How COVID-19 related policies reshaped organic aerosol source contributions in central London

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Abstract

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Organic aerosol (OA), a major component of submicron particulate matter (PM₁), has significant impacts on both human health and climate. Quantifying its sources is therefore crucial for developing effective mitigation strategies. Particulate matter (PM) poses both health and climate risks. Understanding pollution sources is therefore crucial for effective mitigation. Positive matrix factorisation (PMF) applied to aerosol chemical speciation monitor (ACSM) mass spectral data offers a robust approach for quantifying OA sources. Positive Matrix Factorization (PMF) of Aerosol Chemical Speciation Monitor (ACSM) data is a powerful tool to quantify organic aerosol (OA) sources. A year-long study of ACSM data from London's Marylebone Road monitoring station during the COVID-19 pandemic provides insights into the impact of lockdown and the Eat Out To Help Out (EOTHO) scheme, which offered support to the hospitality industry during the pandemic, on PM composition and OA sources. Five OA sources were identified including hydrocarbon-like OA (HOA, traffic-related, 11% to OA), cooking OA (COA, 20%), biomass burning OA (BBOA, 12%), more-oxidized oxygenated OA (MO-OOA, 38%), and less-oxidized oxygenated OA (LO-OOA, 21%). Lockdown significantly reduced HOA (-52%), COA (-67%), and BBOA (-42%) compared to their pre-COVID levels, while EOTHO doubled COA (+100%) compared to the post-lockdown period. However, MO-OOA and LO-OOA were less affected, as these primarily originated from long-range transport. This research has highlighted-demonstrated the importance of commercial cooking as a significant source of OA (20%) and PM₁ (9%) in urban areas. The co-emission of BBOA with COA observed in Central London demonstrates showed a similar diurnal cycle and response to the EOTHO policy, indicating that cooking activities might be currently underestimated and contribute to urban BBOA. Therefore, more effort is required to

- quantify this source and develop targeted abatement policies to mitigate emissions as currently
- 39 limited regulation is in force.

1 Introduction

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42 Atmospheric particulate matter (PM) are tiny particles suspended in the air, which can not only 43 impact the climate directly and indirectly (IPCC, 2021; Seinfeld et al., 2006), but also and cause 44 adverse human health effects to human (Kelly and Fussell, 2012; World Health Organization, 45 2021). The PM present in urban areas, such as London, consist of various constituents is emitted 46 directly or indirectly from a wide range of natural and anthropogenic sources, can be changed 47 through atmospheric reactions and remain airborne for many days. It is consequently a complex 48 mixture -including inorganic species (metals, minerals, black carbon, nitrate, sulphate, etc.) and 49 thousands of organic species (complex mixture of thousands of compounds whose origins remain 50 too complicated to fully quantify). PM_{2.5} (PM with aerodynamic diameter smaller than 2.5 µm) is 51 strongly associated with increased risks of cardiovascular and respiratory related mortalities and 52 hospital admissions (Dominici et al., 2006; Joo et al., 2024; Pye et al., 2021; Wei et al., 2022, 2024)... Some studies (Lippmann et al., 2013; UK Health Security Agency (UKHSA), 2022) have 53 begun to demonstrate that some PM constituents and sources have stronger associations with a 54 55 range of health endpoints metrics, including mortality, morbidity, and toxicities although the 56 evidence remains inconsistent (Kelly and Fussell, 2012; Liu et al., 2023; Vasilakopoulou et al., 2023). With European Environment Agency has reported that 99% of the urban population in 57 Europe are still exposed to polluted air with annual PM_{2.5} (PM with aerodynamic diameter smaller 58 than 2.5 µm) concentrations exceeding the WHO air quality guideline, 5 µg/m³ (European 59 60 Environment Agency, 2024; World Health Organization, 2021), delivering clean air is a target for 61 European and international governments according to the EU air quality directive (European 62 Union, 2024). As the most health-relevant air pollutant, PM_{2.5} has shown strong associations with cardiovascular and respiratory related mortalities and hospital admissions. Several studies have 63

64 demonstrated that different constituents/sources contribute to health effects differently with 65 varying toxicities. However, delivering publicly acceptable policies to improve air quality remains challenging (Mebrahtu et al., 2023; Oltra et al., 2021). TTherefore, targeting the specific 66 composition/sources of PM that are most health-relevant could be athe morest coost-effective (Wu 67 et al., 2023) way to mitigate its adverse health effects, more easily communicated and more 68 69 publicly acceptable approaches (Pinakidou, 2025) to improve public health. It is therefore 70 important to better quantify the sources of PM and understand how they respond to policy interventions. 71 72 The COVID-19 pandemic is a natural experiment to assess the impact of policies which, while not 73 aiming to reduce PM_{2.5} concentrations, Click or tap here to enter text. Coronavirus disease 19 (COVID-19) started to spread rapidly worldwide since the first case was identified in Wuhan, 74 75 China late in 2019. Many countries implemented measures to contain COVID cases, which significantly restricted social and economic activities and consequently reduced emissions. In 76 During the UK national lockdown, starting from the end of Mar 26th, 2020, people were ordered 77 to stay at homehome, and all non-essential businesses were closed, including pubs, cafes and 78 restaurants from the end of Mar 26th, 2020. Non-essential shops were allowed to open from on-Jun 79 15th, and the first national lockdown came to an end on Jun 23rd, 2020. However However, and, 80 pubs, restaurants, and cafes were only allowed to open from July 4th, 2020. Subsequently, the Eat 81 Out to Help Out (EOTHO) Scheme was designed to help the hospitality industry; offering a 50% 82 meal discount up to a maximum of £10 and operated Monday to Wednesday during from Aug 3rd 83 to Aug 31st, 2020; https://www.gov.uk/guidance/get-a-discount-with-the-eat-out-to-help-out-84 85 scheme.

86 The UK recorded a 2.5% drop in Gross Domestic Product (GDP) in the first quarter of 2020, partly 87 as people reduced their own activity prior to the legally enforced lockdown measures introduced on Mar 26thnational lockdown. This accelerated to a 19.8% fall in GDP in April to June 2020 and 88 89 household spending fell by over 20% over this period, the largest quarterly contraction on record, 90 which was driven by falls in spending on restaurants, hotels, transport, and recreation (ONS, 2022). 91 The UK Government Eat Out to Help Out (EOTHO) Scheme is examined specifically in this study 92 as it influenced emissions from the commercial cooking sector. It was designed to help the 93 hospitality industry recover from the financial impact of the national lockdown, offering a 50% meal discount up to a maximum of £10, which operated Mon to Wed, Aug 3rd to Aug 31st, 2020; 94 https://www.gov.uk/guidance/get-a-discount-with-the-eat-out-to-help-out-scheme. 95 96 While the impact of these lockdown policies on some air quality metrics was smaller than expected 97 given the large change in emissions (Shi et al., 2021), the abrupt nature of the intervention ensures 98 it is easier to detect than other air quality policies that are more incremental in nature (Mudway et 99 al., 2019). 100 Some studies have investigated the lockdown impacts on chemical composition and sources of 101 PM, which mainly focused on cities in China (Hu et al., 2022; Tian et al., 2021; Xu et al., 2020), 102 a kerbside site in Toronto, Canada (Jeong et al., 2022), and an urban background site in Paris, 103 France (Petit et al., 2021). These studies all resolved primary sources including traffic related 104 emissions, biomass burning emissions from residential heating, cooking emissions (except Paris), 105 and secondary sources from PMF analysis on organic aerosol (OA). Traffic and cooking emissions 106 appeared to decrease during the lockdown in all sites, while biomass burning predominately from 107 residential heating sources in Chinese cities increased as result of remote work and rather early 108 lockdown measures (Jan-Feb 2020) compared to France. Secondary organic aerosol (SOA)

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showed a more complex phenomenon given its abundance in organic components and dynamic spatiotemporal conditions. Overall, the lockdowns resulted in decreased SOA in both northwest cities in China (Tian et al., 2021; Xu et al., 2020) and Paris (Petit et al., 2021) due to lower primary emissions, and therefore fewer SOA formation products. However, Beijing experienced a large increase in SOA concentrations due to increased fossil fuel and biomass burning emissions, longrange transport influences as well as favourable meteorological conditions (high RH, low wind speed and low boundary layer height) for SOA formation during the lockdown period (Hu et al., 2022). Therefore, the lockdown effects on the SOA were dependent on the abundance of primary emissions, long-range transported air masses, and meteorological conditions. To date, there are few studies that investigate how COVID-related policies could have impacted PM chemical composition and sources. Petit et al. (2021). (2021) and Gamelas et al. (2023) are the only two studies in Europe. The unique COVID-related policies in the UK therefore provide d-a rare opportunity to investigate the impacts these policies had on chemical composition and OA sources. To address these issues, wWe used highly time resolved measurements from an air quality supersite located in the Central London from 2019 to 2020, and advanced source apportionment approaches to quantify the PM composition and OA sources before, during and after the UK nationalthe influence of the first lockdown and EOTHO scheme on the PM composition and OA sources. This study provides valuable unique insight into PM sources and composition in a global mega city and how air quality responds to abrupt changes in emissions from different sources. #Importantly, it helps to establish the importance of cooking as a source of PM and uniquely associates biomass burning organic aerosol with commercial cooking emissions. This provides crucial information to policy makers as they attempt to reduce exposure to air pollution in urban areas.-

2 Methodology

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133 2.1 Air quality monitoring supersite in central Central London

The London Marylebone Road supersite (MY, 51.52 N, -0.15 E) is a kerbside air quality monitoring site, one meter away from a busy 6-lane road in central Central London. It is a wellestablished air quality supersite that has consistently generated high-quality air pollution data since 1997 including mass concentration of bulk PM₁, PM_{2.5}, and PM₁₀, as well as PM composition including black carbon, heavy metals, nitrate (NO₃), sulphate (SO₄), ammonium (NH₄), OA, Chloride (Cl), etc. More details of this site can be found at https://ukair.defra.gov.uk/networks/site-info?site id=MY1.

2.2 Instrumentations

Quadrupole ACSM (Q-ACSM, Aerodyne, Ltd., Ng et al. (2011)) with a standard vaporizer provides 30-min mass loadings of chemical species within non-refractory submicron aerosol (NR-PM₁), including NH₄, NO₃, SO₄, Cl, and OA. Sampled particles are focused into a narrow beam using the aerodynamic lens and impacted on a filament surface at 600 °C, where the NR-PM₁ is vaporised and ionised instantly by an electron impact source (70eV). These ions are detected by the RGA quadrupole mass spectroscopy to provide a mass spectrum of NR-PM₁ up to a mass-to-charge ratio (*m/z*) of 148 Th. The mass concentration of different chemical species are calculated using the fragmentation table developed by Allan et al. (2004), updated for Cl following suggestions provided by Tobler et al. (2020), and a composition-dependent collection efficiency (CDCE) correction suggested by Middlebrook et al. (2012) by following the ACTRIS standard operation procedure (https://www.actris-ecac.eu/pmc-non-refractory-organics-and-inorganics). With co-located black carbon (BC) measurement using a PM_{2.5} cyclone with AE33 (Aerosol

Magee Scientific, Ltd.) and PM_1 measurements using FIDAS (Palas, GmbH), we conducted the mass closure for fine particles measurements. The sum of NR-PM₁ and BC (in PM_{2.5}) reproduces PM_1 concentrations well, with a slope of 1.13 and an R^2 of 0.73 (Fig. S1).

2.3 Sampling periods and COVID-related policies

PM₁ chemical composition from Aug 1st, 2019 to Oct 22nd, 2020, was analysed as this covered the first lockdown period (Mar 26th–23 Jun 23rd, 2020) and the EOTHO Scheme (Mon-Wed during from Aug 3rd to Aug 31st, 2020, <u>Table 1 Table 1</u>). In order to isolate the seasonal effects on the PM chemical composition and OA sources from the COVID-related policies, we further split the data based on seasons (<u>Table 1 Table 1</u>). In addition, deweathering analysis has been conducted using "worldmet" R package to remove the meteorological effects (i.e., relative humidity, wind speed, wind direction, and air temperature) on all PM/OA species as shown in the SI (Fig. S8 and Fig. S9). Meteorological effects were considerable, especially for Pre lockdown Spring period, while it does not change the conclusion of the effects from lockdown and EOTHO policies. Therefore, the main results presented in this study are based on the original measurements.

Table 1 Dates of the COVID-related policies in London

COVID Policies	COVID Policies		
	Summer	Aug 1 st –Aug 31 st , 2019	
Duo I caledorum	Fall	Sep 1 st –Nov 30 th , 2019	
Pre-Lockdown	Winter	Dec 1st, 2019–Feb 28th, 2019	
-	Spring	Mar 1 st –Mar 25 th , 2020	
Lockdown	Spring	Mar 26 th –May 31 st , 2020	

Summer	Jun 1 st –Jun 23 rd , 2020
Pre-EOTHO	Jun 24 th –Aug 2 nd , 2020
ЕОТНО	Aug 3 rd –Aug 31 st , 2020
Post-EOTHO	Sep 1 st –Oct 22 nd , 2020
	Pre-EOTHO EOTHO

2.4 Source apportionment

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Source apportionment is a common but powerful approach to identifying and quantifying the emission sources and atmospheric constituents of PM based on measurements. As the sources of inorganic species (black carbon, ammonium, nitrate, chloride, sulphate, etc.) are relatively wellstudied, most of the studies are focused on deconvoluting the sources of OA, which contains thousands of compounds. Positive matrix factorization (PMF) is one of the receptor models that is widely utilized in the field to conduct source apportionment analysis (Jimenez et al., 2009; Zhang et al., 2007). Typically, an Aerodyne aerosol mass spectrometer (AMS, Aerodyne Ltd., USA, Jayne et al., 2000) is used to measure the time series of both inorganic and organic species of nonrefractory PM, in which, organic mass spectra are used for PMF analysis. However, operating an AMS is labour-intense and expensive. In contrast, the aerosol chemical speciation monitor (ACSM, Aerodyne, Ltd., Fröhlich et al., 2013; Ng et al., 2011) has been designed for long-term monitoring purposes with less maintenance and lower capital cost, which has gained popularity across Europe (Chebaicheb et al., 2024; Chen et al., 2022; Laj et al., 2024) and the U.S. (https://ascent.research.gatech.edu/, (Hass-Mitchell et al., 2024; Joo et al., 2024)). Chen et al. (2022) demonstrated a robust protocol to conduct advanced PMF analysis on long-term ACSM datasets, which delivers high-quality and consistent source apportionment results. This study follows this standardized protocol to resolve the OA sources in London by implementing advanced PMF techniques.

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Advanced source apportionment approaches have been used in this study, including rolling positive matrix factorization (PMF), ME-2 with random a-value approach, bootstrap and criteriabased selections (Canonaco et al., 2021; Chen et al., 2022). The standardized protocol of rolling PMF as presented in Chen et al. (2022) was used to ensure high-quality and comparable sources of OA were retrieved in London. Specifically, PMF was firstly done on four different seasons as suggested in Chen et al. (2022) to determine the optimum number of factors. A total of 5 OA factors were identified: hydrocarbon-like OA (HOA), cooking-like OA (COA), biomass burning OA (BBOA), more-oxidized OOA (MO-OOA) and less-oxidized OOA (LO-OOA). Adding an additional factor resulted in splitsplitting of COA factor, decreasing it to four factors caused mixing between the MO-OOA and COA factors. Therefore, 5 factor-solution was determined across the whole year. In addition, site-specific factor profiles were derived for HOA, COA, and BBOA through a seasonal bootstrap PMF analysis for winter (Dec, Jan, and Feb) and used as constraints as suggested in Chen et al. (2022) and Via et al. (2022). However, the MY site is surrounded by many restaurants with prevalent cooking emissions. Thus, the chemical fingerprint for both HOA and COA might not be fully separated. Therefore, we constrained the trend of NO_x time series, BBOA and COA profiles from a previous winter bootstrap solution collected in London North Kensington (2015-2018, Chen et al., 2022) to retrieve environmentally reasonable results with five factors in winter data, so-called base case solution. Then, a bootstrap resampling analysis with 100 iterations and five factors was conducted by constraining the factor profiles of HOA, COA, and BBOA from the base case with random a-value from 0.1-0.5 with step of 0.1. It results in stable factor profiles of these three primary sources as shown in Fig. S2, which shows good agreements with published reference profiles (Chen et al., 2022; Crippa et al., 2013).

Rolling PMF was conducted with a time window of 14 days and a step of 1 day by constraining primary factor profiles of HOA, COA, BBOA in Fig. S2 (averaged bootstrap results) and two additional unconstrained factors with bootstrap resampling and the random a-value option (0.1-0.5, step of 0.1, 50 iterations/window). A criteria list including selections based on both time series and factor profiles as shown in Table S1 was applied as per Chen et al. (2022). With the help of *t*-test in temporal-based criteria (1-3), we can minimize subjective judgements in determining the environmentally reasonable results. Eventually, 3,166 runs (14.1%) of the PMF runs were selected across different rolling windows across the whole year to average as the final results (utilized a-values were averaged to two decimal places) with 4.9 % unmodelled data points, which is comparable with other rolling PMF analyses (Chen et al., 2022).

2.5 Meteorological normalisation using boot regression tree model

Meteorological normalisation, also known as deweathering analysis, has been conducted using the "worldmet" R package (Carslaw, 2025) to build boot regression tree (BRT) models for all resolved OA factors from PMF as well as chemical species measured. Considered variables, included relative humidity, wind speed, wind direction, and air temperature trend, hours of the day (local time), day of the week, Julian dates, week of the year as suggested by (Carslaw, (2025). While Grange and Carslaw (2019) have also suggested boundary layer height, air mass cluster, or back trajectory information would be beneficial to include to deal with pollutants primarily controlled by regional scale process. However, the aim of this study is understanding how COVID-related policies affect primary/local emission sources (i.e., BC, HOA, COA, and BBOA), which will not be affected significantly regional process, therefore additional metrics will most likely not improve the quantification of these PM components. It is consistent and comparable with previous studies (Font et al., 2022; Grange et al., 2021; Yao and Zhang, 2024) that includes similar meteorological

parameters (i.e., wind speed, wind direction, relative humidity, temperature). In addition, since the trained BRT models are sufficiently good even without considering boundary layer height and back trajectories, performing random forest model will not improve the model significantly, nor change the results drastically as suggested by Yao and Zhang (2024). Thus, in this study, only BRT models were trained and the meteorological effects subsequently removed (i.e., relative humidity, wind speed, wind direction, and air temperature) on all PM/OA.

3 Results and Discussions

242 <u>3.1 Model performance of meteorological normalisation</u>

The performance of each model (for individual species/source) is shown in Table S2 with slopes from 0.97 to 1.02 and R² (Pearson) from 0.77 to 0.94, which have similar or somewhat better performances compared with previous studies (Font et al., 2022; Grange et al., 2018, 2021; Grange and Carslaw, 2019; Krechmer et al., 2018; Shi et al., 2021; Yao and Zhang, 2024). As shown in the SI (Fig. S8 and Fig. S9) and the lower panel of Figure 5, Yao and Zhang (2024) meteorological effects were generally considerable, especially for Pre-lockdown Spring period, while it does not change the conclusion of the effects from lockdown and EOTHO policies. Therefore, the main results presented in this study are based on the original measurements.

3.13.2 Chemical composition of submicron PM for different periods around the COVID-19 Lockdown

The average PM_1 mass concentration at MY site was $11 \mu g/m^3$ for the study period with 44% OA, 21% NO₃, 15% SO₄, 16% BC, 5% NH₄, and 0.6% Cl. The distribution of the chemical composition on PM_1 varied depending on the season and variation was associated with the lockdown and

EOTHO policies (Figure 1). PM₁ increased by 95% in lockdown spring (Mar 26th–May 256 31st, 2020) compared to pre-lockdown spring (Mar 1st-Mar 25th, 2020). Specifically, Org, SO₄, 257 NO₃, NH₄, and Cl all increased by 87%, 211%, 73%, 237%, and 132%, respectively. Except for 258 259 BC, which decreased by 52%. This is due to the polluted airmass originating from mainland 260 Europe and the enhanced agricultural emissions in spring from the UK and wider continental 261 Europe (Aksoyoglu et al., 2020). It was further confirmed, through back trajectory analysis, that 262 elevated PM₁ events (Mar 25th–Mar 28th, Apr 8th–Apr 10th, and Apr 15th–Apr 17th), where the result 263 of airmasses passing over northern continental Europe (Fig. S6). In addition, the Org, SO₄, NO₃, NH₄, and Cl were only increased by 21%, 107%, 50%, and 28% respectively after the 264 deweathering analysis (Fig. S8), suggesting significant meteorological influences during this 265 266 period. NO₃ concentration reduced in summer 2019 and 2020 as expected compared to spring or 267 fall seasons due to the volatility semi-volatile nature of NH₄NO₃ and lower agricultural emissions, 268 while SO₄ concentrations increased in summer due to enhanced photochemistry across Europe 269 (Bressi et al., 2021; Chen et al., 2022). During the lockdown in spring, SO₄ concentrations 270 remained high, which was associated with long-range transport and marine aerosols (e.g., 271 methanesulfonic acid, MSA) (Fig. S7).

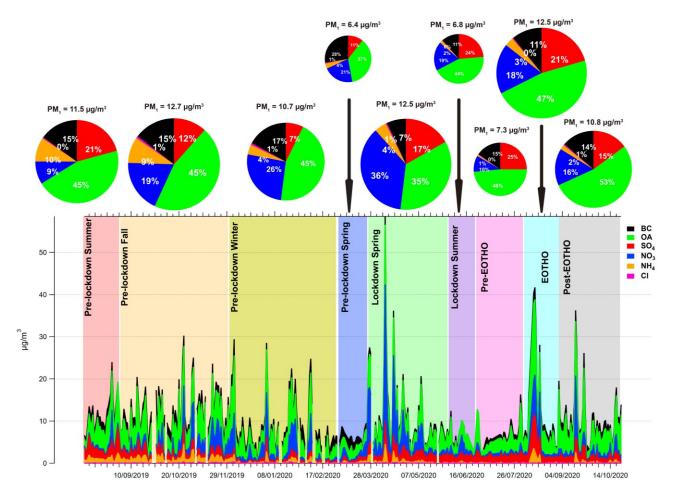


Figure 1 Chemical compositions of PM₁ at MY from Aug 2019 to Oct 2020 (daily resolution) and averaged for the different periods as shown in <u>Table 1</u>.

BC concentrations during the spring lockdown (Mar 26th–May 31st 2020) reduced from 1.78 to 0.86 μg/m³ (-52%) compared to the pre-lockdown level in spring (Mar 1st–Mar 25th), due to the significant reduction in traffic during the first lockdown (Transport for London, 2020). Similar decreasing of BC has been observed elsewhere during COVID lockdown as described in introduction (Gamelas et al., 2023; Jeong et al., 2022; Petit et al., 2021; Tian et al., 2021; Xu et al., 2020). It is worth noting that the BC concentration had already reduced by 3% in pre-lockdown spring (Mar 1st–Mar 25th) compared to the pre-lockdown winter. This is likely due to vehicle mileage reducing as the UK government implemented travel restrictions and advised people to work from home on Mar 16th, 2020 (Transport for London, 2020). BC increased to 1.13 μg/m³

(+44%) after the lockdown and before the EOTHO (Jun 24th–Aug 2nd,2020, pre-EOTHO in Figure 1Figure 1) as people returned to work and travel. However, BC concentrations remained 34% lower than the pre-lockdown summer (Aug 1st–Aug 31st, 2019) concentration of 1.72 μg/m³ (Fig. S9), which suggests that the traffic emissions reduced considerably as the fewer economic activities even after the ease of the first lockdown (e.g., suggestions of hybrid working mode, restricted international travel, reduced tourism, limited access to entertainments). BC also increased to 1.35 μg/m³ (+19%) during the EOTHO scheme (Aug 3rd–Aug 31st, 2020). This was not only because of increased traffic emission during this period, but may also result from cooking activities (e,g, barbecuing or wood-fired cooking styles) in Central London (Defra, 2023). Since the EOTHO was only in place from Mon to Wed, BC concentrations (likely due to increased traffic and cooking emissions) increased on Mon-Wed compared to post-lockdown but before EOTHO (Jun 24th–Aug 2nd, 2020) (Fig. S10).

3.23.3 OA sources in Central London

As mentioned above, the rolling PMF analysis resolved 5 factor solutions, including HOA, COA, BBOA, MO-OOA, and LO-OOA as shown in Figure 3Figure 3 and Figure 4Figure 4. The left panel of Figure 3Figure 3 shows the yearly averaged factor profiles of resolved PMF factors and total OOA calculated as the sum of LO-OOA and MO-OOA. All factors show good agreements with previous studies in terms of key m/z tracers. In addition, as shown in Figure 2Figure 2, the contribution to total OA concentrations from HOA, BBOA, and LO-OOA was consistent at different OA concentrations. However, the contribution of COA increased as total OA concentrations increased. This suggests that cooking emissions in Central London are responsible for elevated OA concentrations, which was also the case in Athens as shown in Chen et al. (2022).

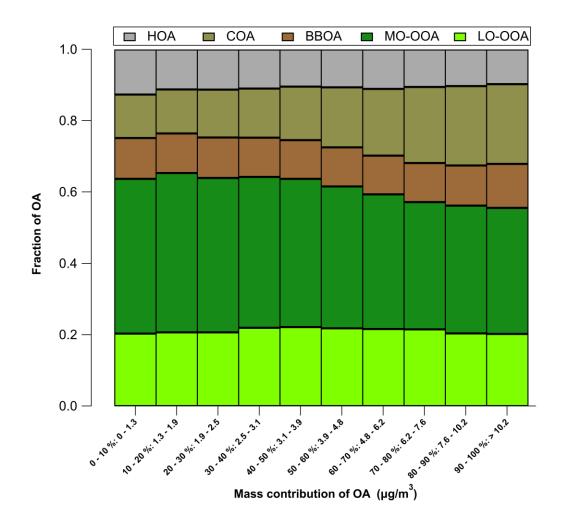


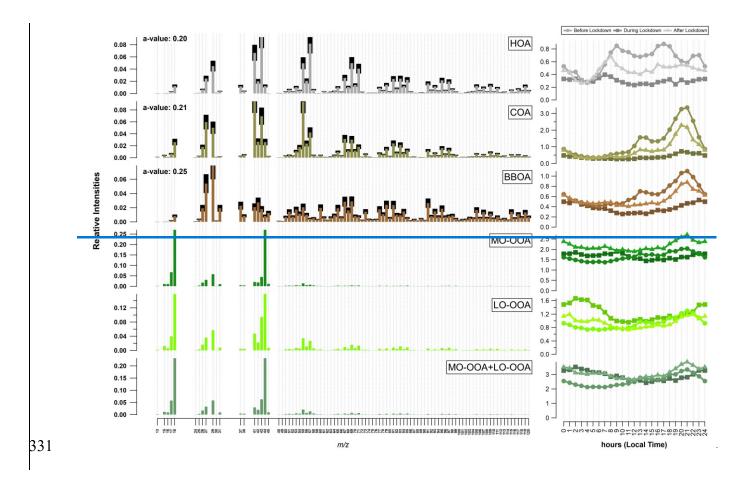
Figure 2 Contributions to total OA from the different identified OA sources at different OA concentrations. Total OA concentrations were split in 10 equally distributed bins.

3.2.13.3.1 General characteristics of OA factors

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The right panel of Figure 3Figure 3 shows time series (daily averaged) and Figure 4Figure 4 shows diurnal cycles for each OA factor. The mean concentrations of HOA, COA, BBOA, MO-OOA, LO-OOA, and OOA (MO-OOA+LO-OOA) were $0.50 \pm 0.1~\mu g/m^3$, $0.93 \pm 0.14~\mu g/m^3$, $0.55 \pm 0.11~\mu g/m^3$, $1.81 \pm 0.41~\mu g/m^3$, $1.00 \pm 0.44~\mu g/m^3$, and $2.80 \pm 0.70~\mu g/m^3$, aonnd contributed to OA (PM₁) with the fractions of 11% (5% to PM₁), 20% (9% to PM₁), 12% (5% to PM₁), 38% (17% to PM₁), 21% (9% to PM₁), and 59% (26% to PM₁), respectively. The concentration of all OA factors shows strong time variations over the year as shown on the Figure 4Figure 4. OA factors also

showed considerable seasonality besides the effects from COVID-related policies (Figure 4Figure 4 and Fig. S3). POA concentrations were generally lower in the warmer seasons than in winter as lower temperature favours particle formation via condensation and dilution and dispersion are reduced due to the lower boundary layer. It's worth mentioning that the reduced POA concentrations in the warm season was not caused by reduced residential heating and energy consummation since Central London mainly uses natural gas and renewable energy instead effather than solid fuel combustions (Cliff et al., 2025). The OOA factor concentrations remain relatively consistent across seasons, while its contributions were was larger during the warmer seasons (Fig. S4). This is because both high temperature and strong irradiation solar radiation will enhance the photochemistry and evaporation of POA sources, and the increased biogenic volatile organic compound (VOC) emissions lead to high OOA production despite the evaporation of semi-volatile OOA (Fig. S4) (Chen et al., 2022)). The temporal variations observed here in central Central London was consistent with the other urban sites across Europe (Chen et al., 2022). Therefore, this study focuses on the impacts of COVID-related polices on OA sources.



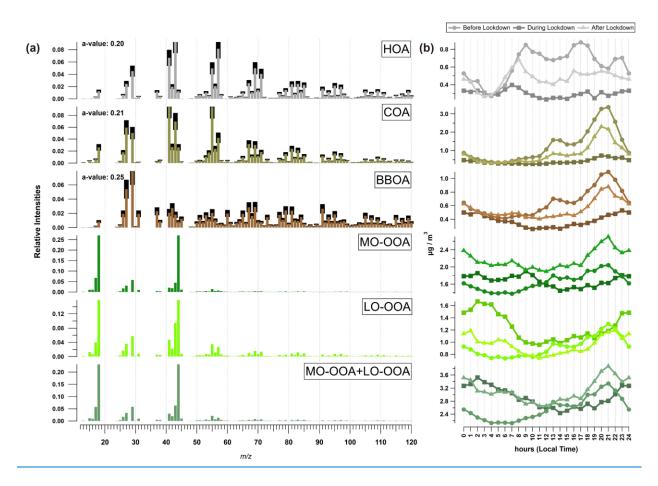


Figure 3 Yearly averaged profiles (lefta) and diurnal cycles (rightb) of resolved factors from the rolling PMF analysis at the MY site. Time is expressed in local time.

The right side of Figure 3 Figure 3 shows the diurnal cycles before, during, and after the lockdown. POA factors showed distinct diurnal variations, in which HOA showed morning and evening rush hour peaks, COA showed distinct lunchtime and evening peaks, and BBOA showed a similar pattern as COA before and after the lockdown. This indicates that the part of what is measured as BBOA in central Central London is most likely co-emitted from cooking activity, most likely from barbecuing style or wood-oven pizza restaurants in the area. A survey about use of domestic fuels in the hospitality sector was conducted by Department for Environment Food and Rural Affairs (Defra), UK suggested that restaurants use solid fuel to cook to provide unique flavours (Defra, 2023). Mohr et al. (2009) showed that meat-cooking can slightly elevate m/z 60, which is an important ion in the BBOA factor profile. OOA factors showed much less diurnal variation

compared to POA factors in all periods, this is in agreement with the other 22 European sites reported in Chen et al. (2022). The MO-OOA showed a smaller diurnal variation compared to LO-OOA. This is because the LO-OOA is also known as semi-volatile OOA (SV-OOA), which evaporate during the day due to higher temperatures and accumulate in the evening due to the shallower boundary layer (Chen et al., 2022).

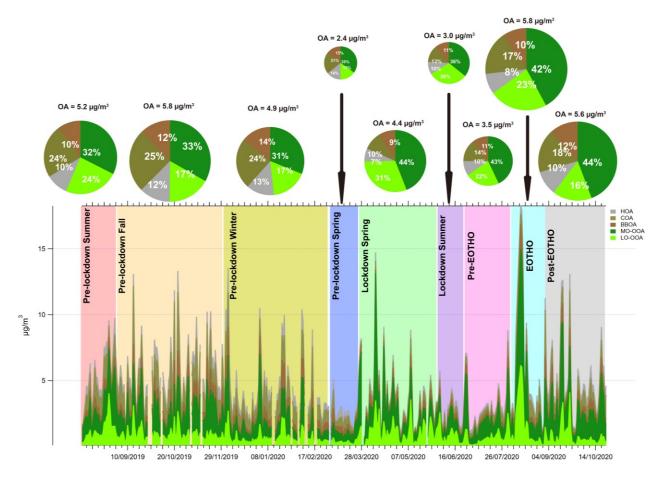


Figure 4 Average mass concentrations for OA sources at MY during different periods from Aug 2019 to Oct 2020

3.2.23.3.2 Pre-lockdown Spring Pre-lockdown Spring

In general, OA concentration decreased by 51% in pre-lockdown spring compared to pre-lockdown winter (Dec 1st, 2019–Feb 28th, 2020) due to seasonality, origins of airmass (Fig. S5), and the impact of lockdown. OOA concentrations also decreased drastically by 50%. Primary emissions were significantly lower due to reduced vehicle mileage and other economic activity

before the official lockdown measure came into force on March 26th 2020 (Figure 3 (a) and Figure 4Figure 4) as suggested by the 1st quarter drop in GDP (ONS, 2022). Atmospheric components related to vehicle emissions (HOA and BC) decreased by 48% and 3% respectively, in early March 2020. COA decreased by 58% due to fewer restaurant activity, BBOA decreased by 50% was likely due to the reduced commercial cooking using charcoal and wood as well as warmer weather requiring less domestic space heating.

3.2.33.3.3 Lockdown

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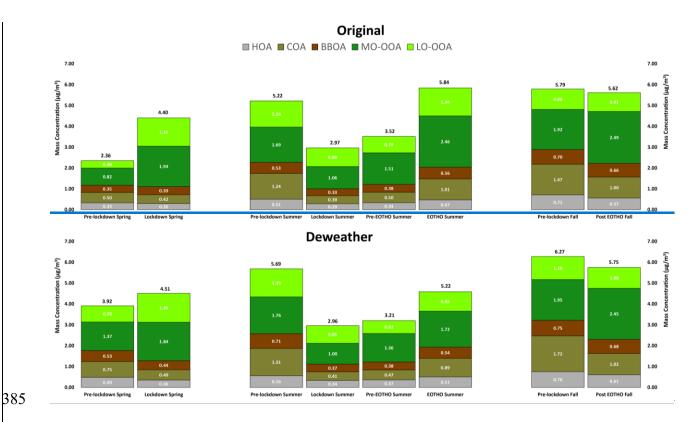
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The diurnal variation of COA and BBOA during lockdown showed much less intensity overall but the distinctive lunchtime peak remained as the pre-lockdown; and the evening peak reduced its intensity (Figure 3 Figure 3 b). HOA retained distinct morning and evening rush hour peaks but at lower mass concentrations during lockdown (Figure 3 Figure 3 b). This is because the takeout activities of some restaurants were still active as well as the potential increases for residential cooking activities during lockdown. After the first lockdown, the distinct lunch and evening peaks in diurnal patterns of COA and BBOA reappeared as the open-up of nearby restaurants. The morning and evening rush hour peaks for HOA enhanced considerably as the ease of the travel restrictions after the first lockdown. However, POA concentrations did not reach pre-COVID levels (Figure 5Fig. 5). This is likely due to widespread hybrid working and the remaining oversea travel restrictions supressing tourism, which reduced traffic activity and restaurants visits. Conversely, OOA concentrations during lockdown were slightly higher than pre-lockdown levels. These were related to long-range transport, with relatively high mass concentrations of MO-OOA and LO-OOA during the lockdown (Fig. S5 and Fig. S6). Compared to the pre-lockdown spring, HOA and COA in the lockdown spring decreased by 11% and 15%, respectively, while BBOA increased marginally by 13% (from 0.35 to 0.39 µg/m³) (Figure 5Figure 5). MO-OOA and LO-

OOA increased by 136% and 279%, respectively due to long-range transportation of airmasses from continental Europe (Fig. S5 and Fig. S6) and increased photochemistry (enhanced temperature and ozone levels in Fig. S4) compared to the first 25 days in Mar 2020. This was

- 383 accompanied by increased SO₄ (+211%), NH₄ (+132%) and NO₃ (+237%) as shown in Fig. S9,
- despite the higher temperature could favour partitioning these species into the gas phase.



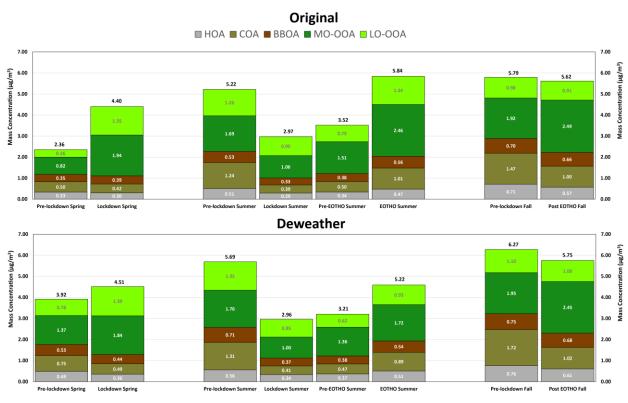


Figure 5 The impacts on OA sources during different periods compared with business-as-usual cases with and without deweathering analysis.

 In June 2020, still in lockdown (Jun 1st Jun 23rd, 2020), POA showed further but marginal decreases (-3%, -8%, and -15% for HOA, COA, and BBOA, respectively, Figure 4Figure 4) compared to the lockdown spring as the enhanced photochemistry leads to increased formation of OOA from the POA. However, the overall mass concentration of MO-OOA and LO-OOA decreased significantly by 45%, and 34%, respectively as the result of fewer long-range transported airmasses (Fig. S5).

3.2.43.3.4 Pre-EOTHO

During pre-EOTHO (Jun 24th–Aug 2nd, 2020) after the lockdown policy was eased, HOA, COA, and BBOA all showed considerably increases of 16%, 30%, and 14%, respectively when compared to lockdown summer period. In which, MO-OOA increased by 45% and LO-OOA decreased by 13%, respectively as the relatively higher temperature and irradiations—solar radiation were favouring—favoured the vaporization—evaporation of LO-OOA and production of MO-OOA from LO-OOA and POA. As shown in Figure 5Figure 5, the POA concentrations were much lower when compared to summer 2019 (Aug 1st–Aug 31st, 2019) as travel and economic activities did not return to pre-COVID levels (ONS, 2022; Transport for London, 2020). Specifically, reduced vehicle mileage resulted in lower HOA (-33%), BC (-37%), %) due to reduced vehicle mileage resulted, and COA (-59%) due to the reduced commercial cooking activity. As BBOA is coemitted with COA during of cooking activities, BBOA also decreased slightly from 0.53 to 0.38 μg/m³ (-28%, Figure 5Figure 5).

408 3.2.53.3.5 Eat Out To Help Out (EOTHO)

EOTHO policy (Aug 3rd–Aug 31st, 2020) had a significant impact on all POA factors. In particular, the COA concentration increased by 100% compared to the post-lockdown period (Pre-EOTHO

Summer) from Jun 24th to Aug 2nd, 2020 (0.5 to 1.0 μg/m³, Figure 6Figure 6). HOA and BBOA concentrations also increased by 40% and 48%, respectively, which suggested the human activities resulting in these emissions recovered slowly after the lockdown (ONS, 2022; Transport for London, 2020). COA was significantly higher due to EOTHO, however, it did not reach pre-COVID concentrations (Figure 5Figure 5 and Figure 6Figure 6) as its level was lower on each weekday except for Mon.

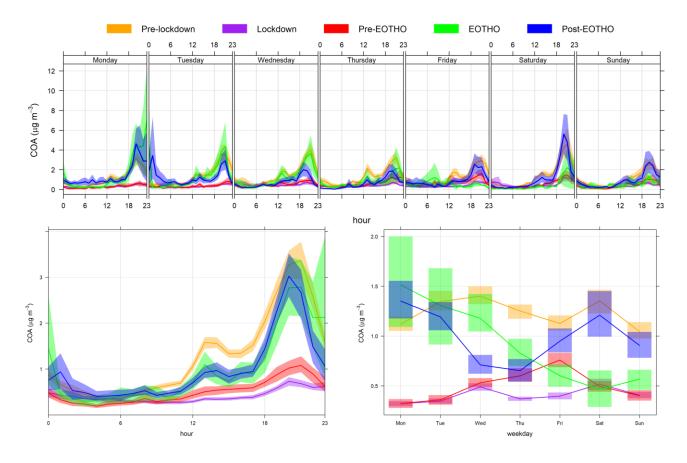


Figure 6 . COA diurnal plots for each weekday, diurnal plots, and weekly plots at different periods in relation with COVIDrelated policies

EOTHO only operated from Mon to Wed, and this was clear in the diurnal plots (Figure 6Figure 6 and Fig. S12) with larger COA concentrations from Mon to Wed, in contrast with larger concentrations over the weekend (Fri to Sun) before EOTHO (Jun 24th–Aug 2nd, 2020). Interestingly, even after the EOTHO policy ceased (Sep 1st–Oct 22nd, 2020), COA levels remained

elevated on Mon and Tue but a much higher level during the weekend was observed. This suggests that EOTHO had an influence on the consumer behaviour even after the lockdown. It is also worth noting that the high concentrations of COA and BBOA (Figure 6Figure 6 and Fig. S11) on Monday night were caused by the last day of EOTHO policy coinciding with a UK public holiday on Aug 31st.

Also, during EOTHO, MO-OOA and LO-OOA increased by 63% and 70% respectively compared to post-lockdown concentrations before EOTHO and correlated with increased NH₄, NO₃ and SO₄ concentrations. This was mainly due to long-ranged transported airmasses (Fig. S5) and enhanced photochemistry with increased temperature and mass concentration of POAs (Fig. S4).

4 Conclusion

This study demonstrates the importance of source apportionment studies to better understand how national and local government policies can impact the PM mixture, and how these effects can be differentiated from the influences of meteorology and large-scale atmospheric processes. PM concentrations increased at the beginning of the lockdown (Mar–Apr 2020) despite reduced economic activities, which was caused by long-range transported airmasses instead of primary emissions. Through examining the source apportionment (and inorganic PM composition), COVID-related policies were found to have profound but largely unintended impacts on air quality. The first lockdown significantly reduced POA sources: including HOA by 52%, COA by 67%, and BBOA by 42%. While all these components reduced dramatically during the lockdown, they only gradually increased again and did not reach pre-COVID levels during the duration of this study (Aug 2019– Oct 2020).

Most significantly, while the Eat Out To Help Out (EOTHO) policy was effective in helping the hospitality industry to recover from economic losses during the lockdown, it had unintended impacts on air quality as cooking emissions increased. Clearly detecting this change confirms the presence of COA (20% to OA) as an important source of OA in London and the importance of commercial cooking as a source. Also of note was the impact that EOTHO had on BBOA concentrations, which increased by 48% while this policy was in place. This establishes a clear link between commercial cooking activity and BBOA measured in cities due to the use of charcoal and wood as cooking fuels (Defra, 2023), as well as potentially emissions from cooking ingredients. Cooking may therefore be underestimated as a source if COA concentrations are considered in isolation, and BBOA is only associated with other sources of solid fuel burning. This emphasises the need to develop policies and technical solutions to mitigate commercial cooking emissions in the urban environment, especially as there are limited regulations on this industry in terms of air pollution. There are filter technologies (e.g., electrostatic precipitators, UV-C lamp exhaust hood, hydrovents) available that have been implemented as law in Hong Kong to effectively control cooking emissions (Hong Kong EPD, 2024). It also demonstrated the importance in continuous monitoring with subsequent source apportionment analysis to better understand the influence of government policies to improve air quality more effectively.

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Code/Data availability

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Rolling PMF analyses is run using SoFi Pro from Datalystica (https://datalystica.com/sofi-pro/,
Datalystica, 2024) under Igor Pro 9 platform from WaveMetrics® (https://www.wavemetrics.com/,
WaveMetrics, 2024) and they are both available for purchase. Raw data/results from the study are
available upon request to the corresponding author Gang I. Chen (gang.chen@imperial.ac.uk).

Author contribution

Gang I. Chen: Writing – review & editing, Writing – original draft, Visualization, Validation,
Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data
curation, Conceptualization. Anja H. Tremper: Writing – review & editing, Methodology,
Formal analysis, Data curation. Max Priestman: Methodology, Formal analysis, Data curation.
Anna Font: Writing – review & editing, Methodology, Formal analysis, Data curation. David C.
Green: Writing – review & editing, Supervision, Project administration, Methodology, Resources,
Funding acquisition, Conceptualization.

Competing interests

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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References

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487 Aksoyoglu, S., Jiang, J., Ciarelli, G., Baltensperger, U., and Prévôt, A. S. H.: Role of ammonia 488 in European air quality with changing land and ship emissions between 1990 and 2030, 489 Atmos Chem Phys, 20, 15665–15680, https://doi.org/10.5194/acp-20-15665-2020, 490 2020. 491 Allan, J. D., Delia, A. E., Coe, H., Bower, K. N., Alfarra, M. R., Jimenez, J. L., Middlebrook, 492 A. M., Drewnick, F., Onasch, T. B., Canagaratna, M. R., Jayne, J. T., and Worsnop, D. 493 R.: A generalised method for the extraction of chemically resolved mass spectra from 494 Aerodyne aerosol mass spectrometer data, J Aerosol Sci, 35, 909–922, 495 https://doi.org/10.1016/j.jaerosci.2004.02.007, 2004. 496 Bressi, M., Cavalli, F., Putaud, J. P., Fröhlich, R., Petit, J. E., Aas, W., Äijälä, M., Alastuey, A., 497 Allan, J. D., Aurela, M., Berico, M., Bougiatioti, A., Bukowiecki, N., Canonaco, F., 498 Crenn, V., Dusanter, S., Ehn, M., Elsasser, M., Flentje, H., Graf, P., Green, D. C., 499 Heikkinen, L., Hermann, H., Holzinger, R., Hueglin, C., Keernik, H., Kiendler-Scharr, 500 A., Kubelová, L., Lunder, C., Maasikmets, M., Makeš, O., Malaguti, A., Mihalopoulos, 501 N., Nicolas, J. B., O'Dowd, C., Ovadnevaite, J., Petralia, E., Poulain, L., Priestman, M., Riffault, V., Ripoll, A., Schlag, P., Schwarz, J., Sciare, J., Slowik, J., Sosedova, Y., 502 503 Stavroulas, I., Teinemaa, E., Via, M., Vodička, P., Williams, P. I., Wiedensohler, A., 504 Young, D. E., Zhang, S., Favez, O., Minguillón, M. C., and Prevot, A. S. H.: A European

505	aerosol phenomenology - 7: High-time resolution chemical characteristics of submicron
506	particulate matter across Europe, Atmos Environ X, 10, 100108,
507	https://doi.org/10.1016/J.AEAOA.2021.100108, 2021.
508	Canonaco, F., Tobler, A., Chen, G., Sosedova, Y., Slowik, J. G., Bozzetti, C., Daellenbach, K.
509	R., El Haddad, I., Crippa, M., Huang, RJ., Furger, M., Baltensperger, U., and Prévôt,
510	A. S. H.: A new method for long-term source apportionment with time-dependent factor
511	profiles and uncertainty assessment using SoFi Pro: application to 1 year of organic
512	aerosol data, Atmos Meas Tech, 14, 923-943, https://doi.org/10.5194/amt-14-923-2021
513	2021.
514	Carslaw, D.: deweather: Remove the influence of weather on air quality data, https://openair-
515	project.github.io/deweather/, 2025.
516	Chebaicheb, H., de Brito, J. F., Amodeo, T., Couvidat, F., Petit, JE., Tison, E., Abbou, G.,
517	Baudic, A., Chatain, M., Chazeau, B., Marchand, N., Falhun, R., Francony, F., Ratier,
518	C., Grenier, D., Vidaud, R., Zhang, S., Gille, G., Meunier, L., Marchand, C., Riffault,
519	V., and Favez, O.: Multiyear high-temporal-resolution measurements of submicron
520	aerosols at 13 French urban sites: data processing and chemical composition, Earth Syst
521	Sci Data, 16, 5089-5109, https://doi.org/10.5194/essd-16-5089-2024, 2024.
522	Chen, G., Canonaco, F., Tobler, A., Aas, W., Alastuey, A., Allan, J., Atabakhsh, S., Aurela, M.,
523	Baltensperger, U., Bougiatioti, A., De Brito, J. F., Ceburnis, D., Chazeau, B.,
524	Chebaicheb, H., Daellenbach, K. R., Ehn, M., El Haddad, I., Eleftheriadis, K., Favez,
525	O., Flentje, H., Font, A., Fossum, K., Freney, E., Gini, M., Green, D. C., Heikkinen, L.,
526	Herrmann, H., Kalogridis, AC., Keernik, H., Lhotka, R., Lin, C., Lunder, C.,
527	Maasikmets, M., Manousakas, M. I., Marchand, N., Marin, C., Marmureanu, L.,

528	Mihalopoulos, N., Močnik, G., Nęcki, J., O'Dowd, C., Ovadnevaite, J., Peter, T., Petit					
529	JE., Pikridas, M., Matthew Platt, S., Pokorná, P., Poulain, L., Priestman, M., Riffault					
530	V., Rinaldi, M., Różański, K., Schwarz, J., Sciare, J., Simon, L., Skiba, A., Slowik, J.					
531	G., Sosedova, Y., Stavroulas, I., Styszko, K., Teinemaa, E., Timonen, H., Tremper, A.					
532	Vasilescu, J., Via, M., Vodička, P., Wiedensohler, A., Zografou, O., Cruz Minguillón,					
533	M., and Prévôt, A. S. H.: European aerosol phenomenology - 8: Harmonised source					
534	apportionment of organic aerosol using 22 Year-long ACSM/AMS datasets, Environ					
535	Int, 166, 107325, https://doi.org/10.1016/j.envint.2022.107325, 2022.					
536	Cliff, S. J., Drysdale, W., Lewis, A. C., Møller, S. J., Helfter, C., Metzger, S., Liddard, R.					
537	Nemitz, E., Barlow, J. F., and Lee, J. D.: Evidence of Heating-Dominated Urban NOx					
538	Emissions, Environ Sci Technol, 59, 4399–4408					
539	https://doi.org/10.1021/acs.est.4c13276, 2025.					
540	Crippa, M., DeCarlo, P. F., Slowik, J. G., Mohr, C., Heringa, M. F., Chirico, R., Poulain, L.					
541	Freutel, F., Sciare, J., Cozic, J., Di Marco, C. F., Elsasser, M., Nicolas, J. B., Marchand					
542	N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E.					
543	Zimmermann, R., Jaffrezo, JL., Prévôt, A. S. H., and Baltensperger, U.: Wintertime					
544	aerosol chemical composition and source apportionment of the organic fraction in the					
545	metropolitan area of Paris, Atmos Chem Phys, 13, 961-981					
546	https://doi.org/10.5194/acp-13-961-2013, 2013.					
547	Defra: Use of domestic fuels in the hospitality sector – a qualitative review of hospitality					
548	businesses, 2023.					
549	Dominici, F., Peng, R. D., Bell, M. L., Pham, L., McDermott, A., Zeger, S. L., and Samet, J					
550	M.: Fine Particulate Air Pollution and Hospital Admission for Cardiovascular and					

551	Respiratory Diseases, JAMA, 295, 1127, https://doi.org/10.1001/jama.295.10.1127,
552	2006.
553	European Environment Agency: Europe's air quality status 2024, https://doi.org/10.2800/5970,
554	2024.
555	European Union: Directive (EU) 2024/2881 of the European Parliament and of the Council of
556	23 October 2024 on ambient air quality and cleaner air for Europe (recast), 2024.
557	Font, A., Ciupek, K., Butterfield, D., and Fuller, G. W.: Long-term trends in particulate matter
558	from wood burning in the United Kingdom: Dependence on weather and social factors,
559	Environmental Pollution, 314, 120105, https://doi.org/10.1016/j.envpol.2022.120105,
560	2022.
561	Fröhlich, R., Cubison, M. J., Slowik, J. G., Bukowiecki, N., Prévôt, A. S. H., Baltensperger, U.,
562	Schneider, J., Kimmel, J. R., Gonin, M., Rohner, U., Worsnop, D. R., and Jayne, J. T.:
563	The ToF-ACSM: a portable aerosol chemical speciation monitor with TOFMS detection
564	Atmos Meas Tech, 6, 3225–3241, https://doi.org/10.5194/amt-6-3225-2013, 2013.
565	Gamelas, C. A., Canha, N., Vicente, A., Silva, A., Borges, S., Alves, C., Kertesz, Z., and
566	Almeida, S. M.: Source apportionment of PM2.5 before and after COVID-19 lockdown
567	in an urban-industrial area of the Lisbon metropolitan area, Portugal, Urban Clim, 49,
568	101446, https://doi.org/10.1016/j.uclim.2023.101446, 2023.
569	Grange, S. K. and Carslaw, D. C.: Using meteorological normalisation to detect interventions
570	in air quality time series, Science of The Total Environment, 653, 578-588,
571	https://doi.org/10.1016/j.scitotenv.2018.10.344, 2019.

572 Grange, S. K., Carslaw, D. C., Lewis, A. C., Boleti, E., and Hueglin, C.: Random forest 573 meteorological normalisation models for Swiss PM 10 trend analysis, Atmos Chem Phys, 574 18, 6223–6239, https://doi.org/10.5194/acp-18-6223-2018, 2018. 575 Grange, S. K., Lee, J. D., Drysdale, W. S., Lewis, A. C., Hueglin, C., Emmenegger, L., and 576 Carslaw, D. C.: COVID-19 lockdowns highlight a risk of increasing ozone pollution in 577 European urban areas, Atmos Chem Phys, 21, 4169–4185, https://doi.org/10.5194/acp-578 21-4169-2021, 2021. 579 Hass-Mitchell, T., Joo, T., Rogers, M., Nault, B. A., Soong, C., Tran, M., Seo, M., Machesky, 580 J. E., Canagaratna, M., Roscioli, J., Claflin, M. S., Lerner, B. M., Blomdahl, D. C., 581 Misztal, P. K., Ng, N. L., Dillner, A. M., Bahreini, R., Russell, A., Krechmer, J. E., 582 Lambe, A., and Gentner, D. R.: Increasing Contributions of Temperature-Dependent 583 Oxygenated Organic Aerosol to Summertime Particulate Matter in New York City, ACS 584 ES&T Air, 1, 113–128, https://doi.org/10.1021/acsestair.3c00037, 2024. 585 Hong Kong EPD: Control of Oily Fume and Cooking Odour from Restaurants and Food 586 Business, 2024. 587 Hu, R., Wang, S., Zheng, H., Zhao, B., Liang, C., Chang, X., Jiang, Y., Yin, R., Jiang, J., and 588 Hao, J.: Variations and Sources of Organic Aerosol in Winter Beijing under Markedly 589 Reduced Anthropogenic Activities During COVID-2019, Environ Sci Technol, 56, 590 6956–6967, https://doi.org/10.1021/acs.est.1c05125, 2022. 591 IPCC: Climate Change 2021 – The Physical Science Basis, Cambridge University Press, 592 https://doi.org/10.1017/9781009157896, 2021. 593 Jayne, J. T., Leard, D. C., Zhang, X., Davidovits, P., Smith, K. A., Kolb, C. E., and Worsnop, 594 D. R.: Development of an Aerosol Mass Spectrometer for Size and Composition

595 Analysis of Submicron Particles, Aerosol Science and Technology, 33, 49-70, 596 https://doi.org/10.1080/027868200410840, 2000. 597 Jeong, C.-H., Yousif, M., and Evans, G. J.: Impact of the COVID-19 lockdown on the chemical 598 composition and sources of urban PM2.5, Environmental Pollution, 292, 118417, 599 https://doi.org/10.1016/j.envpol.2021.118417, 2022. 600 Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H. H., Zhang, Q., Kroll, J. 601 H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. 602 603 R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., 604 Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., 605 Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. 606 I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., 607 Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., 608 609 Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., 610 Middlebrook, A. M., Kolb, C. E., Baltensperger, U., Worsnop, D. R., Dunlea, E. J., 611 Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., 612 Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., 613 Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. 614 Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., 615 Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of Organic Aerosols in the 616

617	Atmosphere, Science (1979), 326, 1525–1529, https://doi.org/10.1126/science.1180353,
618	2009.
619	Joo, T., Rogers, M. J., Soong, C., Hass-Mitchell, T., Heo, S., Bell, M. L., Ng, N. L., and Gentner,
620	D. R.: Aged and Obscured Wildfire Smoke Associated with Downwind Health Risks,
621	Environ Sci Technol Lett, 11, 1340-1347, https://doi.org/10.1021/acs.estlett.4c00785,
622	2024.
623	Kelly, F. J. and Fussell, J. C.: Size, source and chemical composition as determinants of toxicity
624	attributable to ambient particulate matter, Atmos Environ, 60, 504-526,
625	https://doi.org/10.1016/j.atmosenv.2012.06.039, 2012.
626	Krechmer, J., Lopez-Hilfiker, F., Koss, A., Hutterli, M., Stoermer, C., Deming, B., Kimmel, J.,
627	Warneke, C., Holzinger, R., Jayne, J., Worsnop, D., Fuhrer, K., Gonin, M., and de Gouw,
628	J.: Evaluation of a New Reagent-Ion Source and Focusing Ion-Molecule Reactor for
629	Use in Proton-Transfer-Reaction Mass Spectrometry, Anal Chem, 90, 12011-12018,
630	https://doi.org/10.1021/acs.analchem.8b02641, 2018.
631	Laj, P., Lund Myhre, C., Riffault, V., Amiridis, V., Fuchs, H., Eleftheriadis, K., Petäjä, T.,
632	Salameh, T., Kivekäs, N., Juurola, E., Saponaro, G., Philippin, S., Cornacchia, C.,
633	Alados Arboledas, L., Baars, H., Claude, A., De Mazière, M., Dils, B., Dufresne, M.,
634	Evangeliou, N., Favez, O., Fiebig, M., Haeffelin, M., Herrmann, H., Höhler, K., Illmann,
635	N., Kreuter, A., Ludewig, E., Marinou, E., Möhler, O., Mona, L., Eder Murberg, L.,
636	Nicolae, D., Novelli, A., O'Connor, E., Ohneiser, K., Petracca Altieri, R. M., Picquet-
637	Varrault, B., van Pinxteren, D., Pospichal, B., Putaud, JP., Reimann, S., Siomos, N.,
638	Stachlewska, I., Tillmann, R., Voudouri, K. A., Wandinger, U., Wiedensohler, A.,
639	Apituley, A., Comerón, A., Gysel-Beer, M., Mihalopoulos, N., Nikolova, N., Pietruczuk,

640	A., Sauvage, S., Sciare, J., Skov, H., Svendby, T., Swietlicki, E., Tonev, D., Vaughan,
641	G., Zdimal, V., Baltensperger, U., Doussin, JF., Kulmala, M., Pappalardo, G., Sorvari
642	Sundet, S., and Vana, M.: Aerosol, Clouds and Trace Gases Research Infrastructure
643	(ACTRIS): The European Research Infrastructure Supporting Atmospheric Science,
644	Bull Am Meteorol Soc, 105, E1098-E1136, https://doi.org/10.1175/BAMS-D-23-
645	0064.1, 2024.
646	Lippmann, M., Chen, L. C., Gordon, T., Ito, K., and Thurston, G. D.: National Particle
647	Component Toxicity (NPACT) Initiative: Integrated Epidemiologic and Toxicologic
648	Studies of the Health Effects of Particulate Matter Components, Boston, 2013.
649	Liu, F., Joo, T., Ditto, J. C., Saavedra, M. G., Takeuchi, M., Boris, A. J., Yang, Y., Weber, R.
650	J., Dillner, A. M., Gentner, D. R., and Ng, N. L.: Oxidized and Unsaturated: Key
651	Organic Aerosol Traits Associated with Cellular Reactive Oxygen Species Production
652	in the Southeastern United States, Environ Sci Technol, 57, 14150-14161,
653	https://doi.org/10.1021/acs.est.3c03641, 2023.
654	Mebrahtu, T. F., McEachan, R. R. C., Yang, T. C., Crossley, K., Rashid, R., Hossain, R., Vaja,
655	I., and Bryant, M.: Differences in public's perception of air quality and acceptability of
656	a clean air zone: A mixed-methods cross sectional study, J Transp Health, 31, 101654,
657	https://doi.org/10.1016/J.JTH.2023.101654, 2023.
658	Middlebrook, A. M., Bahreini, R., Jimenez, J. L., and Canagaratna, M. R.: Evaluation of
659	Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass
660	Spectrometer using Field Data, Aerosol Science and Technology, 46, 258-271,
661	https://doi.org/10.1080/02786826.2011.620041, 2012.

062	Mohr, C., Huffman, J. A., Cubison, M. J., Aiken, A. C., Docherty, K. S., Kimmel, J. R., Ulbrich,
663	I. M., Hannigan, M., and Jimenez, J. L.: Characterization of primary organic aerosol
664	emissions from meat cooking, trash burning, and motor vehicles with high-resolution
665	aerosol mass spectrometry and comparison with ambient and chamber observations,
666	Environ Sci Technol, 43, 2443–2449
667	https://doi.org/10.1021/ES8011518/SUPPL_FILE/ES8011518_SI_001.PDF, 2009.
568	Mudway, I. S., Dundas, I., Wood, H. E., Marlin, N., Jamaludin, J. B., Bremner, S. A., Cross,
669	L., Grieve, A., Nanzer, A., Barratt, B. M., Beevers, S., Dajnak, D., Fuller, G. W., Font,
570	A., Colligan, G., Sheikh, A., Walton, R., Grigg, J., Kelly, F. J., Lee, T. H., and Griffiths,
571	C. J.: Impact of London's low emission zone on air quality and children's respiratory
572	health: a sequential annual cross-sectional study, Lancet Public Health, 4, e28-e40,
573	https://doi.org/10.1016/S2468-2667(18)30202-0, 2019.
574	Ng, N. L., Herndon, S. C., Trimborn, A., Canagaratna, M. R., Croteau, P. L., Onasch, T. B.,
575	Sueper, D., Worsnop, D. R., Zhang, Q., Sun, Y. L., and Jayne, J. T.: An Aerosol
576	Chemical Speciation Monitor (ACSM) for Routine Monitoring of the Composition and
577	Mass Concentrations of Ambient Aerosol, Aerosol Science and Technology, 45, 780-
578	794, https://doi.org/10.1080/02786826.2011.560211, 2011.
579	Oltra, C., Sala, R., López-asensio, S., Germán, S., and Boso, À.: Individual-Level Determinants
580	of the Public Acceptance of Policy Measures to Improve Urban Air Quality: The Case
581	of the Barcelona Low Emission Zone, Sustainability 2021, Vol. 13, Page 1168, 13, 1168
582	https://doi.org/10.3390/SU13031168, 2021.
583	ONS: GDP and events in history: how the COVID-19 pandemic shocked the UK economy,
584	2022.

685 Petit, J.-E., Dupont, J.-C., Favez, O., Gros, V., Zhang, Y., Sciare, J., Simon, L., Truong, F., Bonnaire, N., Amodeo, T., Vautard, R., and Haeffelin, M.: Response of atmospheric 686 687 composition to COVID-19 lockdown measures during spring in the Paris region 688 (France), Atmos Chem Phys, 21, 17167–17183, https://doi.org/10.5194/acp-21-17167-689 2021, 2021. 690 Pinakidou, S.: People's perceptions of air pollution and their awareness of official indexes at 691 the start of the twenty-first century: a review, Discover Environment 2025 3:1, 3, 1–20, 692 https://doi.org/10.1007/S44274-025-00213-X, 2025. 693 Pye, H. O. T., Ward-Caviness, C. K., Murphy, B. N., Appel, K. W., and Seltzer, K. M.: 694 Secondary organic aerosol association with cardiorespiratory disease mortality in the 695 United States, Nat Commun, 12, 7215, https://doi.org/10.1038/s41467-021-27484-1, 696 2021. 697 Seinfeld, J. H., Wiley, J., and Pandis, S. N.: ATMOSPHERIC From Air Pollution to Climate 698 Change SECOND EDITION, 628-674 pp., 2006. 699 Shi, Z., Song, C., Liu, B., Lu, G., Xu, J., Van Vu, T., Elliott, R. J. R., Li, W., Bloss, W. J., and 700 Harrison, R. M.: Abrupt but smaller than expected changes in surface air quality 701 attributable to COVID-19 lockdowns, Sci Adv, 7, 6696-6709, 702 https://doi.org/10.1126/SCIADV.ABD6696/SUPPL FILE/ABD6696 SM.PDF, 2021. 703 Tian, J., Wang, Q., Zhang, Y., Yan, M., Liu, H., Zhang, N., Ran, W., and Cao, J.: Impacts of 704 primary emissions and secondary aerosol formation on air pollution in an urban area of 705 lockdown, Environ China during the COVID-19 Int, 150, 106426,

https://doi.org/10.1016/j.envint.2021.106426, 2021.

706

707 Tobler, A. K., Skiba, A., Wang, D. S., Croteau, P., Styszko, K., Necki, J., Baltensperger, U., 708 Slowik, J. G., and Prévôt, A. S. H.: Improved chloride quantification in quadrupole 709 aerosol chemical speciation monitors (Q-ACSMs), Atmos Meas Tech, 13, 5293–5301, 710 https://doi.org/10.5194/amt-13-5293-2020, 2020. 711 Transport for London: Travel in London Report 13, 2020. 712 UK Health Security Agency (UKHSA): Statement on the differential toxicity of particulate 713 constituents, matter according source or to 714 https://www.gov.uk/government/publications/particulate-air-pollution-health-effects-715 of-exposure/statement-on-the-differential-toxicity-of-particulate-matter-according-to-716 source-or-constituents-2022, 27 July 2022. 717 Vasilakopoulou, C. N., Matrali, A., Skyllakou, K., Georgopoulou, M., Aktypis, A., Florou, K., 718 Kaltsonoudis, C., Siouti, E., Kostenidou, E., Błaziak, A., Nenes, A., Papagiannis, S., 719 Eleftheriadis, K., Patoulias, D., Kioutsioukis, I., and Pandis, S. N.: Rapid transformation 720 of wildfire emissions to harmful background aerosol, NPJ Clim Atmos Sci, 6, 218, 721 https://doi.org/10.1038/s41612-023-00544-7, 2023. 722 Via, M., Chen, G., Canonaco, F., Daellenbach, K. R., Chazeau, B., Chebaicheb, H., Jiang, J., 723 Keernik, H., Lin, C., Marchand, N., Marin, C., O'dowd, C., Ovadnevaite, J., Petit, J.-E., 724 Pikridas, M., Riffault, V., Sciare, J., Slowik, J. G., Simon, L., Vasilescu, J., Zhang, Y., 725 Favez, O., Prévôt, A. S. H., Alastuey, A., and Cruz Minguillón, M.: Rolling vs. seasonal 726 PMF: Real-world multi-site and synthetic dataset comparison, Atmos Meas Tech, 15, 727 https://doi.org/10.5194/amt-15-5479-2022, 2022. 728 Wei, Y., Qiu, X., Yazdi, M. D., Shtein, A., Shi, L., Yang, J., Peralta, A. A., Coull, B. A., and

Schwartz, J. D.: The Impact of Exposure Measurement Error on the Estimated

729

730 Concentration-Response Relationship between Long-Term Exposure to PM2.5 and 731 Mortality, Environ Health Perspect, 130, https://doi.org/10.1289/EHP10389, 2022. 732 Wei, Y., Feng, Y., Danesh Yazdi, M., Yin, K., Castro, E., Shtein, A., Oiu, X., Peralta, A. A., 733 Coull, B. A., Dominici, F., and Schwartz, J. D.: Exposure-response associations 734 between chronic exposure to fine particulate matter and risks of hospital admission for 735 major cardiovascular diseases: population based cohort study, BMJ, e076939, 736 https://doi.org/10.1136/bmj-2023-076939, 2024. World Health Organization: WHO global air quality guidelines. Particulate matter (PM2.5 and 737 738 PM10), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide., Geneva, 2021. 739 Wu, D., Zheng, H., Li, Q., Wang, S., Zhao, B., Jin, L., Lyu, R., Li, S., Liu, Y., Chen, X., Zhang, 740 F., Wu, Q., Liu, T., Jiang, J., Wang, L., Li, X., Chen, J., and Hao, J.: Achieving health-741 oriented air pollution control requires integrating unequal toxicities of industrial 742 particles, Nature Communications 2023 14:1, 14, 1–12, https://doi.org/10.1038/s41467-743 023-42089-6, 2023. 744 Xu, J., Ge, X., Zhang, X., Zhao, W., Zhang, R., and Zhang, Y.: COVID-19 Impact on the 745 Concentration and Composition of Submicron Particulate Matter in a Typical City of 746 Northwest China, Geophys Res Lett, 47, https://doi.org/10.1029/2020GL089035, 2020. 747 Yao, X. and Zhang, L.: Identifying decadal trends in deweathered concentrations of criteria air 748 pollutants in Canadian urban atmospheres with machine learning approaches, Atmos 749 Chem Phys, 24, 7773–7791, https://doi.org/10.5194/acp-24-7773-2024, 2024. 750 Zhang, Qi., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. 751 R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., 752 DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A.,

753	Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S.,							
754	Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin,							
755	R. J., Rautiain	nen, J., Sun,	J. Y., Zhang, Y	Y. M., and V	Worsno	p, D. R	.: Ubic	luity and
756	dominance of	oxygenated	species in organi	ic aerosols in	n anthro	opogenio	cally-in	ıfluenced
757	Northern H	emisphere	midlatitudes,	Geophys	Res	Lett,	34,	n/a-n/a,
758	https://doi.org	/10.1029/200	07GL029979, 200	07.				
759								