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# Comparative receptor modelling for the sources of fine particulate matter ($PM_{2.5}$) at urban sites in the UK

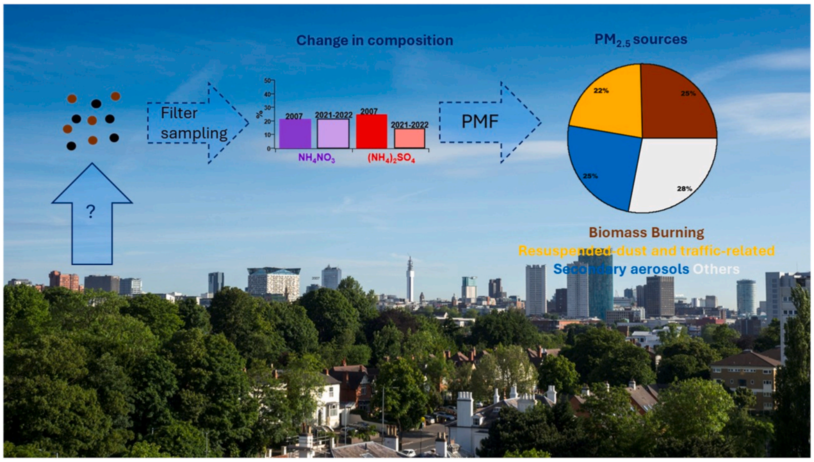
Deepchandra Srivastava [a], Supattarachai Saksakulkrai [a], W. Joe F. Acton [a], Daniel J. Rooney [a], James Hall [b], Siqi Hou [a], Mark Wolstencroft [c], Suzanne Bartington [d], Roy M. Harrison [a,1], Zongbo Shi [a,*], William J. Bloss [a]

[a] *School of Geography Earth and Environmental Science, University of Birmingham, Edgbaston, Birmingham, B15 2TT, UK*
[b] *Health Economics Unit, Institute of Applied Health Research, University of Birmingham, Edgbaston Park Road, Birmingham, B15 2TT, UK*
[c] *Environmental Health Regulation & Enforcement Division City Operations Directorate, Birmingham City Council, Birmingham, B2 2AE, UK*
[d] *Institute of Applied Health Research, University of Birmingham, Edgbaston Park Road, Birmingham, B15 2TT, UK*

## HIGHLIGHTS

- Declining $(NH_4)_2SO_4$ contribution to $PM_{2.5}$ from 25% in 2007 to 15% in 2021/2022.
- Consistent $NH_4NO_3$ contribution to $PM_{2.5}$ (20–22%) despite air quality measures.
- Key sources: Biomass burning, resuspended dust and traffic-related, secondary aerosols.
- Biomass burning $PM_{2.5}$ in 2021/2022 is 7 times higher than in 2008–2010.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The concentration of fine particulate matter ($PM_{2.5}$) in the UK has been decreasing in the last few decades but remains the leading environmental health risk. As a consequence of changing population, behaviour, activity factors, emissions and regulation, it is likely that the sources of $PM_{2.5}$ in the UK are changing but a quantitative source apportionment has not been reported for many years. This study analyses the characteristics and sources of $PM_{2.5}$ collected in 2021 and 2022 at two urban background sites: Birmingham Air Quality Supersite (BAQS) and Ladywood (LW) in Birmingham. Results indicate a notable decrease in the contribution (concentration) of

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* Corresponding author.
*E-mail address:* z.shi@bham.ac.uk (Z. Shi).

[1] Also at: Department of Environmental Sciences, Centre of Excellence in Environmental Studies, King Abdulaziz University, PO Box 80203, Jeddah, 21589, Saudi Arabia.

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$(NH_4)_2SO_4$ to $PM_{2.5}$ from 25% (2.9 μg $m^{-3}$) in 2007 to 15% (1.5 μg $m^{-3}$) in 2022. In contrast, the contribution of $NH_4NO_3$ (20–22%) to $PM_{2.5}$ remains consistent with that in 2007, despite various air quality actions implemented over the years. These shifts are attributed to changes in $SO_2$ and $NO_x$ emissions, coupled with relatively stable $NH_3$ levels—key precursors for $(NH_4)_2SO_4$ and $NH_4NO_3$, which altered the formation dynamics of these compounds, ultimately affecting their contribution to $PM_{2.5}$ concentrations. Positive matrix factorization (PMF) analysis showed that biomass burning factors (25%), resuspended dust and traffic-related emissions (22%), and secondary aerosols (25%) are the major contributors to $PM_{2.5}$ mass. Furthermore, PMF and Aethalometer-model analyses consistently showed that biomass burning aerosol concentrations are approximately seven times higher (1.5 μg $m^{-3}$) than those observed in studies from 2008 to 2010. These findings suggest that targeted measures to reduce wood burning and road traffic have the greatest potential to reduce $PM_{2.5}$-related health risks in the West Midland region.

## 1. Introduction

Atmospheric particulate matter (PM) affects air quality (and consequently human health), visibility, and ecosystems (Bellouin et al., 2020). Through the absorption and scattering of solar radiation and by affecting cloud formation and albedo, PM also has a major impact on climate (Bellouin et al., 2020). PM with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$) is given special attention due to its adverse effects on human health, as it can penetrate deep into the lungs when inhaled (Pope and Dockery, 2006). Following the UK Environment Act in 2021, an annual average $PM_{2.5}$ target level for England of 10 μg $m^{-3}$ is to be achieved by 2040, with an interim target of 12 μg $m^{-3}$ intended to be achieved by January 2028. The World Health Organisation (WHO) recently updated its global guidelines for $PM_{2.5}$ concentration (5 μg $m^{-3}$ annual mean) in 2021 reflecting estimation of the lowest level at which there is clear evidence of risk to health. These guidelines are exceeded in most urban areas in England. Current air quality action plans in the UK primarily focus on the reduction of nitrogen dioxide ($NO_2$), a major component of traffic emissions, through interventions such as clean air zones (CAZ). However, $PM_{2.5}$ is a major driver of the health impacts of air pollution, and there are many sources of $PM_{2.5}$ other than road traffic, suggesting that other targeted measures are needed. Quantitative source apportionment to determine the origins of the atmospheric $PM_{2.5}$ burden provides key information to support such efforts. The health effects of short- and long-term exposure to fine particles ($PM_{2.5}$) are well-documented and include increased respiratory and cardiovascular morbidity and mortality (Thangavel et al., 2022). Exposure to $PM_{2.5}$ is also associated with low birthweight and an increased risk of lung cancer (Balakrishnan et al., 2023; Johnson et al., 2021). There is emerging evidence of impacts on cognitive function (COMEAP, 2022; Zare Sakhvidi et al., 2022). In the UK, 28,000–36,000 premature deaths each year are attributable to exposure to air pollution (COMEAP, 2022). Consequently, air pollution is responsible for direct or indirect costs of billions of pounds every year to national health services. According to Public Health England, the total cost of $PM_{2.5}$ exposures to national health services and social care were estimated to be £1.5 billion by 2025, and £5.1 billion by 2035 (PHE, 2018).

Receptor models such as positive matrix factorization (PMF), chemical mass balance (CMB), and UNMIX are commonly used for source apportionment of $PM_{2.5}$ (Belis et al., 2013; Srivastava et al., 2021; Xu et al., 2021). Among them, PMF is a widely used multivariate method that can resolve the dominant positive factors without prior knowledge of sources (Hopke et al., 2020). Many studies identifying sources of $PM_{2.5}$ using online and offline measurements have been conducted in the UK (Charron et al., 2007, 2013; Crilley et al., 2015; Font et al., 2022; Harrison et al., 1996, 2011, 2012; Harrison and Yin, 2008; Heal et al., 2011; Yin et al., 2010). The use of organic molecular markers in PMF has enhanced our understanding as they can be source-specific and can offer a clearer link between factors and sources (Schauer et al., 2007; Shrivastava et al., 2007; Srivastava et al., 2018, 2021). However, the PMF model has certain limitations which can result in mixing of sources (Srivastava et al., 2021). In these scenarios, other source apportionment methods such as the Aethalometer model based on using absorption coefficient data have gained attention to distinguish combustion sources like fossil fuel and biomass burning (Fuller et al., 2014; Sandradewi et al., 2008). It is worth noting that sources identified using these approaches reflect what is present in the air. This includes particles influenced by both local and regional emissions, secondary formation, advection and deposition, and by chemical and physical transformations.

This study presents the results obtained from the PMF model applied to a filter-derived dataset collected at two urban background sites in the West Midlands. The West Midlands conurbation, located in central England, UK, is home to approximately 2.9 million people who are exposed to $PM_{2.5}$ concentrations in exceedance of WHO guidelines. The study provides source apportionment results from two locations, including their temporal and spatial variation. The study also presents changes in $PM_{2.5}$ chemical composition and sources over the years (2005–2020), along with a comparison with independent measurements for certain sources (biomass burning vs traffic). The study highlights the implications of these results for policy including a health impact assessment.

## 2. Methodology

### 2.1. Sampling site and sample collection

$PM_{2.5}$ sampling was conducted at two urban background sites, Birmingham Air Quality Supersite (BAQS) and Ladywood (LW) in the West Midlands, UK. Monitoring was carried out from January 4, 2021 to February 27, 2022 and 4th January to December 15, 2021 at BAQs and Ladywood, respectively (Fig. S1). Both sites are in Birmingham, the largest administrative component of the West Midlands conurbation. BAQS (52.45° N, 1.93° W) is described in Yin et al. (2010), and is a NERC funded supersite, located on the University of Birmingham's Edgbaston campus. BAQS is situated approximately 3.5 km southwest of Birmingham city centre and is away from any major roads. The nearest anthropogenic sources include a nearby railway track, a moderately trafficked road (120 m away), and activities arising from local residents and the university campus. The instruments at BAQS measure regulated air pollutants ($NO_x$, $O_3$, $SO_2$, $PM_{2.5}$ mass concentration etc), online PM composition, $PM_{2.5}$ samples collected for offline chemical characterisation, and meteorological parameters. Instruments are audited via quality checks with the National Physical Laboratory every six months for NO, $NO_x$, $O_3$, $SO_2$, and $CO_2$. The Ladywood site is operated as part of the UK government DEFRA Automatic Urban and Rural Network (AURN) (https://uk-air.defra.gov.uk/networks/). Unlike BAQS, most AURN sites only provide measurements for regulated air pollutants without detailed chemical characterisation. Ladywood is located on a green space next to a primary school, adjacent to a residential (low traffic) Summer Hill Road. The nearby surroundings include open green space and residential buildings. Ladywood is situated within Birmingham city centre (~1.4 km to the north of BAQS), and at about 600 m to the inner ring road.

24-h $PM_{2.5}$ samples (total filter samples = 120 (BAQS = 69 and Ladywood = 51)) were collected on pre-baked quartz filters (Pallflex, 8 × 10 inch) at BAQS (Tisch, USA, flow rate of 1.1 $m^3$ $min^{-1}$) and

Ladywood (Tisch, USA, flow rate of 1.1 $m^3\ min^{-1}$) using high-volume air samplers. Field blanks were also collected during the sampling campaign at both sites. Sampled filters were prepared and conserved following the same protocols reported elsewhere (Srivastava et al., 2023). Online measurements of $PM_{2.5}$ mass and Black Carbon (BC) concentrations were conducted using TEOM-FDMS and Aethalometer (Model AE33) at both sites (BAQS and Ladywood). High-time resolution measurements of organics, sulphate, nitrate, ammonium and chloride and metals were also available at BAQS using the Aerosol Chemical Speciation Monitor (ACSM) and Xact 625i Ambient Metal Monitor. Meteorological data: relative humidity (RH), temperature (T), horizontal wind speed (ws), wind direction (wd), and cloud layer height were recorded at BAQS.

### 2.2. *Analytical procedure*

The quantified chemical species in each $PM_{2.5}$ sample from BAQS and Ladywood included elemental carbon (EC)/organic carbon (OC), 36 organic tracers, 7 major inorganic ions ($Na^+$, $K^+$, $Ca^{2+}$, $NH_4^+$, $Cl^-$, $NO_3^-$ and $SO_4^{2-}$) and 17 metallic elements (V, Cr, Co, Mn, Ni, Cu, Zn, As, Br, Sr, Ag, Cd, Sn, Sb, Ba, Hg and Pb). The analysis of all $PM_{2.5}$ chemical species followed the procedure described previously (Srivastava et al., 2023; Xu et al., 2021), and therefore, only essential information is presented in this section.

Briefly, EC/OC measurements (0.5 $cm^2$ quartz filter punch) were performed using a DRI multi-wavelength thermal-optical carbon (model 2015, EUSAAR2 transmittance protocol) analyser. Major inorganic ions and metallic elements were analysed using an ion chromatograph (IC, Dionex, Sunnyvale, CA, USA) and inductively coupled plasma-mass spectrometer (ICP-MS). For inorganic ions analysis, a 2 $cm^2$ quartz filter punch was extracted with 10 ml of deionised water using a sonication technique (1 h @ 27 °C), followed by filtration through a 0.45 μm syringe filter to remove any traces of filter particles. The final solution was then stored at 4 °C before analysis. For metallic elements analysis, a 6 $cm^2$ quartz filter punch was extracted with 10 ml of a 2% $HNO_3$ solution and subjected to microwave digestion (CEM, MARS 6 iWave) at 200 °C, followed by serial dilutions to prepare the final volume before analysis. Additionally, organic tracers were analysed using gas chromatography-mass spectrometry. Details on the organic species analysis are provided in the Supplementary information (SI, Section S1).

### 2.3. *Positive matrix factorization*

Positive matrix factorization (PMF) is an extensively used receptor modelling approach and applies the concept of a weighted least squares fit (Paatero and Tapper, 1994). The weightings used in the PMF are derived from the analytical uncertainty of the measurements. We determined the analytical uncertainties for the filter-based measurements using the procedure explained previously (Polissar et al., 1998). The U.S. Environmental Protection Agency (US-EPA) PMF 5.0 software was used in this work to perform the source apportionment of airborne particles.

The selection of input species for the PMF analysis was based on the same set of criteria used in our previous work (Srivastava et al., 2021). A total of 24 species were used in the model ($PM_{2.5}$, OC, EC, $Ca^{2+}$, $NH_4^+$, $NO_3^-$, $SO_4^{2-}$, $Cl^-$, Ti, V, Mn, Ni, Zn, Pb, Cu, Fe, Ba, Sn, levoglucosan (LG), galactosan (GL), mannosan (MN), succinic acid (SA), dehydroabietic acid (SA) and methylerythritol (MET)) with 120 data points (combined dataset from BAQS and Ladywood). The concentration of $PM_{2.5}$ was included as a total variable in the model (with large uncertainty, to minimise impact on the fit) to directly determine the source contributions to the daily mass concentrations. The selection of the optimum PMF solution included a detailed investigation of factor chemical profiles, temporal trends, correlations with external tracers, bootstrap runs for solution stability and modelled vs. measured comparison.

## 3. Results

### 3.1. *$PM_{2.5}$ chemical composition*

$PM_{2.5}$ mass was reconstructed using a pragmatic mass closure analysis following the same procedure explained previously (Harrison et al., 2003). The comparison of the reconstructed $PM_{2.5}$ mass at BAQS and Ladywood, and the $PM_{2.5}$ mass measured by online instruments (TEOM-FDMS), showed good agreement ($r^2 = 0.7–0.8$, slope = 1.2–1.4, $p < 0.05$, ODR – orthogonal distance regression) (Fig. S2). On an annual basis, the results suggest that the highest contribution to $PM_{2.5}$ mass comes from organic matter (36–38%), followed by secondary inorganics (35–36%, the sum of ammonium sulphate and ammonium nitrate), soil dust (10–11%), and EC (7–10%) (Fig. 1). Individual $PM_{2.5}$ components, including OM, EC, ammonium nitrate, ammonium sulphate, sea salt, dust and calcium salt, were compared between BAQS and Ladywood sites (see Fig. S2). Correlations coefficient for these components ranged from 0.6 to 0.9 between the two sites, except for calcium salts, confirming the consistency of sources between the sites. Overall, results showed comparable contributions of different $PM_{2.5}$ components at both sites, suggesting the influence of similar sources or activities. Note, this represents sources of $PM_{2.5}$ present in the air, including local and regional activities, as well as chemical reactions that can take place during the transport of air masses (i.e., secondary PM formation, processing). This is not equivalent to the PM directly emitted, referred to as primary emissions.

### 3.2. *$PM_{2.5}$ source apportionment in the West Midlands*

PMF analyses were performed on the combined chemical composition datasets from BAQS and Ladywood. Based on the factor chemical and temporal profiles, seven sources were identified, which included two biomass burning factors, resuspended dust and traffic-related emissions, fuel oil combustion, sea salt, secondary aerosols, and biogenic secondary organic aerosols (SOA). Here, 'fuel oil combustion' serves as an operational term to aid in understanding this source more broadly. For more detailed insights, please see the text below. For the selection of the final solution, we also investigated solutions with a different number of factors. All solutions, other than the seven-factor solution, were less explanatory, often mixed (by decreasing the number of factors) or split (by increasing the number of factors) into non-meaningful factor profiles. The comparison between the modelled $PM_{2.5}$ contributions from all sources and the measured $PM_{2.5}$ concentrations showed good agreement ($r^2 = 0.8$, slope = 1.2, Fig. S3). In addition, constraints were applied on the base run to produce a clean profile and no significant difference ($p > 0.05$) was observed between the base and constrained runs. To assess the stability of the final solution, bootstrapping was performed, showing stable results with bootstrap-mapped factors greater than 85 (Table S1).

The two biomass burning factors, resuspended dust and traffic-related emissions, and secondary aerosols accounted for approximately 72% of $PM_{2.5}$ mass, followed by fuel oil combustion (9%), biogenic SOA (10%), and sea salt (9%) (Fig. 1). Please note that the comparison for the source contributions was conducted using only data collected during overlapping sampling times at both sites.

***Biomass Burning1:*** A high contribution to the total concentrations of anhydrous sugars, such as mannosan (63%) and galactosan (32%), was observed in the chemical profile, together with a significant contribution to OC (18%) and EC (35%) (see Fig. 2). These sugars are formed via the pyrolysis of cellulose, an element present in biomass, and are therefore emitted in large quantities during biomass burning events. Surprisingly, the contribution to levoglucosan from this factor was found to be low (7%). Levoglucosan is a major pyrolysis product of cellulose and has been proposed as a molecular marker of biomass burning aerosols (Simoneit, 1999). The observed low ratio (<1.5) of levoglucosan to mannosan suggests the dominance of softwood burning. This was

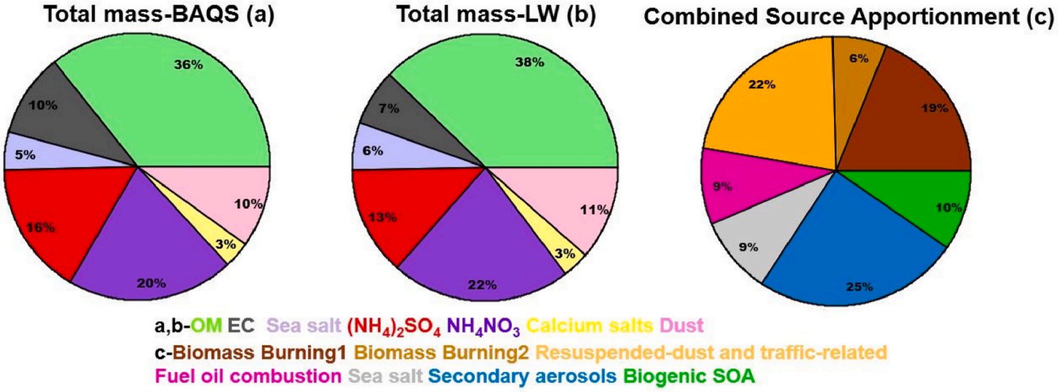

**Fig. 1.** The contribution of different $PM_{2.5}$ components to total mass from the mass closure model (a, b); The average percentage contribution of sources to the total $PM_{2.5}$ mass at both sites from PMF (c).

confirmed, as this factor also showed a substantial contribution to dehydroabietic acid (resin acid, 70%), a specific marker of softwood burning (Simoneit and Elias, 2001).

In addition, $NO_3^-$ (27%) and $NH_4^+$ (24%) contributed significantly to this factor, and their presence in biomass burning plumes has been reported previously (Paulot et al., 2017; Srivastava et al., 2021; Zhao et al., 2020). The factor followed typical seasonal variations with a higher contribution during the cold period, suggesting it could be linked to heating (residential) activities in winter (Fig. 3 and S4). This factor accounted for 19% (annual average) of the $PM_{2.5}$ mass (1.5 μg $m^{-3}$) (Fig. 1).

***Biomass Burning2:*** The second biomass burning factor was characterized by high contributions to levoglucosan (76%) and galactosan (42%) (see Fig. 2). As discussed above, these are cellulose pyrolysis products found abundantly in biomass burning smoke. The source did not show clear seasonal pattern (Fig. 3 and S4) and accounted for 6% (0.5 μg $m^{-3}$) of the $PM_{2.5}$ mass annually (Fig. 1). Additionally, the contribution to $PM_{2.5}$ mass at both sites ranged between 8 and 10% in winter, 4% in spring, 4–7% in summer, and 5–7% in autumn. This suggests that this source could be linked to other activities such as garden waste burning, biomass boilers or barbecues.

***Resuspended dust and traffic-related emissions:*** This factor showed relatively high contributions to the water-soluble ion $Ca^{2+}$ (39%) and metallic elements such as Ti (67%), Fe (73%), Cu (71%), Sn (48%), Ni (44%), and Ba (66%) (Fig. 2). Cu and Ba can originate from brake wear (especially), lubricants, and corrosion of vehicular parts, as well as tailpipe emissions (Goddard et al., 2019; Grigoratos and Martini, 2015; Pant and Harrison, 2012, 2013). Diurnals of Cu and Ba, investigated using the high-resolution data obtained from an online instrument (Xact) at BAQS, showed a similar behaviour to traffic, with morning and evening peaks as expected (Fig. S5). This was further confirmed by the similar diurnal pattern exhibited by $NO_x$, a gaseous pollutant often regarded as a tracer for traffic pollution in urban/suburban environments such as these. Additionally, the seasonal temporal profile of the retrieved factor showed good agreement with $NO_x$, indicating its association with traffic emissions (Fig. S6). Elements such as Ca, Ti, and Fe have been found in abundance in resuspended dust (Pant et al., 2015; Pant and Harrison, 2012; Viana et al., 2008). These elements have been found to be related to tyre and road surface wear particles (TRWP), generated by the shear force between the tyre tread and the road surface (Pant and Harrison, 2013). Laboratory studies have suggested that Cu, Fe, Ti, Ba, Sn and Ni are also linked to brake pads (Fussell et al., 2022). This suggests that the factor presents the characteristics of both traffic-related emissions and resuspended dust.

The contribution of this source to $PM_{2.5}$ mass the two sites (Ladywood and BAQS) ranged between 9-13% in winter, 25–33% in spring, 33–42% in summer, and 19–24% in autumn and accounted for 22% (1.7 μg $m^{-3}$) of the $PM_{2.5}$ mass annually (Fig. 1). The source did show a pattern with higher concentrations observed in spring and summer, which may be reflective of dust resuspension. (Fig. 3).

**Sea salt:** This factor was characterized by high contributions to $Cl^-$ (81%) and $Mg^{2+}$ (43%) as shown in Fig. 2. $Na^+$ is another abundant ion observed in the sea salt chemical profile (Seinfeld and Pandis, 2016). However, we excluded $Na^+$ concentrations from the PMF analysis due to contamination in measurements at Ladywood. Furthermore, $Na^+$ and $Cl^-$ follow a similar trend at BAQS, suggesting that these ions are related to sea salt (Fig. S7). Sea salt was also estimated based on the concentrations of $Na^+$ and $Cl^-$ at BAQS, using a similar approach as explained by Quinn et al. (2007). A good correlation was observed between the estimated sea salt and this factor at BAQS ($r^2 = 0.8$, slope $= 1.2$), further indicating that this factor can be attributed to sea salt particles (Fig. S8). This source accounted for 9 % of the total $PM_{2.5}$ mass annually, equivalent to 0.7 μg $m^{-3}$ (Fig. 1).

***Fuel oil combustion:*** This factor makes a significant contribution to elements, such as V (71%), Ni (30%), and Sb (36%) (see Fig. 2), with a small contribution to $SO_4^{2-}$ (10%). Previous studies suggested that Ni/V are commonly associated with the combustion of fuel oil from industrial or shipping activities including some influence from crustal dust (Jang et al., 2023; Viana et al., 2008; Xiao et al., 2018). NWR (non-parametric wind regression) plots, derived using high-resolution data of V and Ni at BAQS (obtained from Xact measurements), suggest dominance from north-westerly (Ni) and north-easterly (V) air masses with minimal influence from maritime air masses (Fig. S9). Additionally, V and Ni do not appear to be correlated at either site, indicating the likely influence of different emission sources (Fig. S10). The presence of Sb in this factor can further confirm the association with anthropogenic activities (Pač et al., 2023). Therefore, the identified source has been labelled as “fuel oil combustion”, but this source may also include industrial processes. This source accounted for 9 % of the total $PM_{2.5}$ mass (0.7 μg $m^{-3}$) annually, with no clear seasonal variation observed at both sites (Figs. 1 and 3 and S4).

***Secondary aerosols*:** This source was characterised by high contributions to $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ (65%, 42% and 70% of the total species mass, respectively) (Fig. 2). This factor also contributed a significant amount to OC (38%) and EC (31%). The factor accounted for 25% (1.9 μg $m^{-3}$) of the annual $PM_{2.5}$ mass, with the highest contribution reported during spring (Ladywood: 35%, BAQS: 31%). The key precursor species for $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ are $NO_x$, $SO_2$ and $NH_3$, respectively. Emissions of $SO_2$ and $NO_x$ nationally derive from a variety of combustion sources such as power generation, industrial combustion, and traffic emissions. The industrial contribution to $PM_{2.5}$ mass is linked to both fuel oil combustion (discussed earlier) and secondary aerosol sources. Therefore, these findings suggest that the overall contribution of industrial activities to PM mass may be higher than presented here by the

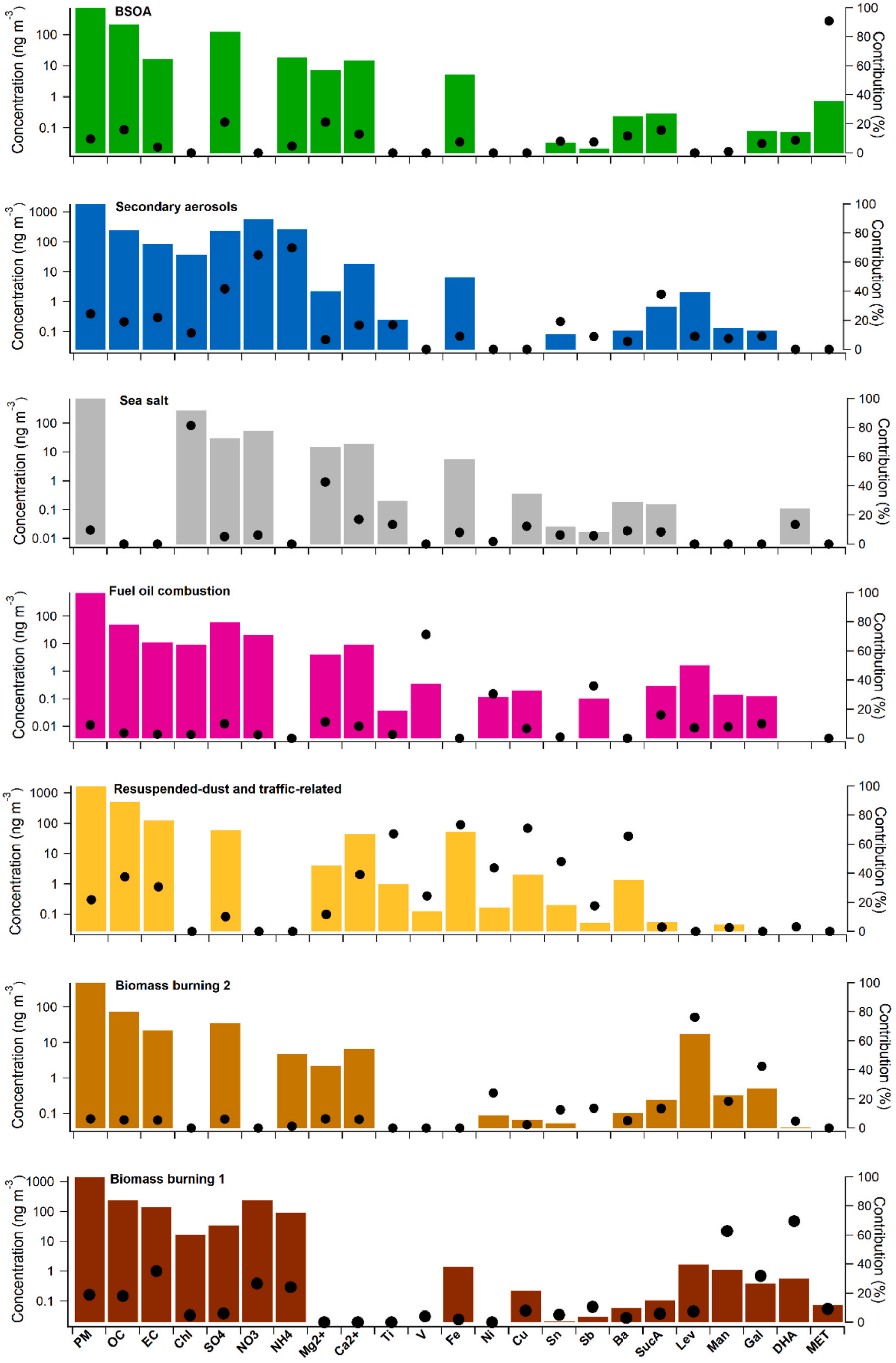

**Fig. 2.** Chemical profiles for the identified factors at BAQS and Ladywood. The bars show the composition profile (left axis) and the dots present the Explained Variation (right axis).

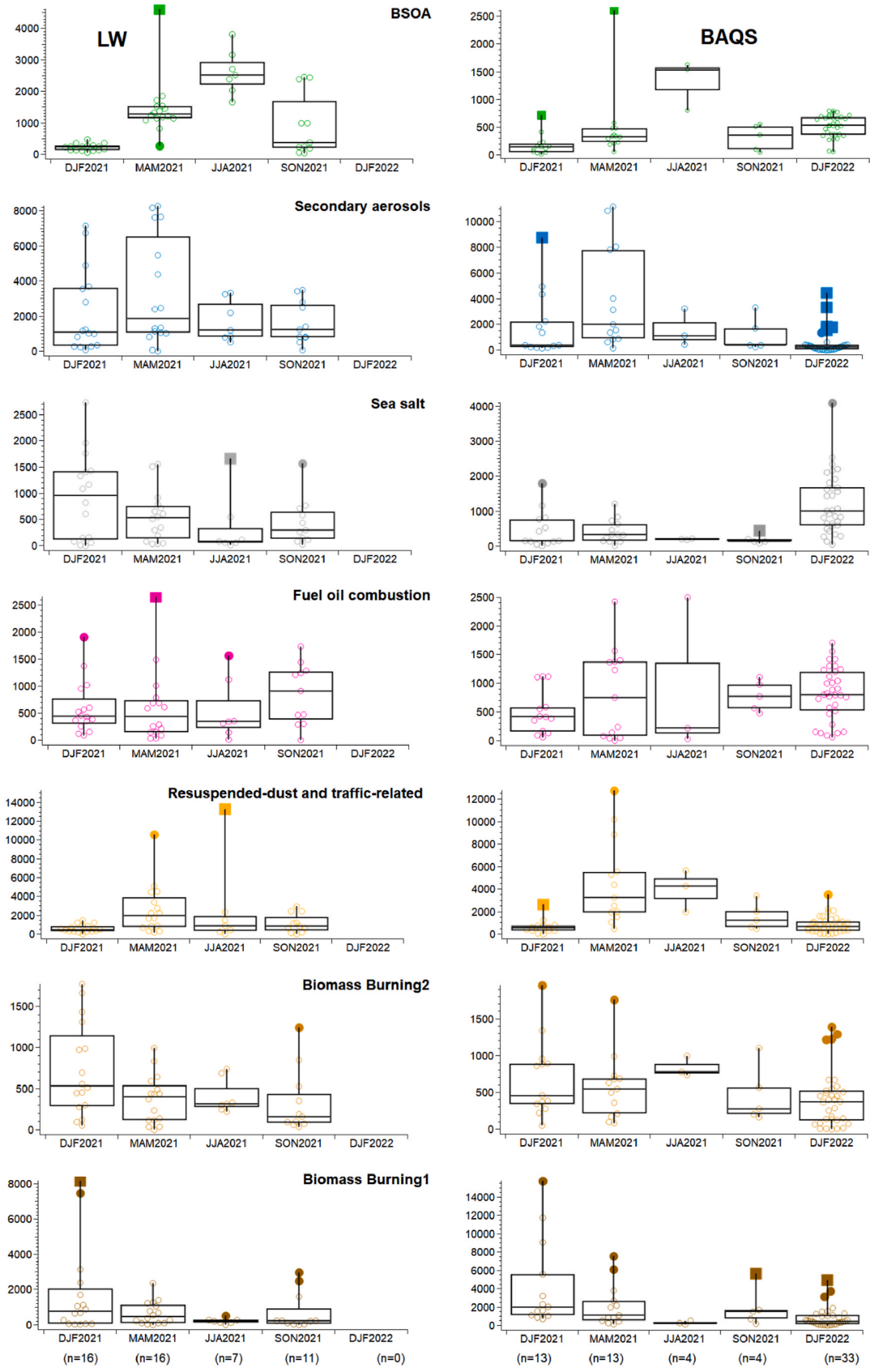

**Fig. 3.** Box plots present the seasonal concentrations of identified factors at BAQS and Ladywood. The number "n" represents the data points taken to draw the box plots for each season. Concentrations are given in ng m$^{-3}$.

fuel oil combustion source alone.

$NH_3$ emissions are mostly from agriculture, with minor contributions from industry and traffic (Jonson et al., 2022). The oxidation of $NO_x$ and $SO_2$ to secondary particles occurs on the timescale of hours to days. As a result, the measured components reflect contributions from both distant and local sources. Traffic can also contribute to $PM_{2.5}$ mass through secondary particle formation (in the form of $NO_3^-$). Therefore, the impact of traffic emissions on $PM_{2.5}$ mass is likely to be higher than that solely indicated by the traffic factor discussed earlier. We have also calculated the degree of neutralisation based on the molar ratio of $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ (Adams et al., 1999). A value of 1.02 was observed, suggesting that $NO_3^-$ and $SO_4^{2-}$ exist as $NH_4NO_3$, and $NH_4(SO_4)_2$. In addition, this factor showed a significant contribution to succinic acid (38%) of the species found within this factor, recognised as a typical secondary organic species formed from the oxidation of anthropogenic precursors (Kawamura and Bikkina, 2016; Kawamura and Ikushima, 1993), indicating the presence of anthropogenic secondary organic matter in this factor.

Polar plots made using high-resolution data obtained for $NO_3^-$, $SO_4^{2-}$ and $NH_4^+$ from the aerosol chemical mass spectrometry at BAQS, suggested the influence of regional transport, highlighting the role of processed aerosols (Fig. S11). The value of online data to better understand the origin of these species is reflected in the strong agreement observed between offline and online measurements at BAQS (Fig. S12).

***Biogenic SOA:*** This factor was characterised by significant contributions to the oxidation product of isoprene (Carlton et al., 2009) (methylerythritol, 91%) (see MET in Fig. 2). This factor accounted for 10% of $PM_{2.5}$ mass, corresponding to a concentration of 0.7 μg $m^{-3}$ at both sites (as an annual mean). As expected, this factor showed high concentrations from early spring to autumn due to an increase in biogenic activity during the warmer period (Fig. 3 and S4). $SO_4^{2-}$ (21%) also contributed to this factor. The presence of $SO_4^{2-}$ has already been shown to have a positive effect on isoprene SOA formation, facilitating the conditions for acid-catalysed reactive uptake and ring-opening reactions of key intermediates in the gas phase or increasing the solubility of polar organic compounds (Li et al., 2018). In addition, high concentrations/contributions were observed at Ladywood compared to BAQS.

The contribution reported in the current study is lower than found previously for the same location in Birmingham (16%) (Heal et al., 2011), which utilized radiocarbon measurements to apportion biogenic SOA in the summer of 2007 and 2008. Radiocarbon measurement distinguishes between fossil and non-fossil carbon sources (Szidat et al., 2004). Non-fossil sources include primary and secondary biogenic OA, vegetative detritus, and cooking. Heal et al. (2011) attributed all non-fossil contributions to secondary biogenic SOA, assuming negligible contributions from other sources. This assumption could lead to the overestimation of identified sources, as has been reported in many other studies. It is also worth noting that the current biogenic factor only represents biogenic SOA formed from isoprene and those other components which correlate with it. The contribution of other biogenic precursors to this factor may not be fully accounted for due to a lack of other marker species or, in cases where they had low concentrations, they were not included in the PMF analysis. Therefore, the identified biogenic SOA only represents a fraction of the total biogenic SOA, explaining the lower contribution compared to Heal et al. (2011)

## 4. Discussion

### 4.1. Change in $PM_{2.5}$ chemical composition over time

$PM_{2.5}$ mass was dominated by the organic fraction (36–38%, 3.3 μg $m^{-3}$), followed by ammonium nitrate (20–22%, 1.9 μg $m^{-3}$) and ammonium sulphate (13–15%, 1.3 μg $m^{-3}$), EC (7–10%, 0.8 μg $m^{-3}$) (Fig. 1). Similar relative contributions were observed in previous studies conducted in Birmingham for carbonaceous material and sea salt using a similar approach (Harrison et al., 2004; Harrison and Yin, 2008; Yin et al., 2010). Sea salt annual concentration/contributions at BAQS (5%, 0.4 μg $m^{-3}$) and Ladywood (6%, 0.5 μg $m^{-3}$) were found to be comparable to previous observations (2007–2008, BAQS, 7%, 0.8 μg $m^{-3}$) reported by Yin et al. (2010) (Table S3). This is consistent with other studies in the UK, such as London (during the summer and winter of 2012) (Crilley et al., 2017; Yin et al., 2015). Additionally, sea salt concentrations (1.1 μg $m^{-3}$) reported using chemical mass balance approach in London (2012) were also similar to the concentrations observed in the current study. These results are also consistent with a recent study conducted by Bousiotis et al. (2023) which reported a marine contribution to $PM_{2.5}$ ranging from 8 to 13% in the central UK for observations recorded between September and October 2020.

The contribution and concentration of $(NH_4)_2SO_4$ to $PM_{2.5}$ has significantly decreased at BAQS from 25% (2.9 μg $m^{-3}$) in 2007–2008 (Yin et al., 2010) to 15% (1.5 μg $m^{-3}$) in this study. In contrast, the $NH_4NO_3$ contribution has remained stable: 20% (1.9 μg $m^{-3}$) at BAQS and 22% (1.9 μg $m^{-3}$) at Ladywood in this study, compared to 15% (2.3 μg $m^{-3}$) in 2000–2002 (Harrison et al., 2012) and (17%, 2.1 μg $m^{-3}$) in 2007–2008 at BAQS (Yin et al., 2010). This indicates no major changes in $NH_4NO_3$ levels despite implementation of various air quality regulations over the past several years. Since 2000, anthropogenic emissions of $SO_2$, $NO_x$, and $NH_3$ (precursor gases for $(NH_4)_2SO_4$ and $NH_4NO_3$) for the whole of the UK have declined by 81 %, 51 %, and 13 %, respectively (NAEI, 2018). The substantial decrease in (national) $SO_2$ and $NO_x$ emissions, and a much smaller decrease in $NH_3$ emissions has also led changes in the formation of $(NH_4)_2SO_4$ and $NH_4NO_3$, subsequently changing the contribution of these species to PM mass and concentrations in the air. The affinity of $H_2SO_4$ (oxidation product of $SO_2$) for $NH_3$ is higher than that of $HNO_3$, which means available $NH_3$ will first react with $H_2SO_4$ to form $(NH_4)_2SO_4$, and the formation of $NH_4NO_3$ will depend on the available excess $NH_3$. This suggests that the large reduction in $SO_2$ may lead to increased available $NH_3$ and hence to more $NH_4NO_3$ formation. This can explain the observed pattern for $NH_4NO_3$ contribution and concentrations in the current study and is consistent with findings of a non-linearity in the $NO_x$-nitrate relationship in two earlier studies (Harrison et al., 2022; Tang et al., 2018) who reported a similar observation based on data collected by UK national networks.

The organic mass fraction was also comparable to data reported at an urban background site in London during the winter period in both studies (33–35%, this study vs 31% London (2012)) (Yin et al., 2015). However, the OM concentration was much higher at a background site in London than compared to this study (4.1 μg $m^{-3}$ > 2.1 μg $m^{-3}$). Calcium salts (3%, 0.3 μg $m^{-3}$) were also found to be consistent with previous studies conducted in Birmingham at BAQS (3%, 0.4 μg $m^{-3}$) (Harrison et al., 2004; Harrison and Yin, 2008; Yin et al., 2010). Additionally, $PM_{2.5}$ concentrations at BAQS (6.8 μg $m^{-3}$) were approximately 60% lower than previous measurements from 2007 to 2008 (BAQS, 11.6 μg $m^{-3}$) reported by Yin et al. (2010). This decline aligns with broader $PM_{2.5}$ trends observed across the UK, suggesting a similar reduction in concentrations (DEFRA, 2024).

### 4.2. Change in concentrations of biomass burning and traffic-related sources

As shown earlier, two biomass burning factors were identified, with biomass burning1 making a high contribution of 19% compared to 6% from biomass burning 2, to $PM_{2.5}$. Biomass Burning1 showed specific chemical characteristics, including a high contribution from dehydroabietic acid and a low ratio (<1.5) of levoglucosan to mannosan. These characteristics, along with the expected higher winter concentrations, suggest its association with local heating-related activities. The contributions of this source to $PM_{2.5}$ mass were higher at BAQS (49%, 19%, 30%) than at Ladywood (27%, 9%, 13%) in winter, spring, and autumn, respectively. The comparison highlights that elevated biomass burning concentrations at BAQS may be significantly influenced by seasonal emissions, potentially from domestic (household) sources.

Higher concentrations are observed at BAQS vs Ladywood, and we note that the former is situated in an area of relatively higher affluence and typically larger, older housing stock, which may be related to a greater preponderance for domestic e.g. wood burning for heating and/or aesthetic purposes. We found that the average concentrations of wood smoke reported by Harrison et al. (2012) were 0.23 μg $m^{-3}$ in Birmingham (2008–2010) (Table 1) and 0.30 μg $m^{-3}$ in London (2010–2011), much lower (7 times) than the concentrations (1.5 μg $m^{-3}$) observed in the current study. Another study conducted by Yin et al. (2015) also observed a low contribution of wood smoke (4%, 0.14–0.16 μg $m^{-3}$) in London in the winter of 2012. Additionally, a recent study analysing data from London North Kensington and Glasgow reported a relatively low contribution of wood burning to $PM_{2.5}$, ranging from 6 to 10% at both sites (Font et al., 2022). This contrasts with the findings of the current work. The observed differences in concentrations or contributions could be attributed to variations in emission patterns over time, differences in wood burning activities, and/or the methodologies employed. Note that earlier studies estimated wood smoke concentrations or black carbon levels using Aethalometer model (Fuller et al., 2014; Sandradewi et al., 2008) or levoglucosan concentrations and a conversion factor (Harrison et al., 2012), whereas the current study employs a more comprehensive and robust approach.

The other biomass burning factor (2) also contained signature species, including a high amount of cellulose pyrolysis products. However, this source showed a similar contribution to $PM_{2.5}$ across different seasons, suggesting it could be associated with burning activities other than heating (Fig. 3). Additionally, the average contribution to $PM_{2.5}$ mass at both sites were 9% in winter, 4% in spring, 6% in summer, and 8% in autumn. Overall, the total biomass burning, including both factors, accounted for 25 % of the total $PM_{2.5}$ mass, indicating significant importance of wood burning in this urban UK location. This was further supported by another recent PMF study conducted at the same site using high resolution data, suggesting the influence of two different types of wood combustion activities (Kamara, 2024).

The identified traffic-related source showed a higher relative contribution (23%) than results reported previously (13% in 2007–2008) by Yin et al. (2010) for the same site (BAQS). As discussed above, the retrieved source in our study is not properly resolved and includes contribution from both traffic, mostly from non-exhaust emissions, and from resuspended dust. Therefore, adding up the absolute contribution from traffic and the dust factor reported by Yin et al. (2010) showed good agreement (20%, 2.09 μg $m^{-3}$ vs 23%, 1.8 μg $m^{-3}$). This further confirms that the identified source represents a mixture characteristic of dust and traffic-related particles.

The use of three-way catalytic converters on gasoline vehicles, and particle filters for both diesel and gasoline (GDI) vehicles, has led to a significant reduction in exhaust emissions of carbon monoxide, hydrocarbons, $NO_x$ and particulate matter (Harrison et al., 2021a). As a result, non-exhaust emissions exceed exhaust emissions (AQEG, 2019). It is important to note that non-exhaust emissions include brake wear, tyre wear and road surface wear particles, including resuspended particles from road dust. This has been confirmed due to the presence of crustal elements in the chemical profile of this source.

### 4.3. Comparison of biomass burning and traffic-related sources with other source apportionment model

The Aethalometer model has been employed to trace the contributions of road traffic and wood burning emissions to equivalent black carbon (BC) (Fuller et al., 2014; Sandradewi et al., 2008). This estimation relies on measured absorption Ångström exponents (αabs) of BC from various sources. In our study, biomass (wood) burning contribution to $PM_{2.5}$ mass (BB-Aethalometer) was determined using the procedure outlined previously (Fuller et al., 2014). We used Angstrom exponent values of 2 for estimating biomass burning contribution. For comparison, the biomass burning sources from PMF analyses (BB-PMF) were summed to account for the total biomass burning contribution. We found a good agreement between the two methods in terms of their temporal correlation and the magnitude of the estimated $PM_{2.5}$ from wood burning (see Fig. 4 and S13, $r^2 = 0.7$–$0.8$, slope = 1.1–1.8), with a slightly higher contribution measured by the Aethalometer model.

The aethalometer model was also used to estimate traffic concentrations using an Angstrom exponent value of 1.07 (Fuller et al., 2014; Sandradewi et al., 2008). We found a good agreement for traffic concentrations ($C_{traffic}$) derived from Aethalometer measurements from the two sites (see Fig. S14, $r^2 = 0.9$, slope = 1.16). Results also suggest a good agreement among $NO_x$ (traffic marker), $C_{traffic}$ and the resuspended dust and traffic-related component from PMF analysis at BAQS (Fig. 5). However, $C_{traffic}$ (1.0 μg $m^{-3}$) apportioned using Aethalometer data showed approximately three-fold lower concentrations than the current source concentrations. The lower value of $C_{traffic}$ is likely because $C_{traffic}$ only represents exhaust emissions. The UK national emission inventory suggests that 67% of all road vehicle PM emissions are defined as emissions from tyres, brakes, and road abrasion (Harrison et al., 2021a) which further confirms the reported lower concentrations.

### 4.4. Secondary OC contribution estimation using the EC tracer method

The EC tracer method has been widely used to estimate the secondary OC (SOC) contribution. SOC can be estimated using the following equations if OC and EC concentrations are available (Turpin and Huntzicker, 1995). We have assumed primary OC from non-combustion sources is negligible.

$$POC = \left(\frac{OC}{EC}\right)_{Pri} {*}EC$$

$$SOC = OC - POC$$

The $(OC/EC)_{pri}$ is the OC/EC ratio in freshly emitted combustion aerosols. To estimate the $(OC/EC)_{min}$ ratio, the scatterplot function was employed (OC vs EC). The lowest point on the scatter plot was identified, with the slope representing the $(OC/EC)_{min}$, while any point above indicated the formation of secondary OC.

The minimum $(OC/EC)_{pri}$ ratios for BAQS (Jan 2021–Dec 2021) and Ladywood (Jan 2021–Dec 2021) are 1.34 and 1.78, respectively (Figure S15). The POC and SOC values, calculated using the $(OC/EC)_{pri}$, were 1.3 μg $m^{-3}$ and 1.1 μg $m^{-3}$ at BAQS and 1.0 μg $m^{-3}$ and 0.7 μg $m^{-3}$ at Ladywood, respectively. POC accounted for 54% and 60% of the total OC at BAQS and Ladywood, respectively. SOC accounted for 46% and

**Table 1**
Change in Biomass Burning contributions/concentrations over time.

| Study year and location | 2008–2010, Birmingham | 2010–2011, London | 2012, winter, London | 2021, Birmingham (BAQS/ Ladywood) |
|---|---|---|---|---|
| Reference work | Harrison et al. (Harrison et al., 2012) | Harrison et al. (Harrison et al., 2012) | Yin et al. (Yin et al., 2015) | This study |
| Biomass Burning (concentrations in μg $m^{-3}$) | *0.23* | *0.3* | *0.14–0.16* | *1.5 (2.4*[a]*)* |
| Biomass Burning (% contribution) | *NA* | *NA* | 4 | 19 |

[a] Winter BAQS; NA: Not available.

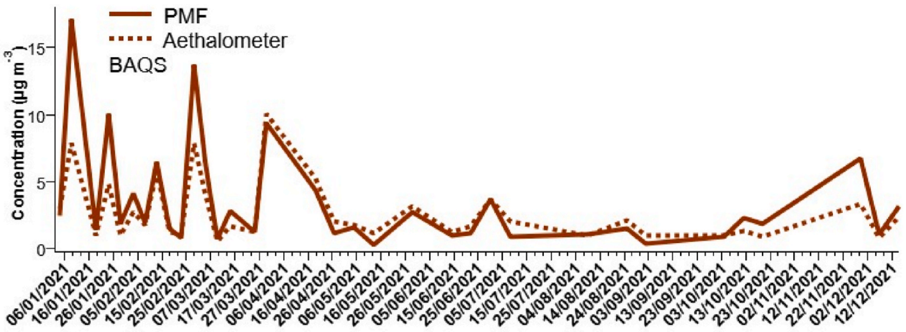

**Fig. 4.** Temporal variation of biomass burning contribution estimated using the Aethalometer approach and PMF analysis. Biomass burning from PMF analysis is the sum of Biomass Burning-1 and Biomass Burning-2.

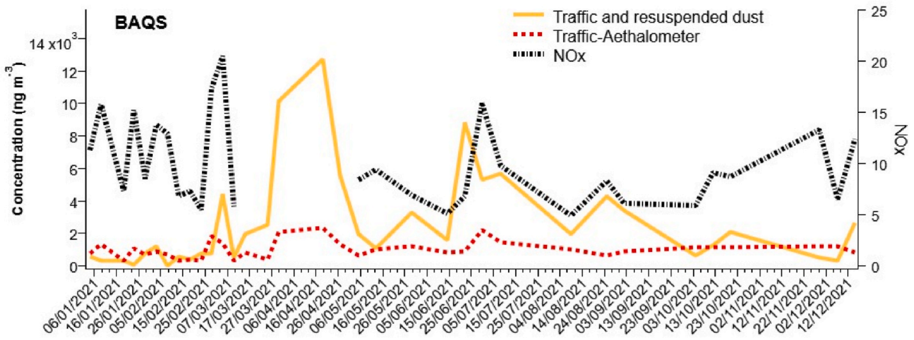

**Fig. 5.** Concentrations of $C_{traffic}$ (Aethalometer), resuspended dust-and traffic-related and $NO_x$ at BAQS.

40% of the total OC at BAQS and Ladywood, respectively. Results suggest that more than 50% of OC is from primary sources in Birmingham.

Additionally, we also computed the secondary contribution based on the PMF analysis. Among the identified seven sources, we assumed that only biogenic SOA and secondary aerosols (only OC has been considered here) represent secondary signatures. Therefore, the contribution of SOC from the PMF analysis (sum of biogenic SOA and secondary aerosols) to the total OC was 35% (0.5 µg $m^{-3}$). This suggests that the PMF analysis underestimated the contribution of SOC. Furthermore, the concentrations of secondary OC reported previously by Yin et al. (2010) showed similar concentrations (0.9 µg $m^{-3}$) at BAQS. However, the relative contribution to total OC was lower (33%).

### *4.5. Implications for policy*

The results highlight that immediate attention should be given to controlling wood burning emissions in urban areas in the West Midlands and beyond. Policies to address wood-burning emissions include strengthening local regulations, such as enhancing and enforcing smoke control areas, and measures to replace wood-burning stoves with cleaner alternatives, such as electric heating or low-emission stoves. Public awareness campaigns could highlight the health impacts of wood burning and promote behaviour changes and adopt best practice, such as burning only dry, seasoned wood and maintaining stoves to reduce pollution (DEFRA, 2023). It should also be noted that not all biomass burning aerosol is from local emissions; while local and regional actions can be effective, wider national policy interventions are also important to reduce biomass burning related $PM_{2.5}$ exposure.

The transport sector remains an important source of $PM_{2.5}$. While ongoing transition to electric vehicles (EVs) will help reduce direct tailpipe emissions (Mehlig et al., 2023; Wolfram et al., 2021), non-exhaust particle emissions such as those from brake wear, tyre wear, and road surface abrasion—are not addressed by this shift (Woo et al., 2022). These non-exhaust emissions have become an increasingly more important source of $PM_{2.5}$ in developed cities (Harrison et al., 2021b). To manage non-exhaust emissions, the key is to promote public transport and active travel and reduce traffic volume (DEFRA, 2019; Hwang and Guhathakurta, 2023). Technical and behaviour measures such as eco-driving (e.g., reducing speed and braking), developing low-wear tyres and road surfaces, implementing road sweeping/washing and applying of dust suppressants to road surfaces may also help (Harrison et al., 2021b).

As mentioned earlier, secondary aerosols accounted for a quarter of the total $PM_{2.5}$ mass. These aerosols are influenced by industrial combustion (such as power generation) and agricultural activities (notably $NH_3$ emissions). Both activities are significantly affected by regional or international transport of air pollutants. Secondary aerosols are also indirectly associated with traffic emissions ($NO_2$ to $NO_3^-$ conversion). Therefore, establishing regional and local emission reduction targets can help in reducing secondary aerosol formation. This could be achieved by promoting more sustainable farming practices and transitioning towards cleaner technologies.

Mazzeo et al. (2022) carried out a modelling study employing the WRF-CMAQ to simulate the effects of emission reductions in the West Midlands (alone). Their results suggest that a combined reduction in wood burning, industrial, and agriculture emissions could lead to a more than 50% reduction in $PM_{2.5}$ concentrations if applied nationally and regionally. This is consistent with the current study, which also confirms that local and regional policy interventions may lead to a major (>50%) reduction in $PM_{2.5}$ concentrations. This could also help achieve air quality targets by reducing $PM_{2.5}$ exposure by 35% by 2040.

We also estimated the approximate number of annual deaths attributed to the identified source factors of the ambient $PM_{2.5}$ sampled in the West Midlands using a novel Air quality Lifecourse Assessment Toll (AQ-LAT) (Hall et al., 2024). The study suggested that the estimated annual mortality attributable to $PM_{2.5}$ in the West Midlands region is 2070. Assuming equal toxicities per unit mass for different $PM_{2.5}$ sources (an assumption which may not be correct), the highest mortality was associated with $PM_{2.5}$ exposure resulting from secondary components and wood burning emissions. As discussed in section 4.2, the majority of wood burning observed in this study was related to residential heating. Therefore, locally targeted interventions, raising awareness about residential wood burning, and enhancing smoke control areas in the West Midlands could significantly reduce the mortality attributable to air pollution and saves lives.

## 5. Conclusion

The study investigates the sources of $PM_{2.5}$ in the West Midlands using multiple methods. We found that organics (36–38%), secondary inorganics (35–36%), EC (6–10%), and dust (10%) comprised most of $PM_{2.5}$ by mass. While $(NH_4)_2SO_4$ contributions decreased significantly, $NH_4NO_3$ levels are similar to those observed more than 10 years ago at the same location, despite reductions in $NO_x$ emissions. The dominant sources of $PM_{2.5}$ include biomass burning (25%), resuspended dust and traffic emissions (22%), and secondary aerosols (25%). We found a strong similarity of biomass burning aerosol estimated from PMF analysis and the Aethalometer model, confirming the substantial increase in the contribution of biomass burning to $PM_{2.5}$ in the urban atmosphere. Additionally, the elevated concentrations of biomass burning at one of the sampling sites may be associated with residents' lifestyle, suggesting that changes in behaviour regarding domestic burning are essential to curb $PM_{2.5}$ concentrations and should be carefully considered in policymaking. The study also suggest that local and regional policy interventions may lead to a major (>50%) reduction in $PM_{2.5}$ concentrations. Health impact assessments indicated that reducing residential wood burning could significantly decrease mortality and life-years lost due to air pollution in the West Midlands region.

## CRediT authorship contribution statement

**Deepchandra Srivastava:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Supattarachai Saksakulkrai:** Formal analysis. **W. Joe F. Acton:** Investigation, Data curation. **Daniel J. Rooney:** Methodology. **James Hall:** Investigation. **Siqi Hou:** Methodology. **Mark Wolstencroft:** Methodology. **Suzanne Bartington:** Writing – review & editing. **Roy M. Harrison:** Writing – review & editing. **Zongbo Shi:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization. **William J. Bloss:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Conceptualization.

## Declaration of competing interest

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2024.120963.

## Data availability

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00001173

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