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Associations between short-term exposure to airborne carbonaceous particles and mortality: A time-series study in London during 2010–2019

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ABSTRACT

Exposure to ambient particulate matter (PM) has been identified as a major global health concern; however, the importance of specific chemical PM components remains uncertain. Recent studies have suggested that carbonaceous aerosols are important detrimental components of the particle mixture. Using time-series methods, we investigated associations between short-term exposure to carbonaceous particles and mortality in London, UK. Daily counts of non-accidental, respiratory, and cardiovascular deaths were obtained between 2010 and 2019. For the same period, daily concentrations of carbonaceous particles: organic (OC), elemental (EC), wood-burning (WC), total carbon (TC) and equivalent black carbon (eBC) were sourced from two centrally located monitoring sites (one urban-traffic and one urban-background). Generalized additive models were used to estimate the percentage change in mortality risk associated with interquartile range increases in particulate concentrations. Lagged effects up to 3 days were examined. Stratified analyses were conducted by age, sex, and season, separate analyses were also performed by site-type. For non-accidental mortality, positive associations were observed for all particle species at lag1, including statistically significant percentage risk changes in WC (0.51% (95%CI: 0.19%, 0.82%) per IQR (0.68 μ g/m³)) and OC (0.45% (95%CI: 0.04%, 0.87% per IQR (2.36 μ g/m³)). For respiratory deaths, associations were greatest for particulate concentrations averaged over the current and previous 3 days, with increases in risk of 1.70% (95%CI: 0.64%, 2.77%) for WC and 1.31% (95%CI: -0.08%, 2.71%) for OC. No associations were found with cardiovascular mortality. Results were robust to adjustment for particle mass concentrations. Stratified analyses suggested particulate effects were greatest in the summer and respiratory associations more pronounced in females. Our findings are supportive of an association between carbonaceous particles and non-accidental and respiratory mortality. The strongest evidence of an effect was for WC; this is of significance given the rising popularity of wood-burning for residential space heating and energy production across Europe.

1. Introduction

Exposure to ambient particulate matter (PM) has been identified as a major global health concern (Murray et al., 2020). Over the past decades, a large literature has amassed documenting the adverse effects of long-term and indeed, short-term exposure to PM on health (US EPA,

2019; Sacks et al., 2019; Chen and Hoek, 2020; Atkinson et al., 2014).

Historically, studies that have investigated the effects of PM have done so by examining measures of particle mass concentration, such as PM_{10} or $PM_{2.5}$ (particles with an aerodynamic diameter of less than 10 μ m or 2.5 μ m, respectively) since these were regulated and monitored. In more recent years, a greater emphasis has been placed on understanding

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Available online 12 August 2024 0269-7491/© 2024 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/bync-nd/4.0/).

the effects of individual particle species, knowledge of which could aid the development of more targeted abatement policies. However, as of yet, there is insufficient evidence to conclude any particular components of PM is more toxic than another (Sacks et al., 2019; COMEAP, 2022).

Among the largest chemical constituents of PM are the carbonaceous aerosols, accounting for approximately 10–50% of the mass of urban particulates (Putaud et al., 2004; Hama et al., 2022). Typically grouped into two fractions: organic carbon (OC) and elemental carbon (EC) (also referred to as black carbon (BC) when measured using light absorption methods), the carbonaceous aerosols are directly released into the atmosphere as a result of combustion processes: EC being mainly associated with fossil fuels and OC being dominant in the burning of wood and other biomass (McDuffie et al., 2020; Bond et al., 2004). OC can also be formed secondarily via gas-to-particle conversion reactions, e.g., the oxidation of volatile organic compounds (VOCs) (Donahue et al., 2012).

Previous studies have demonstrated relationships between shortterm exposure to carbonaceous particles and adverse health, especially cardio-respiratory diseases. In the meta-analysis by Yang et al., which reviewed evidence from existing time-series of PM composition and health, both BC and OC were shown to be robustly associated with cardiovascular and respiratory mortality, and cardiovascular hospital admissions (Yang et al., 2019). Furthermore, the health risks associated with carbonaceous particles, specifically BC, have been shown to be similar per interquartile range increase in mass concentration (μ g/m³) to those of PM₁₀ and PM_{2.5} (Janssen et al., 2011).

In spite of these findings, there has been limited assessment of the short-term effects of carbonaceous particles in the UK, largely because long time series of data are not readily available. Of the few studies that have been conducted, the primary focus has been traffic emissions, considering only EC/BC, with estimates based on only 2-years of data (Atkinson et al., 2016; Samoli et al., 2016). In this study, we used time-series methods to evaluate associations between daily concentrations of carbonaceous particles and daily all-cause and cause-specific mortality in London, UK, based on data collected over the 10-year period, 2010–2019.

2. Methods

2.1. Health data

For the study period January 1, 2010 to November 17, 2019 (3608 days), details of all deaths registered in the administrative region of Greater London were obtained from the Office for National Statistics (ONS). This information was used to establish a database of daily counts of mortality for all non-accidental (International Classification of Diseases, version 10 (ICD-10: A00-R99), cardiovascular (ICD-10: I00-I99), and respiratory (ICD-10: J00-J99) causes. Sub-aggregations of the data were also created stratifying counts by age (<65 years and \geq 65 years) and sex (male/female).

2.2. Exposure data

Particulate data were sourced from two fixed-site monitoring stations within the London Air Quality Network: Marylebone Road (urban traffic) and North Kensington (urban background). These data consisted of daily average concentrations of organic carbon (OC), elemental carbon (EC), total carbon (TC), and PM mass - including PM_{10} and $PM_{2.5}$. Measurements of equivalent black carbon (eBC) were also obtained. At both sites particulate concentrations were collected according to standardized measurement procedures and, where necessary, were aggregated into daily metrics in line with Local Air Quality Management (LAQM) guidance (Tompkins et al., 2021; Defra, 2021). Further details are provided in Supplementary Table S1.

Measurement data were obtained for the whole study period, January 2010–November 2019, with the exception of EC, OC, and TC at North Kensington, that were only available up to November 2018 due to relocation of monitoring equipment (see Supplementary Table S2 for data availability).

Where possible, missing values were imputed for days where particle concentrations were available at only one of the two study monitoring sites. Here, we opted for a simple ratio-based approach with missing values at site *X* calculated by multiplying the corresponding values at site *Y* by the overall monthly average between the two sites. See equation below:

$$C_{xmd} = C_{ymd} * (C_{xm}/C_{ym})$$

where x/y represent the two monitoring stations, *m* the month and *d* the day of measurement.

For cases where missing values fell within months with less than 50% data availability, multiplier ratios were based on seasonal as opposed to monthly averages.

To estimate a daily average city-wide concentration, mean measurements was taken across the two sites.

As an additional metric of carbonaceous exposure, daily concentrations of PM from wood-burning, hereafter wood-burning carbon (WC), were also obtained for the study period (available from January 2010–November 2019). These measurements were derived using dual wavelength aethalometers to estimate the concentration of PM_{2.5} from wood and solid fuel burning. The method relies on PM_{2.5} from wood and solid fuel burning being more strongly light absorbing at short wavelengths when compared with soot from diesel emissions. Notably, values were only able to be reliably calculated for North Kensington. Further details regarding the derivation of WC concentrations can be found elsewhere (Sandradewi et al., 2008; Fuller et al., 2014; Font et al., 2022), a brief summary of the measurement methodologies is provided in Supplementary Table S1.

2.3. Meteorology

Meteorological data, including daily mean temperature (°C) and relative humidity (%), for London City Airport were extracted from the NOAA Integrated Surface Database (ISD).

2.4. Statistical analysis

Generalized additive quasi-Poisson regression models (GAMs) (Hastie, 2017) were used to quantify associations between daily concentrations of carbonaceous particles and mortality by cause. GAM extends the traditional generalized linear modelling approach by replacing linear predictors with unspecified nonparametric functions (Dominici et al., 2002). The GAM approach has been widely adopted in time-series studies of air pollution and health, mainly due to its inherent flexibility, enabling nonparametric adjustments for nonlinear confounding effects of seasonality, trends, and weather variables (Dominici et al., 2002). In the current study, we specified a natural spline function of calendar time with 8 degrees of freedom (df) per year to control for seasonality and long-term trends. Meteorological confounding was addressed using smooth splines of daily mean temperature (temp_t) and relative humidity (rht) with 6 df and 3 df respectively. The smoothing applied to each of the splines was selected according to previous literature (Bhaskaran et al., 2013; Touloumi et al., 2006). Additional adjustments for day of the week (dow) and holiday effects (hols) were incorporated into the models as indicator variables. The basic form of the models was as follows:

 $log[E(Y_t)] = intercept + \beta X_t + s(time, df = 8/year) + s(temp_t, df = 6) + s(rh_t df = 3) + dow + hols$

Where, $E(Y_t)$ denotes the expected number of deaths on day t, X the concentration of carbonaceous particulates on day t, and β the effect estimate.

To investigate for latency in the exposure-response, particle

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concentrations were lagged between the current and previous 3 days (single-day: lag0 to lag3 and moving average: lag03). This period was selected in accordance with previous studies, which have shown associations between PM and mortality to be greatest within the 0–3-day lag interval (Atkinson et al., 2016; Atkinson et al., 2010).

In line with previous studies, age- and sex-specific analyses were performed to identify potentially vulnerable groups. Stratified analyses were also performed by season (warm: May to October or cold November to April), with models adjusted for time segmentation, temperature, and humidity. To assess whether estimates differed according to exposure site type, analyses were repeated using measurement data from Marylebone Road and North Kensington individually.

Two-pollutant models were used to evaluate the extent to which estimated associations were confounded by particle mass, with EC, OC, and TC being adjusted for PM_{10} and eBC and WC for $PM_{2.5}$ reflecting the particle size for each measurement. Sensitivity to modelling

assumptions was tested by varying the degree of smoothing applied to both meteorological (2–10 df for daily mean temperature and relative humidity) and temporal trend adjustments (4–12 df/year). Analyses were also performed using the raw exposure data, that is, prior to the imputation of any missing values. Diagnostic plots were used to assess for deviances from linearity.

All models were fitted in R (version 4.0.4; R Development Core team (2021)), using functions from the GAM package (version 1.20; T. Hastie (2020)). Effect estimates were expressed as a percentage change in the risk of mortality per interquartile range (IQR) increase in mass particle concentrations. Significance was evaluated at a confidence level of 95%.

Table 1

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Organic Carbon (24-h Avg. µg/m ³) North Kensington 96% 5.99 2.63 1.34 5.45 29.04 3.04 Marylebone Road 96% 3.47 2.09 0.58 2.91 29.65 2.08 City-Average 99% 4.76 2.02 1.18 4.22 2.945 2.36 City-Average 99% 4.76 2.02 1.88 4.22 2.945 2.36 Total Carbon (24-h Avg. µg/m ³) 4.11 2.74 0.78 3.69 3.6.68 2.76 City-Average 99% 7.60 3.41 1.57 6.94 37.40 4.12 Equivalent Black Carbon (24-h Avg. µg/m ³) 7.60 3.66 2.34 0.78 3.69 3.6.68 2.76 North Kensington 100% 1.22 1.01 0.13 0.93 12.81 0.90 City-Average 92% 0.86 0.82 0.002 0.60 1.002 0.68 Voot-burning Carbon (24-h Avg. µg/m ³) 2.4 8.4 <td< td=""><td>City-Average</td><td>99% ^b</td><td>2.85</td><td>1.61</td><td>0.32</td><td>2.54</td><td>10.74</td><td>2.10</td></td<>	City-Average	99% ^b	2.85	1.61	0.32	2.54	10.74	2.10
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Total Carbon (24-h Avg, µg/m ³) International and the second secon	City-Average	99% ^b	4.76	2.20	1.18	4.22	29.45	2.36
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Note that again the problem of the	North Kensington	93%	19 40	10.92	3.16	16.61	84.65	10.90
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Mortality DataInternational and the late of the late	City-Average	96%	15.57	9.66	2.76	12.73	98.98	9.31
Non-accidental Total 100% 126 18.79 76 124 215 24 <65 years	Mortality Data							
Total100%126 18.79 76 124 215 24 <65 years	Non-accidental							
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$\begin{array}{c c c c c c c c c c c c c c c c c c c $	<65 years	100%	22	5.11	8	22	46	7
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Cardiovascular Total 100% 37 7.69 16 36 74 11 <65 years	Female	100%	64	11.50	31	63	126	15
Total 100% 37 7.69 16 36 74 11 <65 years	Cardiovascular							
<65 years 100% 5 241 0 5 17 3	Total	100%	37	7.69	16	36	74	11
	<65 years	100%	5	2.41	0	5	17	3
> 65 years 100% 31 7.05 13 31 66 10	> 65 years	100%	31	7.05	13	31	66	10
Male 100% 19 4.93 5 19 39 7	Male	100%	19	4.93	5	19	39	7
Female 100% 18 4.96 3 17 39 7	Female	100%	18	4.96	3	17	39	7
Respiratory	Respiratory							
Total 100% 18 6.39 4 17 54 7	Total	100%	18	6.39	4	17	54	7
<65 years 100% 2 1.34 0 1 9 1	<65 years	100%	2	1.34	0	1	9	1
≥ 65 years 100% 16 5.96 4 15 50 8	\geq 65 years	100%	16	5.96	4	15	50	8
Male 100% 9 3.68 0 8 30 5	Male	100%	9	3.68	0	8	30	5
Female 100% 9 4.06 0 9 32 5	Female	100%	9	4.06	0	9	32	5
Meteorological Data (24-h Avg)	Meteorological Data (24-h Avg)							
Temperature (°C) 100% 12.24 5.54 -3.00 12.21 29.06 8.44	Temperature (°C)	100%	12.24	5.54	-3.00	12.21	29.06	8.44
Relative humidity (%) 100% 73.33 9.99 41.93 73.68 99.48 14.51	Relative humidity (%)	100%	73.33	9.99	41.93	73.68	99.48	14.51

Footnote.

Abbreviations: Avg = Average, IQR = Inter-Quartile Range, $PM_{2.5}$ = Particulate matter with aerodynamic diameter <2.5 µm, in mass concentration (µg/m³), PM_{10} = Particulate matter with aerodynamic diameter <10 µm, in mass concentration (µg/m³), SD = Standard Deviation.

^a Availability of data is expressed as a percentage of the total number of study days (3608 days) and was calculated following the imputation of missing values. ^b Measurement data available between January 1, 2010–November 12, 2018 - availability evaluated up to last recorded date.

3. Results

3.1. Descriptive analyses

Descriptive characteristics of the study exposure and health outcome data are shown in Table 1. Between January 1, 2010 and November 17, 2019, a total of 456,300 non-accidental deaths were registered in Greater London, equating to an average of 126 deaths per day (IQR = 24). The mean daily number of deaths from cardiovascular causes was 37 (IQR = 11) and 18 (IQR = 7) for respiratory causes.

Particulate concentrations at Marylebone Road were greater than those at North Kensington, as expected. Across the sites, daily mean concentrations (and IQR) of EC, OC, eBC, and TC were 2.85 (2.10), 4.76 (2.36), 3.66 (3.11) and 7.60 (4.12) μ g/m³, respectively, while the average daily concentration of WC at North Kensington was 0.86 (0.68) μ g/m³. Particle concentrations varied seasonally, peaking in the winter, this was particularly evident for WC and eBC (Supplementary Fig. S1) as observed and discussed in Font et al. (2022) (Font et al., 2022). Downward trends in particle concentration were observed over the study period, most notably for eBC, with annual average concentrations decreasing from 5.16 μ g/m³ in 2010 to 1.42 μ g/m³ in 2019. Large reductions were also observed in EC (from 3.88 μ g/m³ in 2010 to 1.69 μ g/m³ in 2018; Supplementary Table S3).

Pearson correlation coefficients (r) between daily concentrations of carbonaceous particles, PM_{10} and $PM_{2.5}$, are shown in Table 2 (site-specific correlations are shown in Supplementary Table S4). High correlations were observed amongst the carbonaceous species, with the exception of WC with EC and eBC (r = 0.38). Daily concentrations of $PM_{2.5}$ were strongly correlated with those of OC (r = 0.79) and TC (r = 0.70) but less so with WC (r = 0.61), eBC (r = 0.47), and EC (r = 0.42). Similar correlations were observed for PM_{10} .

3.2. Regression analyses

Fig. 1 shows the estimated associations between daily concentrations of carbonaceous particles and daily mortality at different lag times. Results are also provided numerically in Supplementary Tables S5–S7.

For non-accidental mortality, associations from single day lagged models peaked at lag 1, including statistically significant percentage risk changes observed for both WC (0.51% (95% Confidence Interval (CI): 0.19%–0.82%)) and OC (0.45% (95% CI: 0.04%–0.87%)). Estimates based on particle concentrations averaged over the current and previous 3 days (lag03) were null, with the exception of WC (0.71% (95% CI: 0.26%–1.16%)).

Associations for respiratory mortality tended to be larger than those

Table 2

Pearson correlation coefficients between daily city-wide average concentrations of carbonaceous particles and PM mass for London between 2010 and 2019.

	EC	OC	TC	eBC	WC	PM_{10}	PM _{2.5}
EC	1.00	-	-	-	-	-	-
OC	0.61	1.00	-	-	-	-	-
TC	0.86	0.92	1.00	-	-	-	-
eBC	0.90	0.60	0.81	1.00	-	-	-
WC	0.38	0.71	0.63	0.38	1.00	-	-
PM_{10}	0.46	0.79	0.72	0.52	0.59	1.00	-
PM _{2.5}	0.42	0.79	0.70	0.47	0.61	0.95	1.00

Footnote: Correlations calculated based on daily average city-wide particle concentrations, except those for WC, where data was only available for North Kensington. Values represent correlations for the full study period (01/01/2010-17/11/2019), except pollutant pairs including OC, EC, or TC which were up to November 12, 2018. Abbreviations: eBC = Equivalent Black Carbon, EC = Elemental Carbon, OC = Organic Carbon, Carbon, PM2.5 = Particulate matter with aerodynamic diameter <2.5 µm, in mass concentration (µg/m³), PM10 = Particulate matter with aerodynamic diameter <10 µm, in mass concentration (µg/m³), TC = Total Carbon, WC = Wood-burning.

for non-accidental mortality, but with wider confidence intervals. Statistically significant percentage risk changes were observed for WC at lag 1, 2, and 3 and OC at lag 2. Estimates from cumulative lag models were larger than those at any single day lag. IQR increases in the concentration of WC lagged at lag03 were associated with a statistically significant increase in the risk of respiratory mortality of 1.70% (95% CI: 0.64%– 2.77%). Similar magnitudes of association were observed for TC (1.28% (95%CI: -0.39%–2.98%)), OC (1.31% (95%CI: -0.08%–2.71%)), and eBC (1.32% (95%CI: -0.92%–3.61%)).

Little evidence was observed for an association with cardiovascular mortality. Estimates were generally small and negative with confidence intervals spanning zero.

3.3. Stratified analyses

Estimates from stratified analyses by age, sex, and season, are shown in Supplementary Figs. S2–S4. No meaningful differences were observed when stratifying by age. In contrast, stratification by sex indicated that associations with respiratory mortality tended to be more pronounced in females, while season-specific analyses revealed effects of OC and TC were greater during the warmer months (May–October). Site-specific analyses were broadly consistent with the main study results (Fig. 2), although more prominent associations were observed for eBC and respiratory mortality across all lags when using data collected at the North Kensington monitoring site (see Fig. 3).

3.4. Sensitivity analyses

Effect estimates were robust to adjustment for PM mass concentration (PM₁₀ or PM_{2.5}) and, in some instances, increased in magnitude (Fig. 2; Supplementary Tables S5–S7). This was most notable for WC with adjustment resulting in significantly increased risk of cardiovascular mortality at single day lags 2, 3, and cumulative lag03 (1.23% 95% CI: 0.19%–2.28%).

Examining sensitivity to key modelling assumptions, such as the degree of smoothing applied to time trends and weather-related variables did not yield any significant differences (data not shown), neither did repeating the analyses using the raw exposure data. Diagnostic plots supported a linear relationship between the carbonaceous particulates and mortality.

4. Discussion

4.1. Main findings and interpretation

In this study, we investigated associations between daily concentrations of carbonaceous particles and daily mortality, using data collected for London between 2010 and 2019. Based on our analyses, we found short-term exposure to carbonaceous particles to be associated with small, but statistically significantly increased risks of mortality from both non-accidental and respiratory causes. Less evidence was found for an association with cardiovascular mortality.

The most convincing of our findings was for an effect of carbonaceous particles from wood-burning, with significantly increased risks of non-accidental and respiratory mortality observed at single-day lag 1, 2, and 3 and cumulative lag03. These associations persisted after adjustment for particle mass and, in some instances, increased in magnitude, indicating a level of specificity of the effects of WC on health.

Most studies that have examined the health effects of wood-burning particles are related to wildfires. Episodic increases in the levels of PM from wildfires have been linked with a variety of detrimental health outcomes, including the exacerbation of respiratory conditions, such as asthma and COPD, and premature deaths (Reid et al., 2016; Karanasiou et al., 2021). Experimental studies have also shown that wildfire PM is more toxic than conventional PM, with a greater capacity to induce inflammation and oxidative stress (Wegesser et al., 2010).



Fig. 1. Percentage change (and 95% CI) in (a) non-accidental, (b) respiratory, and (c) cardiovascular mortality per IQR increase in the concentrations of carbonaceous particles lagged between the current and previous 3 days. All estimates are based on daily city-wide particle concentrations for London, except those for WC (measurement data only available for North Kensington). IQRs for EC, OC, eBC, TC, and WC were 2.10, 2.36, 3.11, 4.12, and 0.68 µg/m3, respectively. Abbreviations: CI = confidence interval, EC = Elemental Carbon, eBC = Equivalent Black Carbon, IQR = interquartile range, OC = Organic Carbon, TC = Total Carbon, WC = Wood-burning Carbon.



Fig. 2. Site-specific percentage changes (and 95% CI) in the risk of (a) non-accidental, (b) respiratory, and (c) cardiovascular mortality per interquartile range increase in the concentrations of carbonaceous particles lagged between the current and previous 3 days. All estimates are based on daily city-wide particle concentrations for London, except those for WC (measurement data only available for North Kensington). IQRs for EC, OC, eBC, and TC at Marylebone Road were 3.66, 3.04, 5.47, and $6.35 \mu g/m^3$, and at North Kensington were 0.65, 2.08, 0.90, and 2.76 $\mu g/m^3$. Abbreviations: CI = confidence interval, IQR = interquartile range, eBC = Equivalent Black Carbon, OC = Organic Carbon, TC = Total Carbon, WC = Wood-burning Carbon.

In London, WC has been shown to originate from diffuse urban sources, with concentrations peaking in the evenings and on weekends, reflective of residential wood combustion (RWC), e.g., from woodstoves and fireplaces (Fuller et al., 2014; Font et al., 2022). With regard to the health effects of PM from RWC, several time-series studies have reported associations between PM and adverse respiratory health outcomes in cities where RWC is a major contributor to outdoor air pollution (Boman et al., 2003; Naeher et al., 2007; Sigsgaard et al., 2015). Similarly, community intervention studies have demonstrated health improvements following woodstove change-out programmes (Johnston et al., 2013), there also exists some evidence linking short-term exposure to potassium (K), a known tracer of RWC, with increased risks of all-cause and respiratory mortality (Basagaña et al., 2015; Li et al., 2015).

The toxicological evidence for the effects of PM from RWC, however, remains unclear. While some studies have reported associations with increased levels of airway inflammation and oxidative stress (Barregard et al., 2008), others have not (Riddervold et al., 2012). These inconsistencies have been attributed, at least in part, to differences in how PM is generated during the experiments (Kocbach Bølling et al., 2009). Studies have shown that the physiochemical properties of PM from RWC can vary greatly according to combustion conditions and technology, as well as fuel type, e.g., wood species and water content (Kocbach Bølling

et al., 2009; World Health Organization, 2021).

Our results were also supportive of an adverse effect of OC on nonaccidental at lag 1 and respiratory mortality at lag 2, with increases of 0.45% (95% CI: 0.04%–0.87%) and 1.00% (95% CI: 0.05%–1.95%) per IQR (2.36 μ g/m³), respectively. These values are consistent with those reported in the recent meta-analysis by Yang et al., which linked OC to increases in non-accidental mortality of 0.35% (95% CI: 0.09%–0.61%) and respiratory mortality of 0.68% (95% CI: 0.97%–1.89%) per median IQR (2.83 μ g/m³) (Yang et al., 2019). Studies have also linked acute exposure to OC with increases in non-accidental (Yang et al., 2019; Zhang et al., 2022) and respiratory hospital admissions (Zhang et al., 2022; Peng et al., 2009). In London, both vehicle exhaust and woodsmoke emissions are considered important sources of OC (Yin et al., 2015); and thus, the observed associations for OC may be more broadly reflective of an adverse effect of primary combustion particles.

We did not find evidence of a significant association between shortterm exposure to EC or eBC and mortality. These components are typically considered markers of road traffic emissions and, contrary to the findings in this study, have been consistently linked with adverse health outcomes (Chen and Hoek, 2020; McDuffie et al., 2020; Achilleos et al., 2017).

Comparing our results to those of an earlier study that investigated



Fig. 3. Comparison of estimates from single-pollutant and PM adjusted models for the percentage change (and 95% CI) in the risk of natural (a), respiratory (b), and cardiovascular (c) mortality associated with an IQR increases in concentrations of carbonaceous particles lagged between the current and previous 3 days. All estimates are based on daily city-wide particle concentrations for London, except those for WC (measurement data only available for North Kensington). IQRs for EC, OC, eBC, TC, and WC were 2.10, 2.36, 3.11, 4.12, and 0.68 μ g/m3, respectively. Abbreviations: CI = confidence interval, EC = Elemental Carbon, eBC = Equivalent Black Carbon, IQR = interquartile range, OC = Organic Carbon, PM =Particle Mass, TC = Total Carbon, WC = Wood-burning Carbon.

associations between EC and BC and mortality in London between 2011 and 2012, the authors did not find evidence of an effect on all-cause or cardiovascular mortality (Samoli et al., 2016). However, they did report significant associations for respiratory mortality (EC: 2.66% (95% CI: 0.11 to 5.28), BC: 2.72% (95% CI: 0.09 to 5.42) per IQR). The differences in the estimated health effects between our study and those described above are likely due to the declines in the concentrations of EC and (e)BC over time (see Supplementary Table S3), primarily driven by the introduction of diesel particle filters as part of the Euro 5 vehicle emission standards (Kamara and Harrison, 2021; Ciupek et al., 2021). When restricting our analyses between 2011 and 2012, we obtained similar results to the study by Atkinson et al. (data not shown). Further confirmatory research is needed to identify whether the introduction of particle filters is causatively linked to the observed reductions in the associations between EC and (e)BC and mortality.

4.2. Interpretation of stratified analyses

In stratified analyses, associations for OC and TC were shown to vary by season, with greater effects observed in the warmer months (May to October). While this seems somewhat counterintuitive, given their concentrations were generally lower in the summer, studies have reported similar results when examining season-specific PM effects in other European cities (Katsouyanni et al., 1997; Stafoggia et al., 2008). One explanation for these findings is that higher temperatures may impair an individual's physiological response to air pollution (Gordon, 2003), thus resulting in greater levels of susceptibility, even on low(er) pollution days. Work on mediation and effect modification by climatic factors remains ongoing. It has also been reasoned that greater summer-time effects may be due to increased outdoor activity, raising exposure time, as well as lower background mortality rates, resulting in a larger pool of susceptible individuals (Chen et al., 2013).

Associations between WC and mortality also appeared to be slightly greater in the warmer season. This finding is unexpected given WC concentrations in summer are minimal, as shown previously in Font et al. (2022) (Font et al., 2022). The sources of WC in the summer likely differ from those in the winter, with a higher contribution from bonfires, fire pits, and patio heaters, rather than emissions from wood-burning stoves. The warmer period is also associated with greater formation of secondary organic aerosol from VOCs; biogenic emissions of VOCs, such as isoprene, are also enhanced in summer (Redington and Derwent, 2013). The increase in the effect size for WC in the warmer period raises the question of whether summer WC is serving as a proxy for emissions from other sources. Differences in the sources of WC between seasons are likely to cause variations in the chemical composition of WC, which may account for the differences observed in the health effects. However, this relationship is yet to be established. In addition, lower concentrations and change in optical properties in the summer months may increase uncertainty in exposure assessment.

Our results also suggested that females were disproportionately affected by exposure to carbonaceous particles, with elevated risks of respiratory mortality. While the literature is inconsistent with regard to sex-specific PM effects (Shin et al., 2022; Clougherty, 2010), the observed heterogeneity may result from differing lifestyle factors and behaviours between men and women, such as occupation, socioeconomic status, and physical activity levels. Biological factors such as lung size/capacity and air-way reactivity may also play a role (Corsini et al., 2019).

Stratified analyses were also conducted by site type. Typically, timeseries studies only use data collected at urban background stations; however, these sites may not entirely capture the day-to-day variability of air pollution in a metropolitan city like Greater London, given residents may live, work, or commute near busy roads. To better reflect the daily average concentrations of air pollution in London, we combined data from both a traffic and background monitor to calculate the main exposure. When comparing the main study results to those from the sitespecific analyses, we observed broadly consistent patterns of association. Having said this, associations between eBC and respiratory mortality were greater at the urban background site than at the traffic site. One explanation for this finding is that the daily variation in eBC at North Kensington may be more representative of the daily variation of the true population exposure to eBC, resulting in less measurement error and, hence, better power to observe an exposure-response association. Further research is needed to clarify these findings.

4.3. Implications for public health and air quality policy

This study contributes towards the growing understanding of the health effects of specific components and sources of PM that can be used to inform more targeted air-quality management policies.

Of particular importance are our findings related to the effects of carbonaceous particles from wood burning. Over the past decade, wood burning for domestic heating and energy production purpose has increased in popularity across Europe and is projected to continue to do so into the near future (Sigsgaard et al., 2015; Corsini et al., 2019). This is partially due to the introduction of climate change policies promoting the burning of wood/biomass as a "green" source of energy production, but also the rising costs of fuel, forcing individuals to seek alternative ways to heat their homes (Fuller et al., 2013; Vouitsis et al., 2015).

The potential upsurge in wood/biomass burning activity has been noted as a cause for concern, threatening to undermine policies aimed at reducing particle emissions in the UK/EU (Sandradewi et al., 2008; Sigsgaard et al., 2015). For London, recent studies have demonstrated downward trends in WC (Fuller et al., 2014), possibly due to a shift towards cleaner wood-burning technologies, but the association between WC and mortality observed in this study indicates that this metric should be monitored going forward. This would enable policymakers to track changes in WC, assess whether the adoption of more efficient woodstoves is sufficient to reduce emissions, and, if not, evidence a need for further policy development.

4.4. Strengths and limitations

A key strength of our study was the length of time over which exposure data were available. Given that the unit of observation in timeseries studies is days, a larger number of study days equates to greater statistical power and thus, the ability to estimate the effects of particle exposures with greater precision.

A further strength of our analysis was the fact that through use of novel exposure assessment methodologies, we were able to directly quantify the effects of PM from RWC on health. This has previously proven difficult due to a lack of long-term measurements of specific indicators of wood-burning particles (Kocbach Bølling et al., 2009).

With regards to the limitations of our study, firstly, we investigated associations between carbonaceous particles and mortality in London; therefore, findings may have limited generalisability to other cities and populations across the UK/Europe. Secondly, using concentration data from two fixed-site monitoring stations as a proxy for population-level exposure will have introduced some degree of exposure measurement error (ME). Exposure ME is a major concern in air pollution epidemiology and is known to bias effect estimates, generally towards the null (Samet et al., 2000; Zeger et al., 2000). The effects of ME are particularly great when considering spatially heterogeneous exposures, e.g., those associated with local sources such as road traffic, and may have led to more pronounced attenuation of estimates for EC/BC (Zeger et al., 2000). While this is less of an issue for WC, given concentrations have been shown to be homogenous across London (Fuller et al., 2014), disparities between ambient concentrations and personal exposures to WC are likely to have resulted in further ME bias (Evangelopoulos et al., 2021). Lastly, having studied the effects of carbonaceous particles on broad categories of disease outcome, namely mortality from all respiratory and cardiovascular causes, we may have failed to uncover relationships for specific sub-types of disease. For example, previous studies have shown significant associations between carbonaceous particles and deaths from ischemic heart disease and stroke (Kim et al., 2015; Lin et al., 2016). Additional work to quantify associations between carbonaceous particles and additional health outcomes would be beneficial.

5. Conclusion

In summary, our results are supportive of an association between short-term exposure to carbonaceous particles and all-cause and respiratory mortality. The strongest evidence of an effect was for WC; this is particularly important given the rising popularity of wood burning for residential heating and energy production across the UK and Europe. Additional studies are required to confirm our findings.

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Ethical approval

Ethical approval was not required. The study was observational and used anonymised mortality data.

CRediT authorship contribution statement

Rachael Piper: Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. Anja Tremper: Writing – review & editing, Resources, Methodology, Data curation, Conceptualization. Klea Katsouyanni: Writing – review & editing, Supervision, Methodology, Conceptualization. Gary W. Fuller: Writing – review & editing, Methodology, Conceptualization. David Green: Writing – review & editing. Anna Font: Writing – review & editing, Resources, Data curation. Heather Walton: Writing – review & editing. Ioar Rivas: Writing – review & editing, Resources. Dimitris Evangelopoulos: Writing – review & editing, Supervision, Methodology, Formal analysis, 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.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2024.124720.

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