. Non-UK European emissions were taken from the EMEP dataset[4]. Annual total emissions were distributed in time using the standard seasonal, day-of-week and hourly profiles incorporated in the EMEP model for small scale combustion (GNFR-C; Figure 4.2). This emission sector also covers heating with gas and oil. The diurnal PM profiles are dominated by solid fuel combustion and therefore show a pattern of increased emissions in the evening. Emissions of gases have larger contribution from gas/oil combustion and this shapes the overall pattern of this sector. In addition, emissions are slightly larger for weekdays (factor 1.08) than for weekend days (factor 0.8). The temporal pattern does not account for the relationship between heating activity and actual outdoor temperature. Emissions were released into the surface layer, at heights of up to 50 m.

Meteorological input data for the EMEP model was generated for 2023 using the community Weather Research and Forecasting (WRF) model version 4.4.2 (Skamarock et al., 2019) constrained with ECMWF reanalysis data (Copernicus

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[4] www.emep.int

Climate Change Service, 2023; Hersbach et al., 2020; Simpson et al., 2012, 2023). This year was chosen to enable comparison between modelled and measured data from Edinburgh (Jan-March 2023). Whilst emissions do not normally change greatly between years, unless regulations change, the timing of concentrations at a particular site are very sensitive to meteorology, such as wind speed and wind direction. WRF version 4.6.1 was used for supplementary EMEP runs for January – March 2024, to allow comparison with the measurements during the campaign at Charlestown.

In total six model runs were conducted to assess the contributions of different fuel types to Scottish air pollution. Emissions were modified only for the domestic solid fuel sector for primary $PM_{2.5}$, primary PM_{10}, BC, CO, VOCs, NO_x and SO_2:

1. Reference run with all emissions included.
2. Rest-of-the-UK run with domestic solid fuel emissions for non-Scottish UK sources removed (i.e. England, Wales & N. Ireland).
3. No-wood run with all wood and wood product emissions removed for Scotland
4. No-coal run with all house coal/lignite/peat emissions removed for Scotland
5. No-MSF run with all MSF emissions removed for Scotland
6. No SF run with all solid fuel emissions removed for Scotland

The contribution of each category to Scottish air pollutant concentrations was quantified by calculating the difference between a scenario run and the reference run. The total contribution of Scottish domestic solid fuel burning was calculated using a run with all solid fuel emissions removed. This allows the model to capture changes in secondary particulate concentrations due to chemical interactions between emissions from different solid fuel types, which lead to non-linearities in the total particulate contributions.

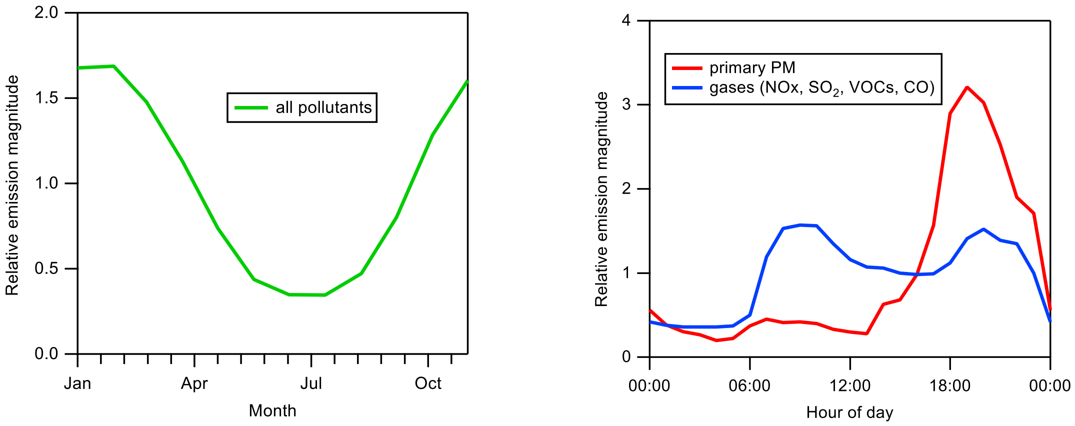

Figure 4.2. Seasonal (left) and diurnal (right) cycles in the residential combustion source sector.

4.2 Modelled pollutant concentrations

The EMEP model output files for 2023 were used to plot maps of annual average and winter average concentrations of $PM_{2.5}$, NO_x, NO_2 and SO_2. These are presented in Figures 4.3 to 4.10, with panels showing the total concentration and the contributions from non-Scottish UK domestic burning, Scottish domestic burning from all fuels combined, and from wood, coal and MSF fuels separately. The winter average maps generally show a similar spatial pattern to the annual average maps, but with slightly higher magnitudes of solid fuel contributions, due to the higher emissions at that time-of-year (Figure 4.2) and meteorological conditions being typically more conducive to the accumulation of pollutants. The $PM_{2.5}$ concentration contributions are more spatially widespread than the gaseous pollutants, which may partly reflect the influence of secondary $PM_{2.5}$ formation from gaseous emissions over moderate distances. However, the magnitude of $PM_{2.5}$ contributions from Scottish domestic solid fuel burning are generally small (<0.5 μg m^{-3}, compared with total concentrations of 4 to 8 μg m^{-3}) except in hotspots around small towns such as Dumfries, Fort William and Ayr. The contribution of non-Scottish domestic solid fuel burning is also generally small and confined to southern Scotland.

The largest absolute contribution of total Scottish domestic solid fuel sources to $PM_{2.5}$, NO_x and NO_2 was identified to occur in (-3.601°, 55.895°), between Bathgate and Livingston, collocated with the maximum contribution from MSF. The largest $PM_{2.5}$ contribution from Scottish coal burning is also predicted in the Central Belt, at (-3.049°, 55.878°), between Dalkeith and Newtongrange. In contrast, the largest $PM_{2.5}$ contribution from Scottish wood burning is modelled at (-5.088°, 56.825°), in Fort William. The maximum contribution from non-Scottish domestic solid fuel burning sources is modelled at (-3.059°, 54.997°), in Gretna near the English border, likely to be dominated by emissions from Carlisle and surroundings. The maximum total $PM_{2.5}$ contribution is collocated with the maximum primary $PM_{2.5}$ contribution for all solid fuel categories except Scottish coal burning, suggesting that the air quality impacts of domestic wood and MSF burning are predominantly local.

The statistics of the grid cell concentration enhancements are shown in Table 4.1 for $PM_{2.5}$ and Table 4.2 for NO_x and NO_2. For $PM_{2.5}$ statistics are shown for primary (emitted) $PM_{2.5}$ only, and as a sum over all modelled $PM_{2.5}$ chemical compounds. Population-weighted mean concentration (PWMC) contributions have been calculated by combining modelled concentrations with gridded population data (Carnell et al., 2025) based on 2021/2022 census data and the UK Land Cover Map.

The mean and median contributions from domestic solid fuel burning are modest compared with the total annual mean $PM_{2.5}$ concentrations and air quality standards/guidelines. The population-weighted mean $PM_{2.5}$ concentration increment from Scottish domestic solid fuel burning is more than four times larger than the simple spatial mean, this reflects the higher emissions in populated areas and predominantly local concentration impacts for this source. Wood burning has the largest population-weighted concentration contribution to $PM_{2.5}$, followed by MSF then coal, but the differences between the fuel types are relatively small for this metric.

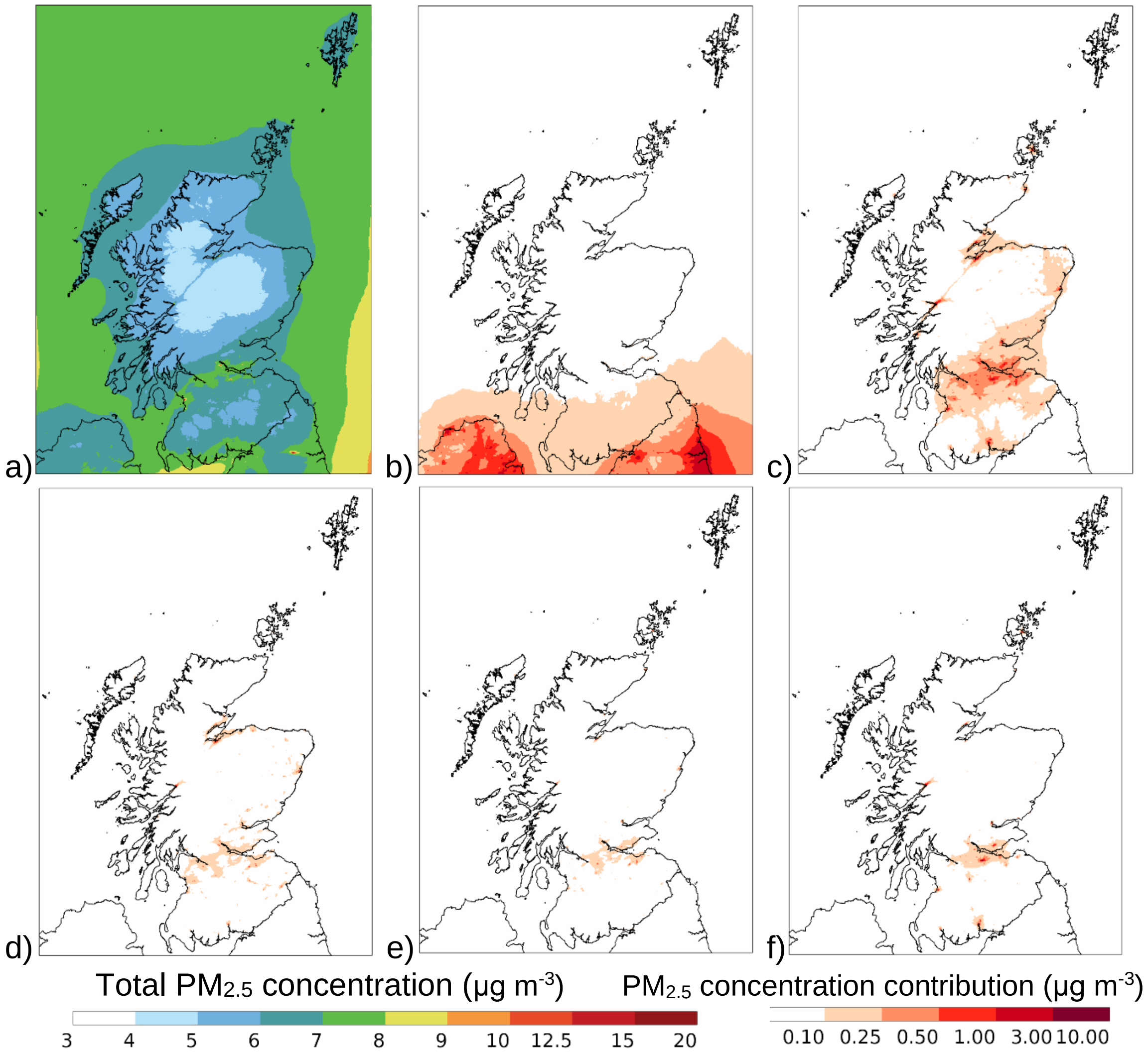

Figure 4.3. Maps of annual average $PM_{2.5}$ (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic solid fuel burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

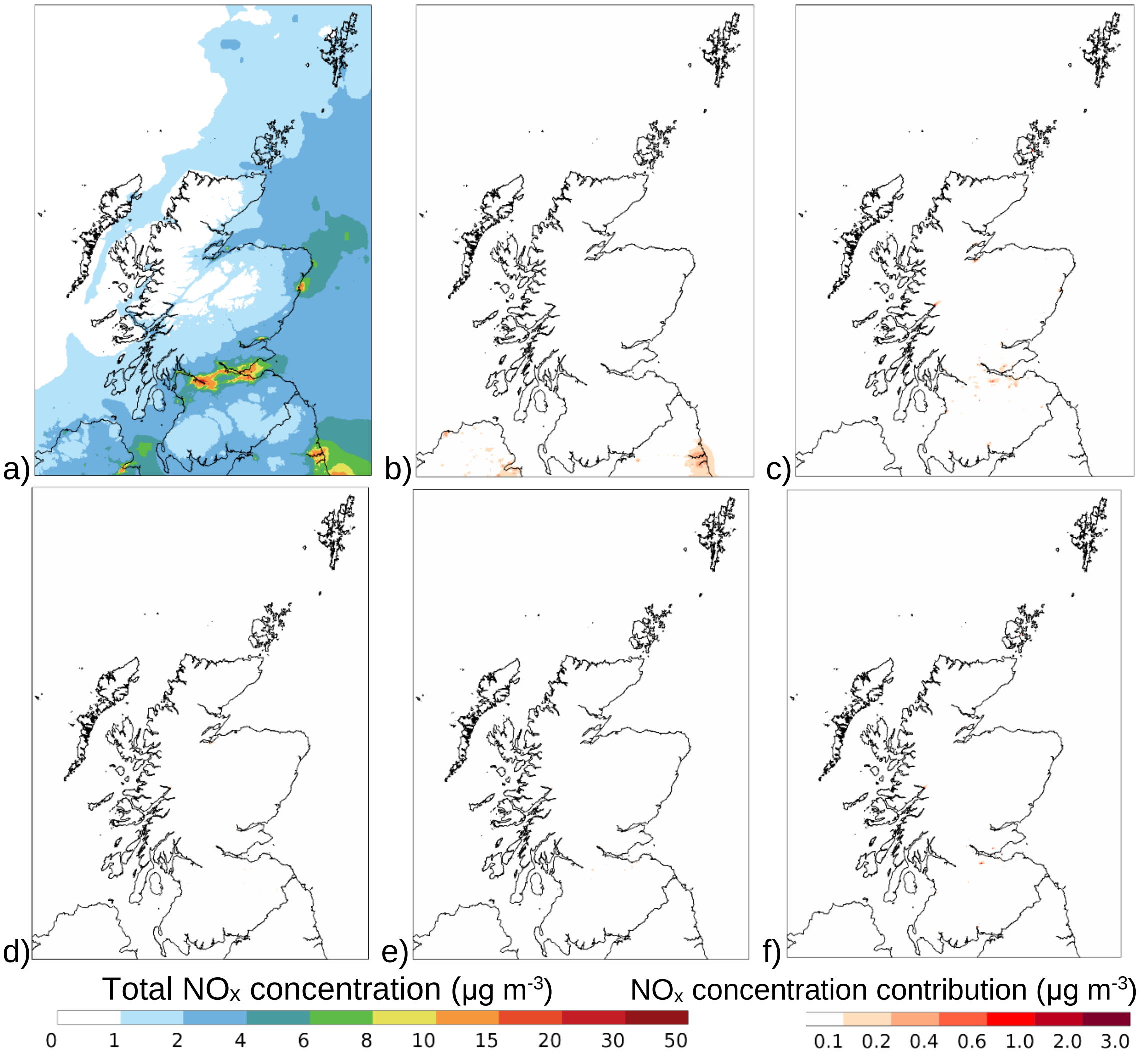

Figure 4.4. Maps of annual average NO_x concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic solid fuel burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

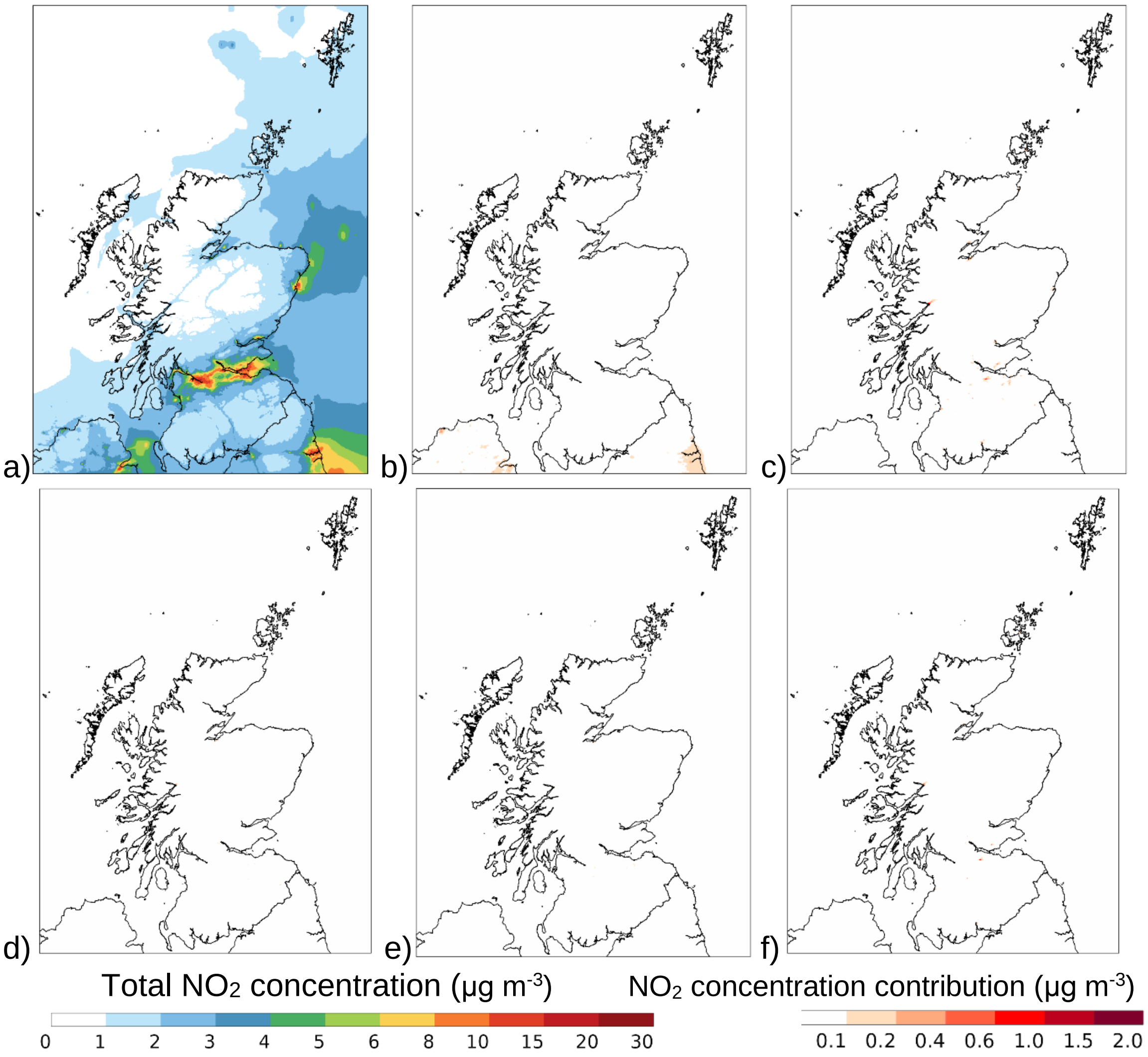

Figure 4.5. Maps of annual average NO_2 concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic solid fuel burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

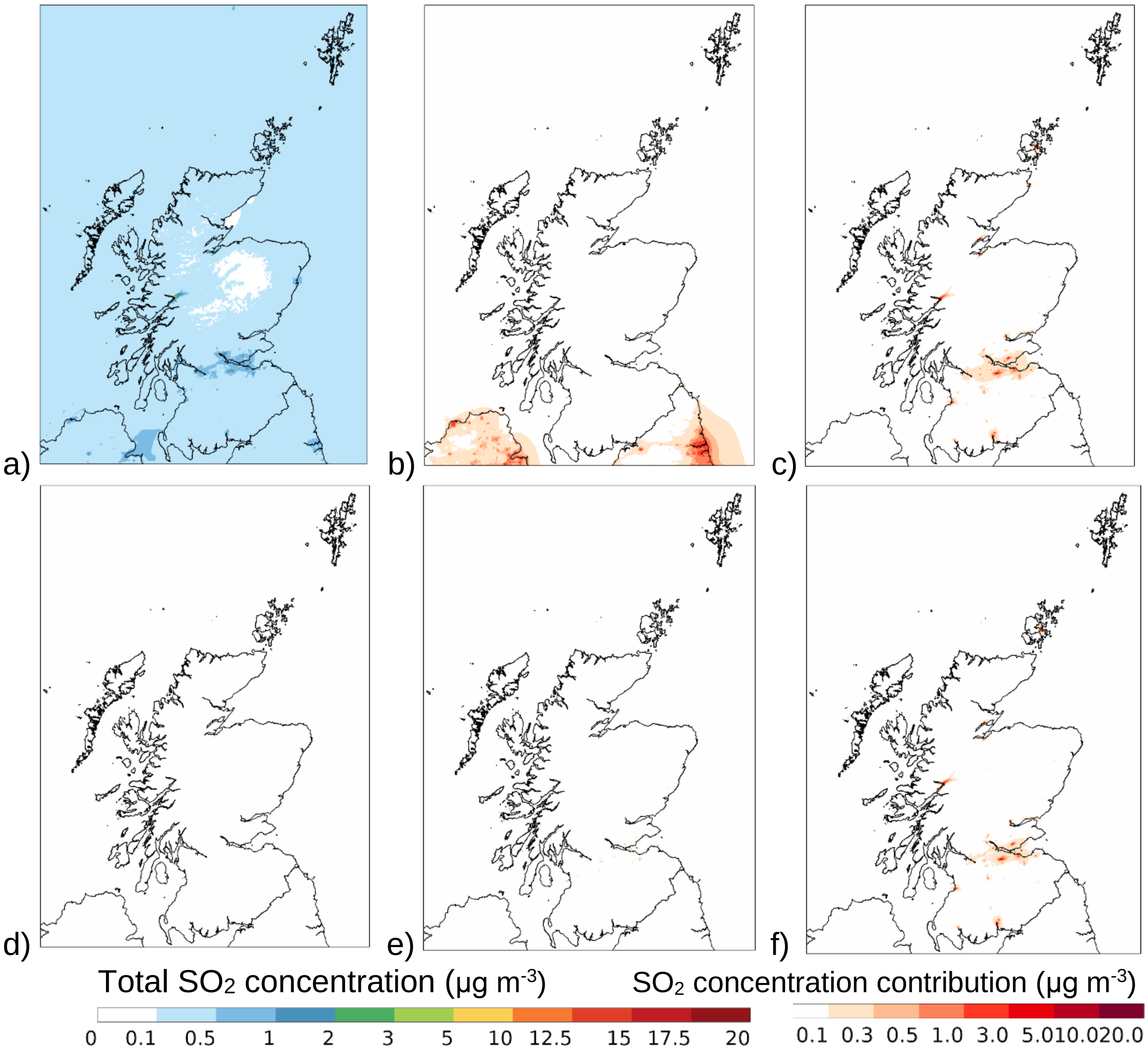

Figure 4.6. Maps of annual average SO_2 concentration (μg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic solid fuel burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

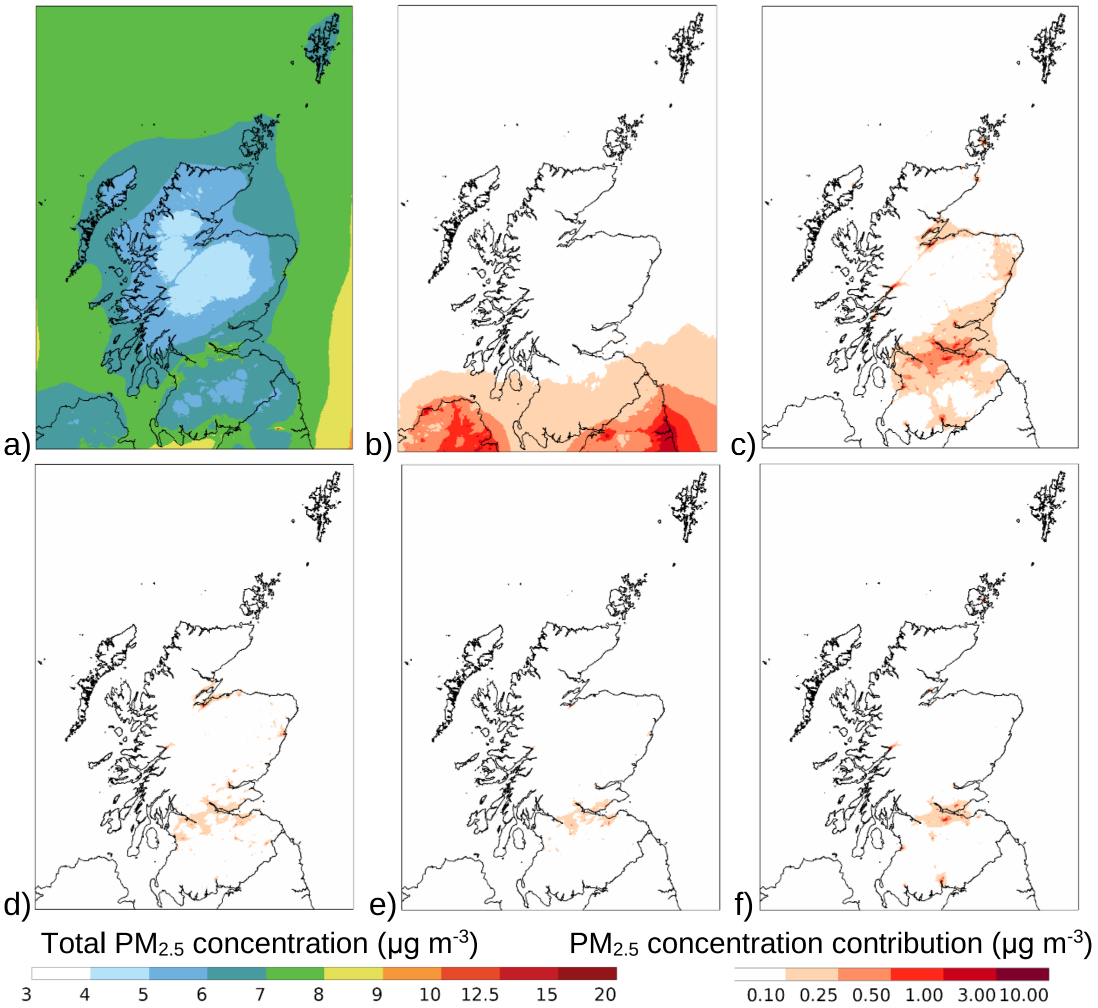

Figure 4.7. Maps of winter average $PM_{2.5}$ concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

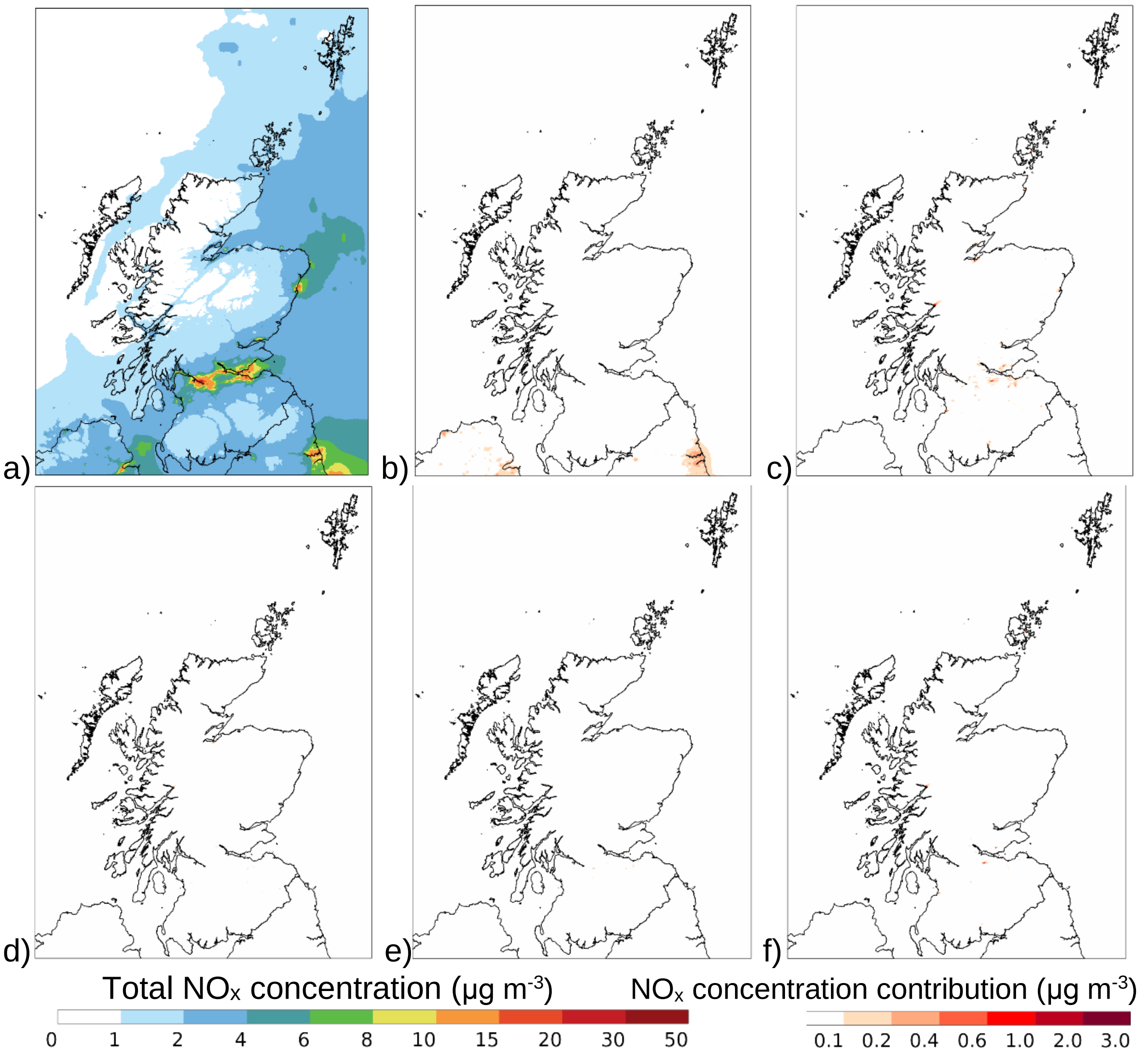

Figure 4.8. Maps of winter average NO_x concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

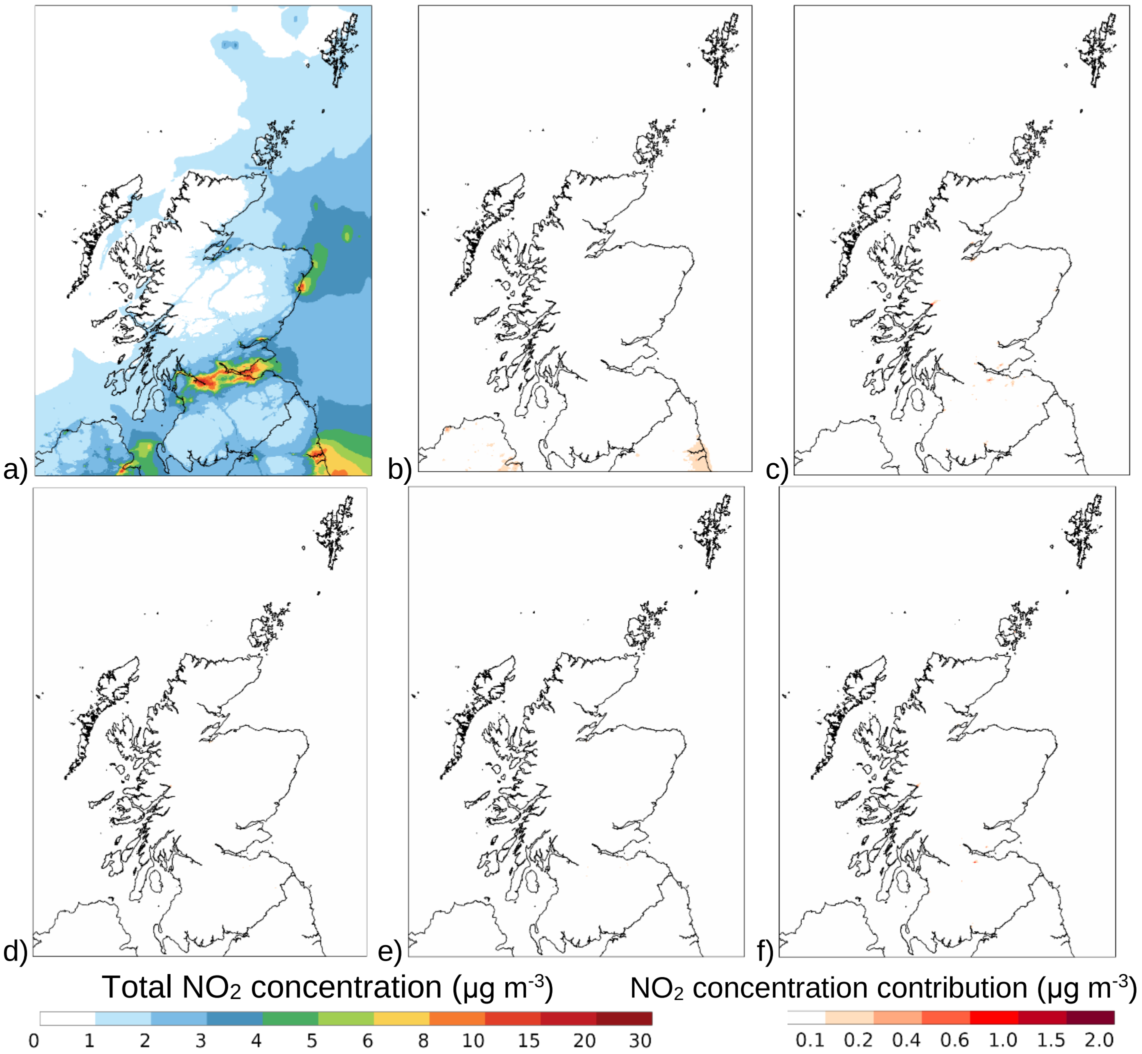

Figure 4.9. Maps of winter average NO_2 concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

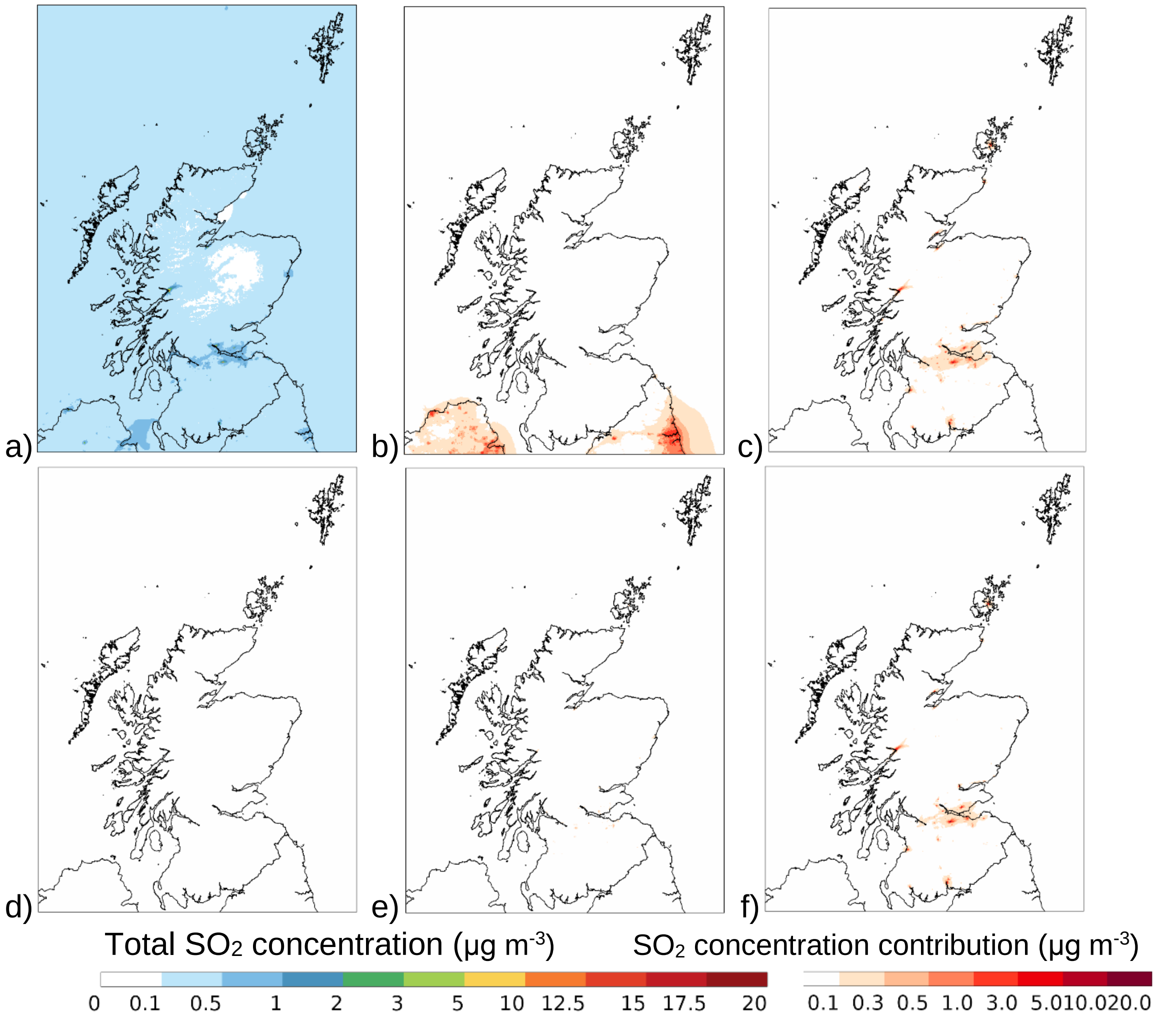

Figure 4.10. Maps of winter average SO_2 concentration (µg m^{-3}) (a) all sources; (b) non-Scottish domestic solid fuel burning; (c) all Scottish domestic burning sources, (d) Scottish domestic wood burning; (e) Scottish domestic coal burning; (f) Scottish domestic MSF burning.

Table 4.1. Statistics of modelled 2023 $PM_{2.5}$ contributions (µg m^{-3}) from domestic solid fuel burning across 1 km x 1 km grid cells (A - annual; W – winter, defined as November – March), separated into primary $PM_{2.5}$ only and total $PM_{2.5}$.

Species	Metric	All sources (reference)		Non-Scottish SF		Total Scottish SF		Scottish Wood		Scottish Coal		Scottish MSF	
		A	W	A	W	A	W	A	W	A	W	A	W
Primary $PM_{2.5}$	Mean	0.404	0.479	0.029	0.040	0.056	0.091	0.028	0.046	0.015	0.024	0.013	0.021
	PWMC	1.19	1.59	0.040	0.056	0.255	0.435	0.108	0.184	0.079	0.135	0.068	0.116
	Median	0.313	0.352	0.020	0.029	0.031	0.046	0.016	0.023	0.007	0.011	0.006	0.009
	Min	0.140	0.135	0.008	0.008	0.003	0.005	0.002	0.002	0.001	0.001	0.001	0.001
	Max	11.0	10.1	0.228	0.350	3.59	6.02	1.40	2.41	0.680	1.11	3.40	5.70
	Stdev	0.267	0.353	0.022	0.028	0.074	0.127	0.032	0.055	0.022	0.038	0.033	0.055
Total $PM_{2.5}$	Mean	5.84	6.22	0.064	0.094	0.086	0.140	0.036	0.057	0.022	0.035	0.028	0.047
	PWMC	7.11	7.95	0.088	0.133	0.359	0.608	0.129	0.217	0.104	0.176	0.124	0.213
	Median	5.82	6.16	0.048	0.074	0.053	0.080	0.021	0.031	0.012	0.018	0.017	0.026
	Min	4.32	4.57	0.009	0.011	0.003	0.004	0.002	0.002	0.001	0.000	-0.001	-0.002
	Max	17.4	17.0	0.388	0.603	5.76	9.62	1.54	2.63	0.802	1.31	5.52	9.21
	Stdev	0.783	0.794	0.045	0.063	0.103	0.177	0.038	0.064	0.028	0.048	0.054	0.091

Table 4.2. Statistics of 2023 NO_x and NO_2 contributions ($\mu g\ m^{-3}$) from domestic solid fuel burning across 1 km x 1 km grid cells (A - annual; W – winter, defined as January – March and November - December)

Species	Metric	All sources (reference)		Non-Scottish SF		Total Scottish SF		Scottish Wood		Scottish Coal		Scottish MSF	
		A	W	A	W	A	W	A	W	A	W	A	W
NOx	Mean	1.80	2.07	0.002	0.003	0.015	0.025	0.007	0.012	0.004	0.007	0.004	0.006
	PWMC	8.06	11.1	0.002	0.004	0.082	0.142	0.032	0.055	0.027	0.046	0.024	0.042
	Median	1.31	1.37	0.001	0.002	0.007	0.012	0.003	0.006	0.002	0.003	0.001	0.002
	Min	0.403	0.308	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000
	Max	30.0	47.4	0.026	0.042	1.55	2.56	0.478	0.802	0.274	0.435	1.500	2.46
	Stdev	1.65	2.33	0.002	0.004	0.025	0.043	0.009	0.016	0.008	0.013	0.013	0.022
NO_2	Mean	1.61	1.91	0.002	0.003	0.013	0.022	0.006	0.010	0.004	0.006	0.003	0.005
	PWMC	6.42	8.74	0.002	0.003	0.060	0.099	0.024	0.040	0.019	0.032	0.017	0.027
	Median	1.19	1.31	0.001	0.002	0.007	0.011	0.003	0.005	0.002	0.003	0.001	0.002
	Min	0.364	0.301	-0.002	-0.004	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000
	Max	18.9	27.9	0.022	0.038	0.922	1.55	0.353	0.593	0.189	0.323	0.880	1.40
	Stdev	1.33	1.86	0.002	0.003	0.019	0.033	0.008	0.013	0.006	0.010	0.010	0.016

The population weighted mean enhancement due to total Scottish domestic indoor solid fuel burning sources was estimated to be 0.26 $\mu g\ m^{-3}$ at the annual mean and 0.44 $\mu g\ m^{-3}$ during winter. Non-Scottish UK solid fuel sources add another 0.04 and 0.06 $\mu g\ m^{-3}$. The total solid fuel burning component therefore represents 6.3% of the total PWM $PM_{2.5}$ PWMC of 7.11 $\mu g\ m^{-3}$ at the annual average and 9.3% of the winter PMWC of 7.95 $\mu g\ m^{-3}$. The spatial maximum values of modelled $PM_{2.5}$ contributions from domestic solid fuel burning are more substantial. MSF burning dominates the overall modelled maximum annual average $PM_{2.5}$ contribution from domestic solid fuel, whereas wood burning has the highest mean and median contribution. The minimum $PM_{2.5}$ concentration contributions from domestic MSF burning are slightly negative, this indicates that removing the MSF emissions leads to a small increase in modelled secondary $PM_{2.5}$ concentrations in some locations and reflects interactions through atmospheric chemistry.

The mean winter $PM_{2.5}$ concentration increment from all Scottish solid fuel burning was modelled to be 62% higher than the annual equivalent. This includes both increased emissions in winter months and reduced dispersion under cold and low wind speed meteorological conditions. The model does not currently take into account short-term dependencies of domestic solid fuel burning activity on meteorological conditions, which could increase the winter concentration contributions further.

For NO_2, the other compound of concern for human health, the PWMC contribution from all solid fuel burning sources is small, 0.062 µg m^{-3} (1.0%) at the annual mean and 0.102 µg m^{-3} (1.2%) in winter, with a maximum contribution of 0.92 µg m^{-3}.

The breakdown of the annual mean $PM_{2.5}$ enhancements across Scotland from non-Scottish, total Scottish and wood, coal and MSF sources into chemical components is shown in Figure 4.11. These results show the fractions that would have been missed by other modelling approaches or by the different measurement methodologies. Modelling approaches that only consider the primary emissions (such as that of Masey et al. (2023) would have missed 18% of the total contribution. None of the measurement approaches discussed in Section 2.1 would have identified the secondary inorganic contribution (NH_4^+ + NO_3^- + SO_4^{2-}) as being dependent on solid fuel burning, accounting for 24% of total $PM_{2.5}$. For the solid fuel pollutants imported into Scotland about half are secondary.

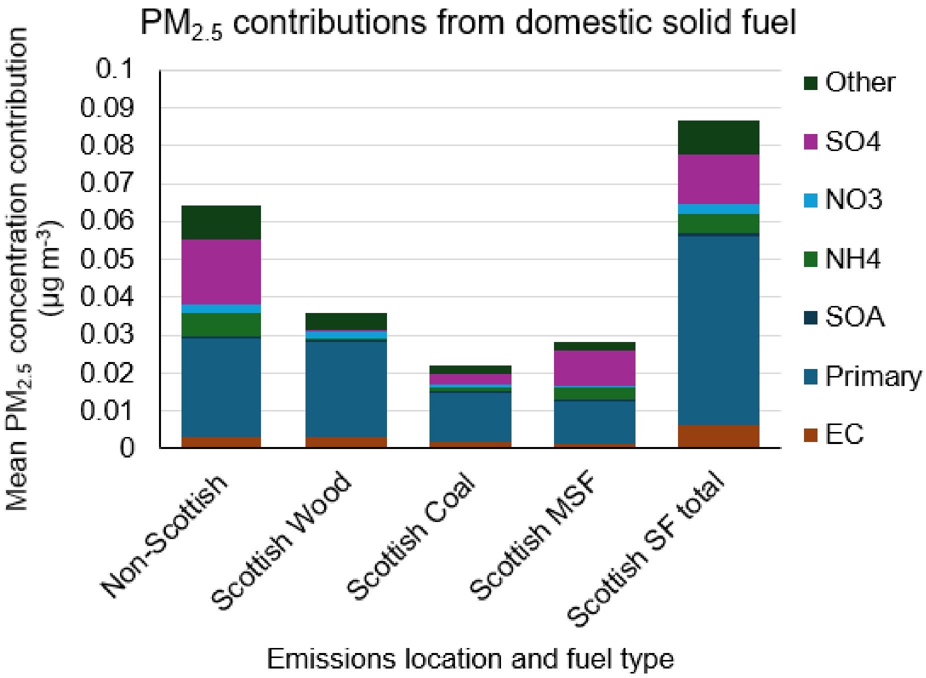

Figure 4.11. Stacked bar chart showing the contribution of different $PM_{2.5}$ components (µg m^{-3}) to mean annual $PM_{2.5}$ increments in Scotland from domestic solid fuel burning. EC: Elemental carbon; Primary: other primary $PM_{2.5}$; SOA: secondary organic aerosol; Secondary inorganic species: NH4 (ammonium, NH_4^+), NO3 (nitrate, NO_3^- including 27% coarse nitrate), SO4 (sulphate, SO_4^{2-}); Other components of $PM_{2.5}$ including particle-bound water at 50% relative humidity.

4.3 Model / measurement comparison of solid fuel $PM_{2.5}$

The model run period was extended until March 2024 to enable comparison of model results with both measurement campaigns.

The upper panels of Figures 4.12 and 4.13 show the comparison of the stacked modelled time-series of the elemental carbon (EC) originating from wood burning (wb), coal burning (cb) and MSF burning (MSF) compared with the BC component attributed to solid fuel burning by the aethalometer analysis (eBC_{wb}).

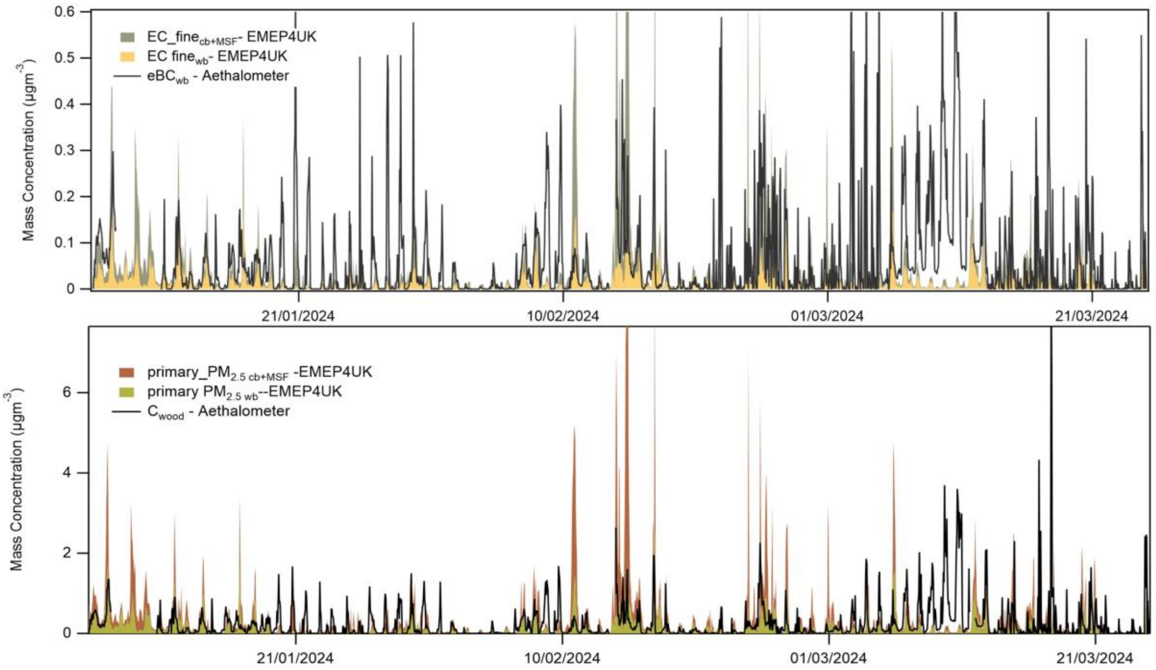

Figure 4.12. Comparison of measured and modelled time-series (hourly values) at Charlestown (Jan-March 2024). Top graph: modelled elemental carbon (EC) from the combustion of various solid fuels (wood, coal+MSF-solid colours) and solid fuel component (eBC_{wb}) of measured eBC (black line). Bottom graph: modelled primary $PM_{2.5}$ from the combustion of solid fuels (wood, coal+MSF-solid colours) and c_{wood} obtained by aethalometer measurements.

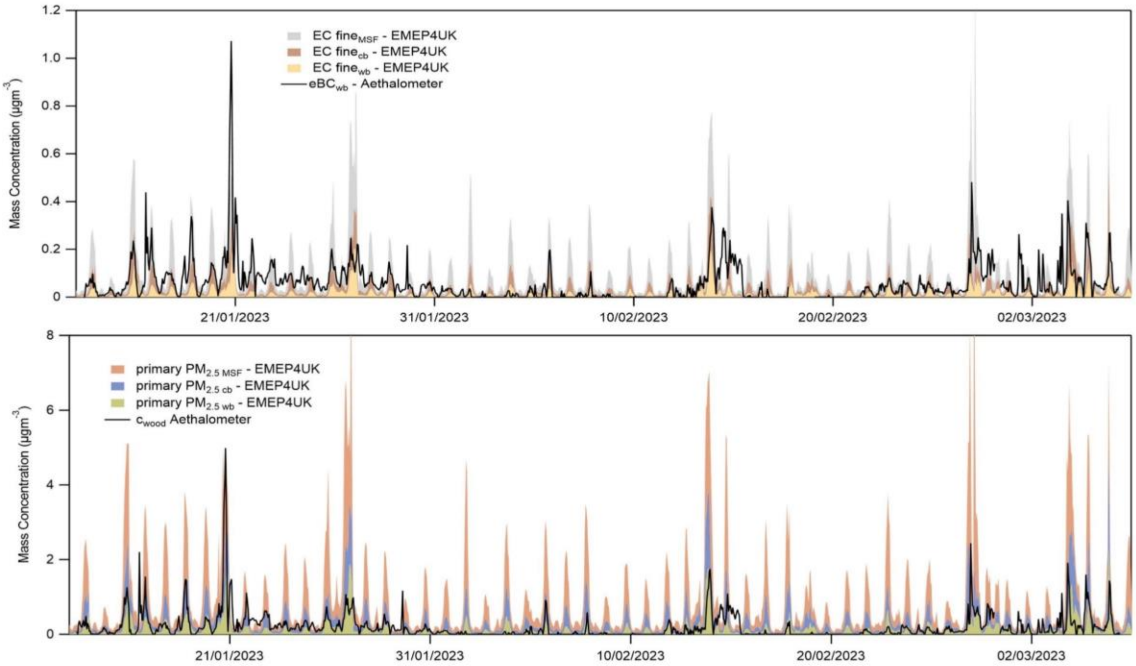

Figure 4.13. Comparison of measured and modelled time-series (hourly values) at Edinburgh (Jan-March 2023). Top graph: modelled elemental carbon (EC) from the combustion of various solid fuels (wood, coal, MSF-solid colours) and solid fuel component (eBC_{wb}) of measured eBC (black line). Bottom graph: modelled primary $PM_{2.5}$ from the combustion of solid fuels (wood, coal, MSF-solid colours) and c_{wood} obtained by aethalometer measurements.

Strictly speaking, EC and BC refer to two different physical concepts, EC referring to the carbon fraction that is refractory (thermally stable) and BC being defined by its light absorbing properties. Whilst the EMEP model reports EC, the emission factors used here were actually measured as BC and therefore the two entities are therefore consistent. The lower panels of Figures 4.12 and 4.13 show a comparison of the stacked modelled primary $PM_{2.5}$ components due to the three fuel categories with the measure of c_{wood} derived from the aethalometer. These are the entities we would expect to be comparable. Overall, there is mostly good agreement between measured and modelled temporal pattern of concentrations, with high concentration periods and low concentration periods, suggesting that meteorological influences (wind speed; transport patterns) on concentrations are well reproduced. There are some periods of disagreement, such as 14-15 Feb 2023 and 8-11 Mar 2024 when the model fails to predict the elevated concentrations that were measured.

Overall biases in the magnitude of modelled concentrations compared with measurements are easier to see in the averaged diurnal cycles presented in Figures 4.14 and 4.15. In both cases the top panels show the averaged diurnal cycles of the data shown in Figures 4.12 and 4.13, whilst the bottom panel additionally shows the consistency between the two independent measurement approaches of the AMS (BBOA, CCOA) and the aethalometer (c_{wood}, eBC_{wb}), in relation to the total primary $PM_{2.5}$ concentration from all fuels predicted by the model. c_{wood} would be expected to represent the sum of BBOA and the eBC$_{wb}$ which is refractory and cannot be detected by the AMS. The degree to which c_{wood} also includes coal burning organic aerosol is less clear (see above).

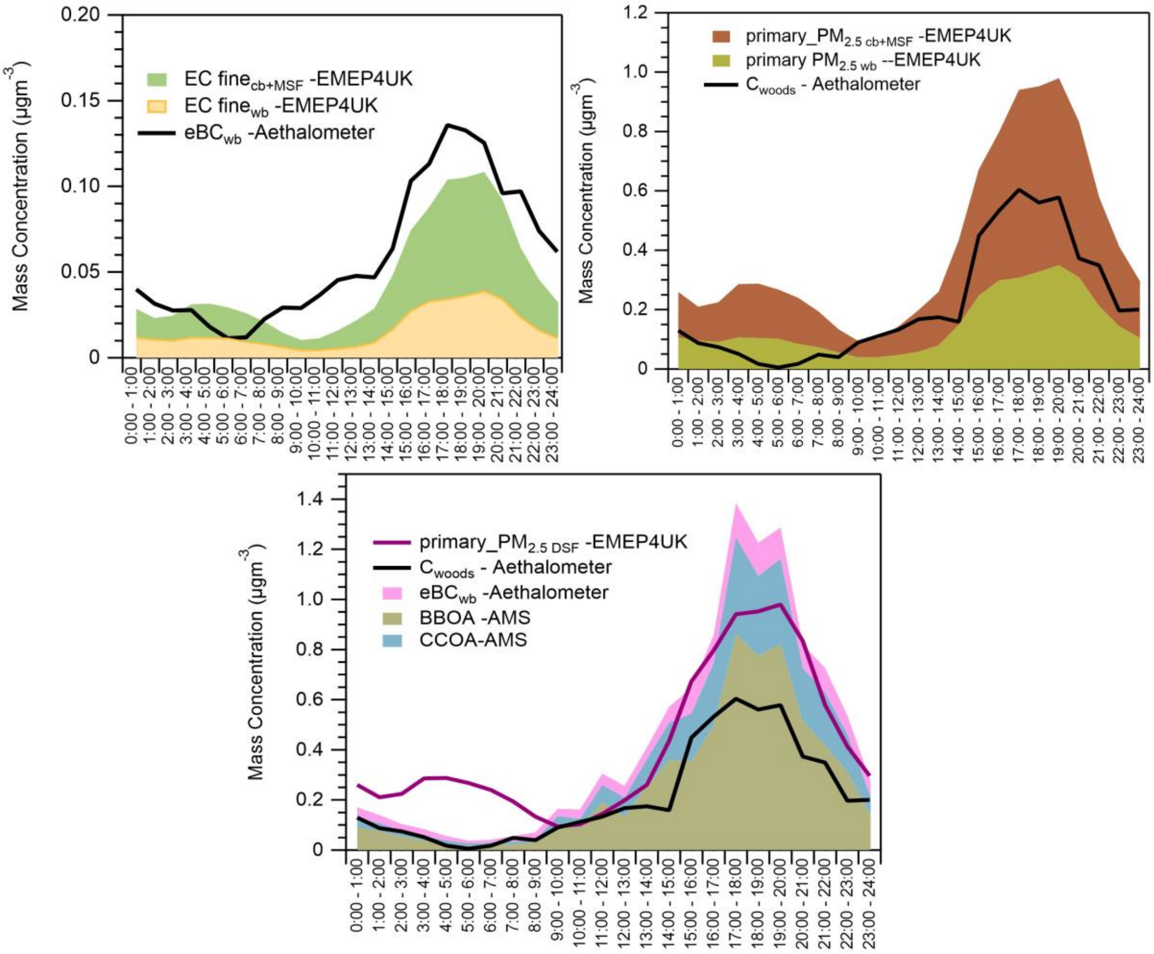

Figure 4.14. Average diurnal patterns of modelled and measured primary PM$_{2.5}$ components from solid fuel burning for the campaign at Charlestown, Fife.

What is already visible in the time-series but becomes clearer in the average diurnal cycles is that the model represents the measured concentrations in the rural / village setting of Charlestown, Fife, really quite well (Fig. 4.14). On average BC/EC concentrations are somewhat underestimated, but this comparison will be strongly influenced by the inability of the model to reproduce the 8-11 March period. The model predicts somewhat higher organic aerosol concentrations on average than the aethalometer's c_{wood} estimate, but somewhat lower concentrations than the sum of the AMS BOA, CCOA and refractory eBC$_{wb}$ component. At this site the AMS clearly derives larger concentrations than the aethalometer. One explanation is that c_{wood} may not fully capture the contribution from coal fuels. The good agreement between modelled and measured estimates seems to some extent fortuitous because in a village setting the measurement is highly dominated by only a few nearby burners and it is difficult for the emissions inventory to correctly represent small number statistics. In terms of diurnal pattern the model does not reproduce the morning peak contained in the measurements. Presumably, in this village setting more people rely on solid fuel as their primary heating source, lighting fires in the morning, than is reflected in the average diurnal cycle used for the emissions (Fig. 4.2).

The agreement between modelled and measured concentrations is much poorer in Edinburgh. Here the BC_{wb} estimated using the default AAE of Font et al. (solid line) is approximately twice as large as the BC the model predicts to originate from wood alone, but significantly smaller than the BC predicted to originate from all three solid fuel types. Note that the order of stacking is arbitrary. The same observation applies to total primary $PM_{2.5}$ where the model overestimates concentrations by an even larger margin than for BC. Based on the findings at Charlestown one explanation could be that c_{wood} does not fully reflect the coal based fuels. However, at the Edinburgh site there was extremely close agreement between c_{wood} and the sum of the AMS-based BBOA and eBC_{wb} and no coal factor (CCOA) was identified in the PMF analysis of the AMS data. There are potential factors that could explain the discrepancy which will be discussed in Section 5.1, but overall it appears that the emission inventory overestimates solid fuel burning emissions in Edinburgh, probably from coal and coal products (including MSF).

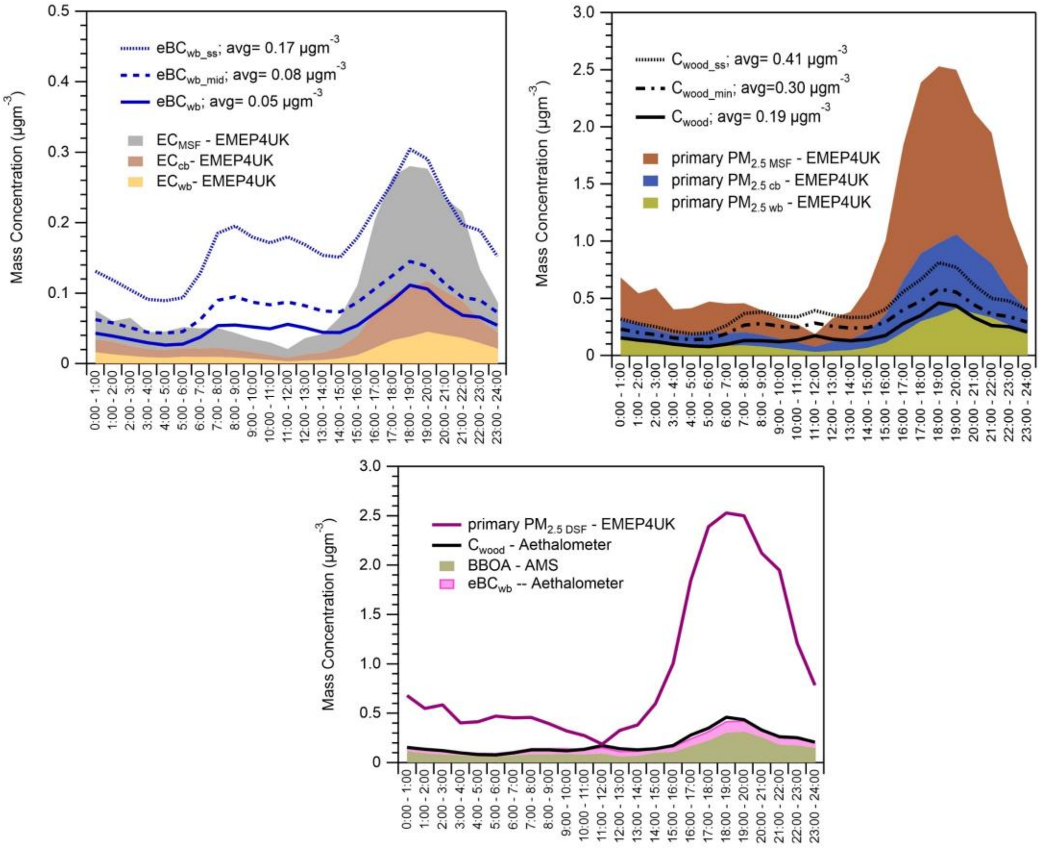

Figure 4.15. Average diurnal patterns of modelled and measured primary $PM_{2.5}$ components from solid fuel burning for the campaign at Infirmary Street, Edinburgh.

The model / measurement comparison for the whole year of 2023 is shown for the long-term BC monitoring sites at Glasgow Townhead and Auchencorth Moss in Figures 4.16 and 4.17, respectively. In Glasgow, during winter, the model predicts somewhat lower BC contributions than were derived from the aethalometer (based on Font's AAE). During summer it predicts much smaller concentrations. The modelled component does not include outdoor burning (e.g. of garden refuse, and of charcoal for barbeques), muirburn or (global) wildfires which will have contributed to the BC. It is also possible that because of the relatively low temperatures, in Scotland more people light indoor fires outside the winter burning season than is reflected in the seasonal emission profile implemented in EMEP (Fig. 4.2). During winter in Glasgow, c_{wood} is well reproduced by the model and emissions from all three fuels are needed to close the budget of c_{wood}. In summer, there is again a contribution to c_{wood} that is not modelled, similar to the BC comparison.

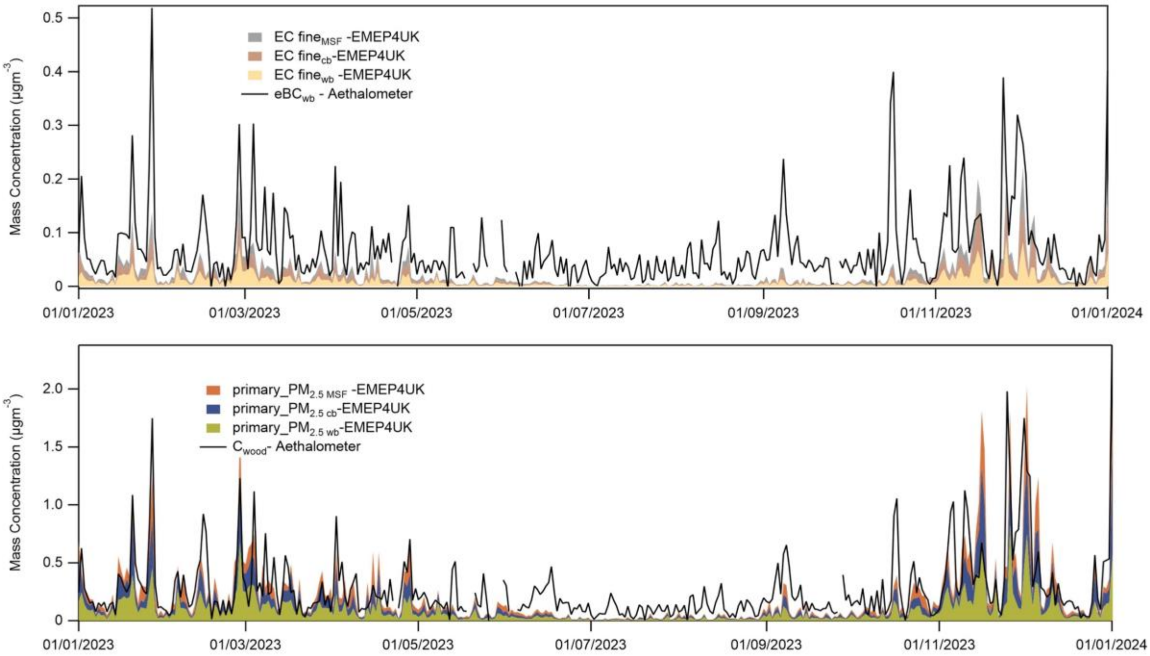

Figure 4.16. Comparison of modelled and measured time-series (daily values) at Glasgow Townhead in 2023. Top graph: modelled elemental carbon (EC) from the combustion of various solid fuels (wood, coal, MSF-solid colours) and solid fuel component (eBC_{wb}) of measured eBC (black line). Bottom graph: modelled primary $PM_{2.5}$ from the combustion of solid fuels (wood, coal, MSF-solid colours) and c_{wood} obtained by aethalometer measurements.

At the more remote site at Auchencorth Moss, 20 km S of Edinburgh, again the model does a reasonable job in reproducing the BC and c_{wood} components from all three fuel types. There are some individual periods where the model does not reproduce peaks in the measurements and some periods where the model overestimates the contribution. This may in part be due to the fact that our temporal emission pattern is not linked to actual meteorology, but could also be impacted by individual local sources. Auchencorth Moss is also likely to see episodes of muirburn in the vicinity (e.g. Pentland Hills; Moorfoot Hills; Southern Uplands).

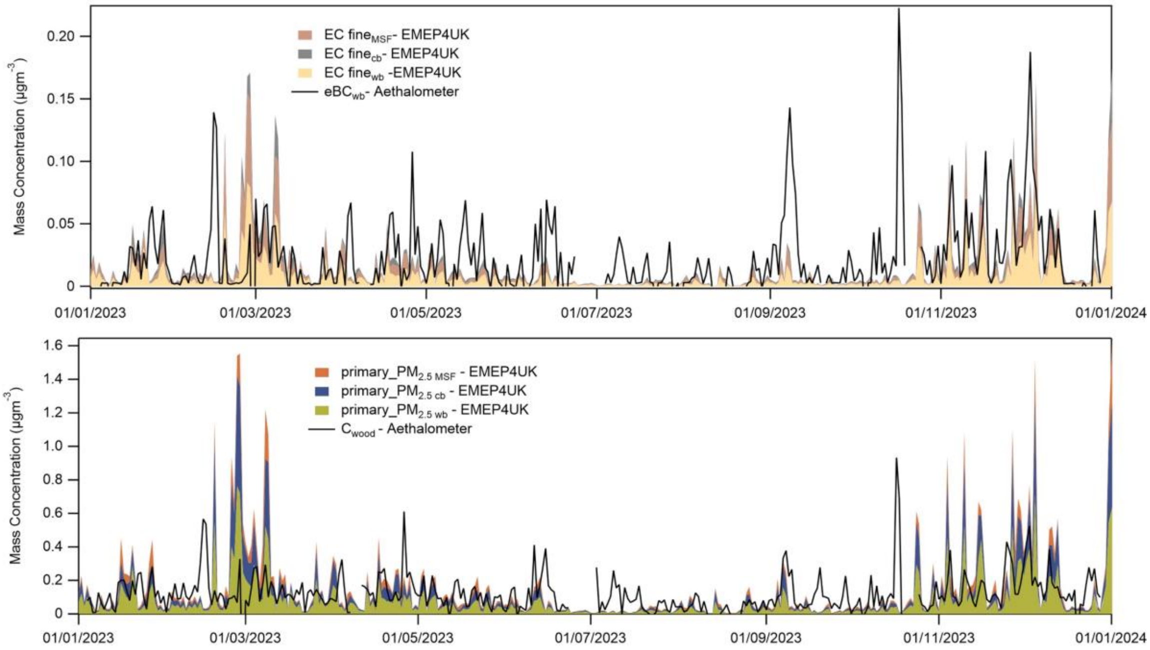

Figure 4.17. Comparison of modelled and measured time-series (daily values) at Auchencorth Moss in 2023. Top graph: modelled elemental carbon (EC) from the combustion of various solid fuels (wood, coal, MSF-solid colours) and solid fuel component (eBC_{wb}) of measured eBC (black line). Bottom graph: modelled primary $PM_{2.5}$ from the combustion of solid fuels (wood, coal, MSF-solid colours) and c_{wood} obtained by aethalometer measurements.

Overall, the model (and by inference the emission inventory) reproduces measured concentrations well at three of the four sites, the exception being Edinburgh. In Edinburgh, the modelled contribution from burning of coal and coal products seems larger than is supported by the measurements. Uncertainties will be discussed in more detail in the following section. It seems that the emission mapping algorithm allocates too much coal and MSF to Edinburgh, presumably reflecting the number of EPC certificates stating solid fuel burning appliances. Possibly, these appliances are used less often or over shorter durations than the allocation process assumes or with cleaner fuels. EPC certificates are usually issued when a property is built, sold or upgraded. It is possible that the prevalence of solid fuel burning appliances in this subset of properties is unrepresentative for the full existing housing stock.

It is unclear whether, if this amount of coal / MSF were to be attributed to other parts of the country instead, this would then lead to the model overestimating the measured contribution at the other sites.

5 Discussion

5.1 Uncertainties

5.1.1 Uncertainties in measurement approaches

Measurement approaches aimed at quantifying the PM component linked to solid fuel burning are subject to significant uncertainties and no standardised procedures exist. Fuel mixes, aerosol properties and the chemical composition of solid fuel derived $PM_{2.5}$ vary between locations and during transport away from the source. One key source of uncertainty in the measurements arises from the Aerosol Mass Spectrometer (AMS) positive matrix factorisation (PMF) analysis, which decomposes complex datasets into contributing source profiles and their temporal contributions. The results are sensitive to the number of factors selected and the rotational ambiguity of the solution. This can lead to variability in source identification and quantification, especially when source profiles overlap or are not very well resolved, like in the case of wood burning and coal combustion during our study in Edinburgh. Another limitation of the technique is that it assumes constant source profile through the duration of the analysis – an assumption which might not always reflect reality.

The aethalometer model, which is commonly used to estimate the black carbon from fossil fuel and from wood burning, carries its own uncertainties. A critical parameter in this method is the AAE coefficient, which is used to differentiate between combustion sources based on their spectral absorption characteristics. These values can vary depending on environmental conditions and on the specific aethalometer wavelengths used to obtain them. This variability complicates the separation of wood and coal combustion sources, particularly in the absence of ancillary tracers such as potassium and levoglucosan, which can provide additional constraints for source attribution. In the case of Edinburgh for example, the ratio of wood combustion (eBC_{wb}) to traffic component (eBC_{tr}) in the black carbon can vary from 16%-84% to 50%-50% using different sets of AAE coefficients, with implications for the quantification of the source contribution to total PM mass. Associated uncertainties and ranges in possible results were explored in Sections 2.3.3 and 2.4.3.

Furthermore, the calculation of c_{wood} relies on an empirical scaling factor such as the C_2 coefficient, which can vary significantly between locations but which also depends on the type of organic matter measurements and on the aethalometer wavelengths used to derive it. Here we used a value from a previous UK-based study on wood smoke pollution rather than a C_2 derived for the field campaigns. Some calibrations for the coefficients used in converting absorption across wavelengths into c_{wood} concentration have been referenced to PMF analysis results of aerosol mass spectrometer data and in these situations the uncertainties around AMS PMF discussed above propagate into the aethalometer-derived estimates.

5.1.2 Uncertainties in emission estimates

Whilst better data are continuing to become available to help estimate emissions from fuel burning the emission estimates continue to be subject to significant uncertainty, both in terms of magnitude and geographical allocation. Uncertainties may be distinguished into (i) the total fuel allocation to Scotland, (ii) the geographical distribution within Scotland, (iii) distribution across appliance types, and (iv) the emission factors.

Fuel amounts are easier to constrain at the UK level (e.g. through national sales statistics) than they are for Scotland. Whilst total UK fuel amounts have stayed relatively consistent, total fuel amounts allocated to Scotland have been revised significantly between the CAS2018/19 and the CAS2022/23 (Defra, 2024b), with an increase of other coal products (incl. MSF) being the most obvious. The fuel volume assessment (Defra, 2024b) raises significant concerns about CAS interviewees having misunderstood the different coal classifications, especially the distinction between house coal and charcoal. This may have led to errors; future CAS studies will attempt to improve the questionnaire in this respect. The CAS is a UK wide study and according to the CAS2022/23, Scotland which accounts for 8.1% of the UK’s population, accounts for 11.5% of the coal/coal product consumption and 12.4% of the wood/wood product use (CAS2018/19 suggested only 2.4% and 6.7%, respectively). A fairly small re-allocation of the total UK consumption therefore leads to a large change for Scotland. Once the number of respondents is broken down into the four nations, urban vs non-urban, urban SCA vs urban non-SCA and by appliance and preferred fuel type, the statistics for Scotland become very limited. The limited sample size not only affects the allocation of fuel to Scotland but also the spatial allocation within Scotland. A larger Scotland-specific fuel survey would be useful for providing a statistically more meaningful data basis for the emission inventory. Due to the sample size, some breakdowns are only available at UK level and this study had to assume that this breakdown also holds for Scotland.

The spatial pattern in emissions has changed significantly between NAEI releases and different studies, indicative of large uncertainties in the UK emission inventory. Whilst the use of information from Energy Performance Certificates (EPCs) in this study should have helped with the allocation, these certificates only cover a subset of properties. It is theoretically possible that this subset of properties is not representative of the total housing stock. EPCs are required when properties are newly constructed, sold or rented out, and as an eligibility requirement for certain subsidies related to energy improvements.

Finally, emissions depend on fuel type, fuel quality / condition, appliances, installations (e.g. chimney draught) and burning practice. Whilst the new EFDSF study has used a common methodology as well as fuels and appliances relevant to the UK, it is necessarily based on only a few fuel samples and appliances to represent broad classes. The examples picked for the EFDSF study may not have been representative, and real-world burning practices may differ from those used during the measurements. The EFDSF study brought up some unexpected observations, including higher emissions from dry than seasoned wood.

5.1.3 Uncertainties in model estimates

Some uncertainties in the model estimates originate from generalised assumptions. For example, time variation factors are defined in the EMEP4UK model for the whole of the GNFR C ‘Other stationary combustion’ sector, so are not specific to domestic solid fuel burning. In particular, the day-of-week variation is higher for weekdays than weekends, appropriate to office or school heating patterns but likely to be a poor representation of secondary domestic heating activities in particular. The diurnal pattern of NOx emission is averaged over all fuels and therefore does not reflect the tendency of solid fuel burning to increase in the evening. By shifting emissions away from evening / night-time periods when wind speeds tend to be lower and the boundary layer shallower, the model may somewhat underestimate the contribution to population weighted mean concentrations of NO_2. The modelling carried out for this project was based on average seasonal emission profiles and did not include the impact of weather on fuel use, such as increased heating activity during short-term low temperature periods. It also does not account for spatial variations in the temporal patterns of domestic heating across Scotland, or spatial gradients in temperature.

The EMEP4UK model represents pollutant emissions and concentrations as uniform within each 1 km x 1 km horizontal grid cell, with a surface layer depth of around 50 m. This cannot capture spatial variations of emissions or concentrations on smaller spatial scales so may not be directly comparable with point measurements, especially when individual local sources may be affecting the measurement location.

The population data used to calculate population-weighted mean concentrations was originally gridded in British National Grid / OSGB coordinates and has been re-gridded to match the model projection and grid. The re-gridding process can smooth out variations and adds spatial uncertainty when matching to concentrations.

The atmospheric chemistry affecting the formation and evolution of secondary particulates is highly complex and model descriptions of these processes are still developing. This means that model predictions of secondary particulate concentrations have additional uncertainty compared to primary particulates. Linked to this, here the primary emissions from solid fuel burning are treated as non-reactive and non-evolving. It is possible that some components of the solid fuel burning particles evaporate to form volatile organic compounds. These would then be chemically processed in the atmosphere, with some re-partitioning into PM to contribute to the secondary organic aerosol component, rather than the primary component.

Ultimately, however, the uncertainty in model outputs is crucially linked to the uncertainty in model inputs and it is likely that uncertainties in emissions dominate the overall uncertainties in the model results.

5.2 Future research priorities

A version of the National Atmospheric Emissions Inventory that takes into account the fuel statistics of the CAS2022/23 and the EDFS emission factors has just been released for 2023. This follows a different methodology in spatially disaggregating emissions across the UK and Scotland. On first glance, the new release appears to have attributed more emissions back into urban centres and Smoke Control Areas, compared to the previous releases (cf Fig. 3.4 and associated text). A comparison of spatial emissions and a rerun through the EMEP4UK atmospheric chemistry and transport model would be very illuminating in highlighting current uncertainties associated with this disaggregation and interpretation / implementation of the new statistics.

An expansion of the model / measurement intercomparison to cover the full UK would provide valuable information about whether the relative attribution of the total fuel to the different parts of the UK derived from the CAS2022/23 is appropriate. For example, overall model overestimation at Scottish sites at the same time as overall underestimation in England might suggest that too much fuel has been attributed to Scotland. Measurement sites available for this exercise include Defra’s full BC network and the four sites performing continuous measurements of PM speciation by Aerosol Chemical Speciation Monitor (ACSM; similar in principle to the AMS) at London Honor Road Park, Chilbolton Atmospheric Observatory, the Manchester Air Quality Site (MAQS) and the Birmingham Air Quality Site (BAQS).

Further information on burning practices and much improved fuel use statistics for Scotland that would allow Scotland-specific attribution of the various fuels across burning areas (rural vs urban vs SCA), appliance types and ages would likely make the single largest change to the confidence in the emissions and therefore model results.

This study has provided state-of-the-art maps of the $PM_{2.5}$ contribution from domestic indoor burning across Scotland which would provide a suitable database to quantify associated health impacts and to assess where interventions might be most effective. In particular, the model results presented in this report could be used to calculate the fraction of total $PM_{2.5}$ concentration contributed by domestic solid fuel and locations where $PM_{2.5}$ is above e.g. WHO thresholds, to assess where changes to domestic solid fuel burning restrictions could alter compliance with standards. It could be used to investigate inequalities with respect to exposure of different demographics, for example given the higher $PM_{2.5}$ solid fuel concentration contributions in small towns such as Dumfries, Ayr and Fort William.

The current study does not include the impact of outdoor burning, muirburn and accidental wildfires (incl. abroad), which are the likely causes for summertime peaks seen in the long-term aethalometer measurements (Section 2.5). A recent study suggests that open fires of sizes detectable through Earth Observation may add about 1 $\mu g\ m^{-3}$ to UK $PM_{2.5}$ on average (Tan et al., 2025a, b). It would be of interest to compare the contributions from domestic indoor burning, domestic outdoor burning and wildfires in one holistic study.

Isotope measurements of the carbonaceous aerosol component would have been useful as an additional constraint of the measurement-derived $PM_{2.5}$ fraction attributed to solid fuel burning and this should be added to future studies where possible.

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