

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

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Abstract

Organic aerosol (OA), a major component of submicron particulate matter (PM_{10}), has significant impacts on both human health and climate. Quantifying its sources is therefore crucial for developing effective mitigation strategies. Positive matrix factorisation (PMF) applied to aerosol chemical speciation monitor (ACSM) mass spectral data offers a robust approach for quantifying 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 demonstrated the importance of commercial cooking as a significant source of OA (20%) and PM_{10} (9%) in urban areas. The co-emission of BBOA with COA observed in Central London 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 limited regulation is in force.

1 Introduction

Atmospheric particulate matter (PM) are tiny particles suspended in the air, which impact the climate directly and indirectly (IPCC, 2021; Seinfeld et al., 2006), and cause adverse human health effects (Kelly and Fussell, 2012; World Health Organization, 2021). The PM present in urban areas, such as London, is emitted directly or indirectly from a wide range of natural and anthropogenic sources, can be changed through atmospheric reactions and remain airborne for many days. It is consequently a complex mixture including inorganic species (metals, minerals, black carbon, nitrate, sulphate, etc.) and thousands of organic compounds whose origins remain too complicated to fully quantify. PM_{2.5} (PM with aerodynamic diameter smaller than 2.5 μm) is strongly associated with increased risks of cardiovascular and respiratory related mortalities and 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 begun to demonstrate that some PM constituents and sources have stronger associations with a range of health metrics, including mortality, morbidity, and toxicities although the evidence remains inconsistent (Kelly and Fussell, 2012; Liu et al., 2023; Vasilakopoulou et al., 2023). With 99% of the urban population in Europe exposed to PM_{2.5} concentrations exceeding the WHO air quality guideline (European Environment Agency, 2024; World Health Organization, 2021), delivering clean air is a target for European and international governments according to the EU air quality directive (European Union, 2024). However, delivering publicly acceptable policies to improve air quality remains challenging (Mebrahtu et al., 2023; Oltra et al., 2021). Targeting the sources of PM that are most health-relevant could be a more cost-effective (Wu et al., 2023), more easily communicated and more publicly acceptable approaches (Pinakidou, 2025) to improve public

health. It is therefore important to better quantify the sources of PM and understand how they respond to policy interventions.

The COVID-19 pandemic is a natural experiment to assess the impact of policies which, while not aiming to reduce PM_{2.5} concentrations, significantly restricted social and economic activities and consequently reduced emissions. During the UK national lockdown, people were ordered to stay at home, and all non-essential businesses were closed, including pubs, cafes and restaurants from the end of Mar 26th, 2020. Non-essential shops were allowed to open from Jun 15th, and the first national lockdown came to an end on Jun 23rd, 2020. However, pubs, restaurants, and cafes were only allowed to open from July 4th, 2020. The UK recorded a 2.5% drop in Gross Domestic Product (GDP) in the first quarter of 2020, partly as people reduced their own activity prior to national lockdown. This accelerated to a 19.8% fall in GDP in April to June 2020 and household spending fell by over 20% over this period, the largest quarterly contraction on record, which was driven by falls in spending on restaurants, hotels, transport, and recreation (ONS, 2022). The UK Government Eat Out to Help Out (EOTHO) Scheme is examined specifically in this study as it influenced emissions from the commercial cooking sector. It was designed to help the 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.

While the impact of these lockdown policies on some air quality metrics was smaller than expected given the large change in emissions (Shi et al., 2021), the abrupt nature of the intervention ensures it is easier to detect than other air quality policies that are more incremental in nature (Mudway et al., 2019). Some studies have investigated the lockdown impacts on chemical composition and sources of PM, which mainly focused on cities in China (Hu et al., 2022; Tian et al., 2021; Xu et al., 2020), a kerbside site in Toronto, Canada (Jeong et al., 2022), and an urban background site in

83 Paris, France (Petit et al., 2021). These studies all resolved primary sources including traffic related
84 emissions, biomass burning emissions from residential heating, cooking emissions (except Paris),
85 and secondary sources from PMF analysis on organic aerosol (OA). Traffic and cooking emissions
86 appeared to decrease during the lockdown in all sites, while biomass burning predominately from
87 residential heating sources in Chinese cities increased as result of remote work and rather early
88 lockdown measures (Jan-Feb 2020) compared to France. Secondary organic aerosol (SOA)
89 showed a more complex phenomenon given its abundance in organic components and dynamic
90 spatiotemporal conditions. Overall, the lockdowns resulted in decreased SOA in both northwest
91 cities in China (Tian et al., 2021; Xu et al., 2020) and Paris (Petit et al., 2021) due to lower primary
92 emissions, and therefore fewer SOA formation products. However, Beijing experienced a large
93 increase in SOA concentrations due to increased fossil fuel and biomass burning emissions, long-
94 range transport influences as well as favourable meteorological conditions (high RH, low wind
95 speed and low boundary layer height) for SOA formation during the lockdown period (Hu et al.,
96 2022). Therefore, the lockdown effects on the SOA were dependent on the abundance of primary
97 emissions, long-range transported air masses, and meteorological conditions. To date, there are
98 few studies that investigate how COVID-related policies could have impacted PM chemical
99 composition and sources. Petit et al. (2021) and Gamelas et al. (2023) are the only two studies in
100 Europe. The unique COVID-related policies in the UK therefore provide a rare opportunity to
101 investigate the impacts these policies had on chemical composition and OA. We used highly time
102 resolved measurements from an air quality supersite located in the Central London from 2019 to
103 2020, and advanced source apportionment approaches to quantify the PM composition and OA
104 sources before, during and after the UK national lockdown and EOTHO scheme. This study
105 provides valuable 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

2.1 Air quality monitoring supersite in 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 London. It is a well-established 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://uk-air.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 PM₁ 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 PM₁ concentrations well, with a slope of 1.13 and an R² 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 EOTH Scheme (Mon-Wed during from Aug 3rd to Aug 31st, 2020, Table 1). 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 (Table 1).

Table 1 Dates of the COVID-related policies in London

COVID Policies		Date
Pre-Lockdown	Summer	Aug 1 st –Aug 31 st , 2019
	Fall	Sep 1 st –Nov 30 th , 2019
	Winter	Dec 1 st , 2019–Feb 28 th , 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

Post-Lockdown	Pre-EOTH0	Jun 24 th –Aug 2 nd , 2020
	EOTH0	Aug 3 rd –Aug 31 st , 2020
	Post-EOTH0	Sep 1 st –Oct 22 nd , 2020

2.4 Source apportionment

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 well-studied, 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 non-refractory PM, in which, organic mass spectra are used for PMF analysis. However, operating an AMS is labour-intensive 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.

Advanced source apportionment approaches have been used in this study, including rolling positive matrix factorization (PMF), ME-2 with random α -value approach, bootstrap and criteria-based 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 splitting 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 α -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 α -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 α -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

3.1 Model performance of meteorological normalisation

The performance of each model (for individual species/source) is shown in Table S2 with slopes from 0.97 to 1.02 and R^2 (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, 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.

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

The average PM_{10} mass concentration at MY site was $11 \mu g/m^3$ for the study period with 44% OA, 21% NO_3 , 15% SO_4 , 16% BC, 5% NH_4 , and 0.6% Cl. The distribution of the chemical composition on PM_{10} varied depending on the season and variation was associated with the lockdown and

228 EOTHO policies (Figure 1). PM_{10} increased by 95% in lockdown spring (Mar 26th–May 31st, 2020)
229 compared to pre-lockdown spring (Mar 1st–Mar 25th, 2020). Specifically, Org, SO_4 , NO_3 , NH_4 ,
230 and Cl all increased by 87%, 211%, 73%, 237%, and 132%, respectively. Except for BC, which
231 decreased by 52%. This is due to the polluted air mass originating from mainland Europe and the
232 enhanced agricultural emissions in spring from the UK and wider continental Europe (Aksoyoglu
233 et al., 2020). It was further confirmed, through back trajectory analysis, that elevated PM_{10} events
234 (Mar 25th–Mar 28th, Apr 8th–Apr 10th, and Apr 15th–Apr 17th), where the result of air masses passing
235 over northern continental Europe (Fig. S6). In addition, the Org, SO_4 , NO_3 , NH_4 , and Cl were only
236 increased by 21%, 107%, 50%, and 28% respectively after the deweathering analysis (Fig. S8),
237 suggesting significant meteorological influences during this period. NO_3 concentration reduced in
238 summer 2019 and 2020 as expected compared to spring or fall seasons due to the semi-volatile
239 nature of NH_4NO_3 and lower agricultural emissions, while SO_4 concentrations increased in
240 summer due to enhanced photochemistry across Europe (Bressi et al., 2021; Chen et al., 2022).
241 During the lockdown in spring, SO_4 concentrations remained high, which was associated with
242 long-range transport and marine aerosols (e.g., methanesulfonic acid, MSA) (Fig. S7).

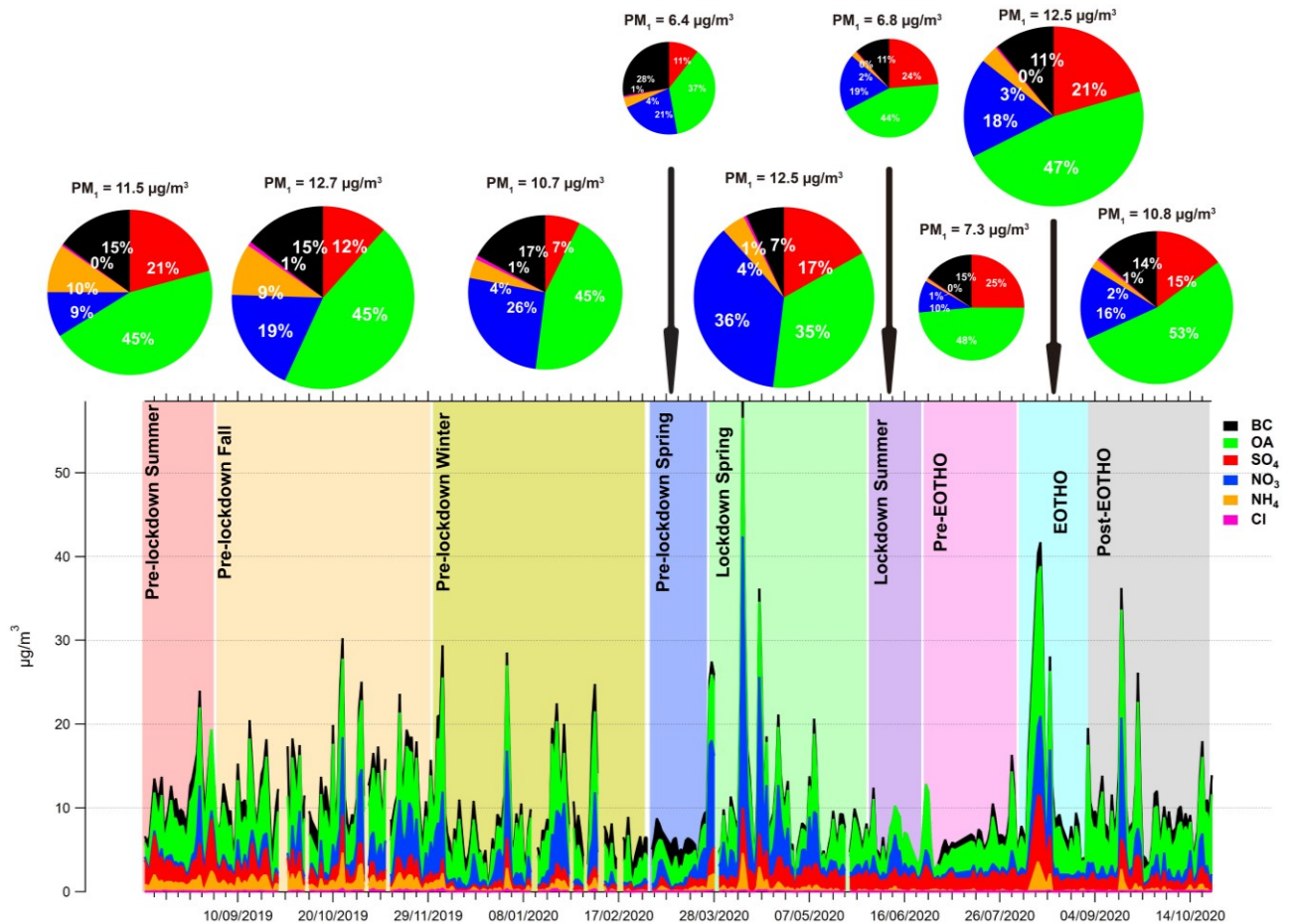


Figure 1 Chemical compositions of PM_{10} at MY from Aug 2019 to Oct 2020 (daily resolution) and averaged for the different periods as shown in Table 1.

BC concentrations during the spring lockdown (Mar 26th–May 31st 2020) reduced from 1.78 to 0.86 $\mu\text{g}/\text{m}^3$ (-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 $\mu\text{g}/\text{m}^3$

(+44%) after the lockdown and before the EOTHO (Jun 24th–Aug 2nd, 2020, pre-EOTHO in Figure 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 $\mu\text{g}/\text{m}^3$ (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 $\mu\text{g}/\text{m}^3$ (+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).

3.3 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 3 and Figure 4. The left panel of Figure 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 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).

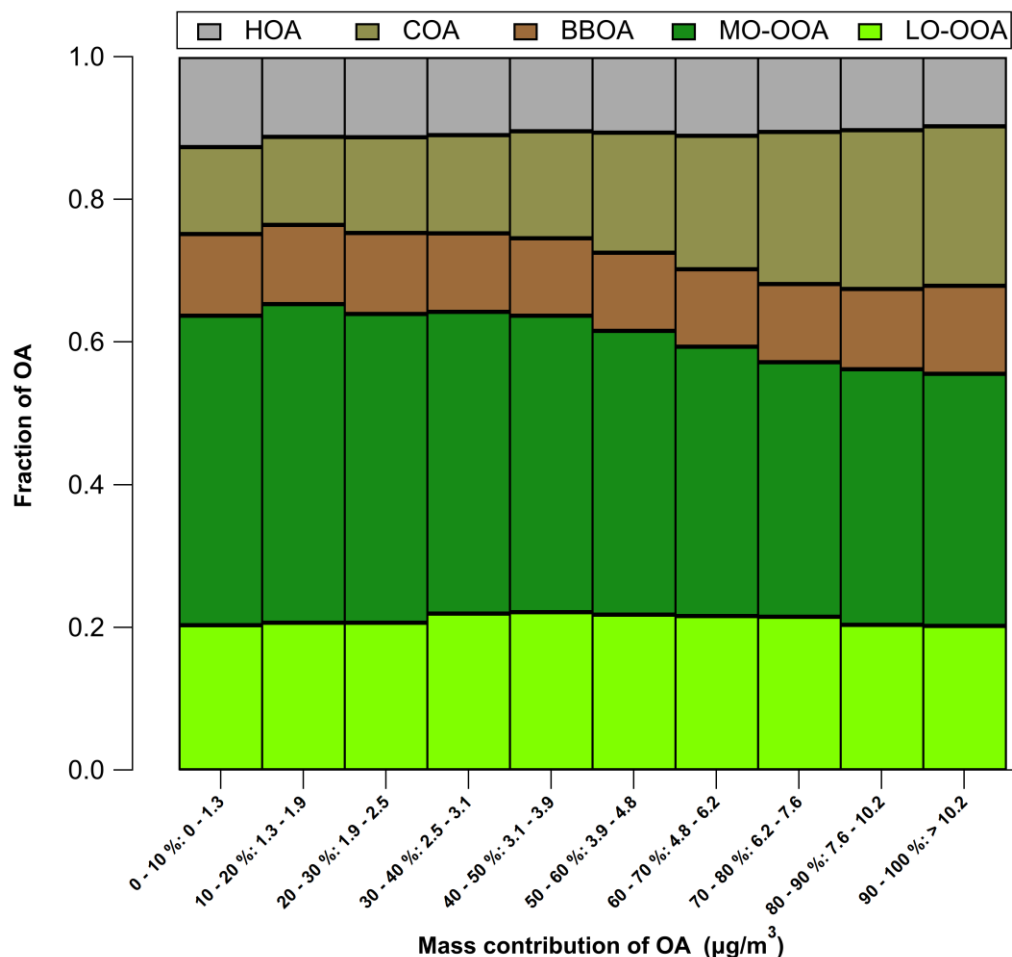


Figure 2 Contributions to total OA from the different identified OA sources at different OA concentrations. Total OA concentrations were split in 10 equally distributed bins.

3.3.1 General characteristics of OA factors

The right panel of Figure 3 shows time series (daily averaged) and Figure 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 $0.50 \pm 0.1 \mu\text{g}/\text{m}^3$, $0.93 \pm 0.14 \mu\text{g}/\text{m}^3$, $0.55 \pm 0.11 \mu\text{g}/\text{m}^3$, $1.81 \pm 0.41 \mu\text{g}/\text{m}^3$, $1.00 \pm 0.44 \mu\text{g}/\text{m}^3$, and $2.80 \pm 0.70 \mu\text{g}/\text{m}^3$, and 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 4. OA factors also showed seasonality besides

the effects from COVID-related policies (Figure 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 consumption since Central London mainly uses natural gas and renewable energy rather than solid fuel combustions (Cliff et al., 2025). The OOA factor concentrations remain relatively consistent across seasons, while its contribution was larger during the warmer seasons (Fig. S4). This is because both high temperature and strong solar radiation will enhance the photochemistry and evaporation of POA sources, and increased 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 London was consistent with the other urban sites across Europe (Chen et al., 2022). Therefore, this study focuses on the impacts of COVID-related policies on OA sources.

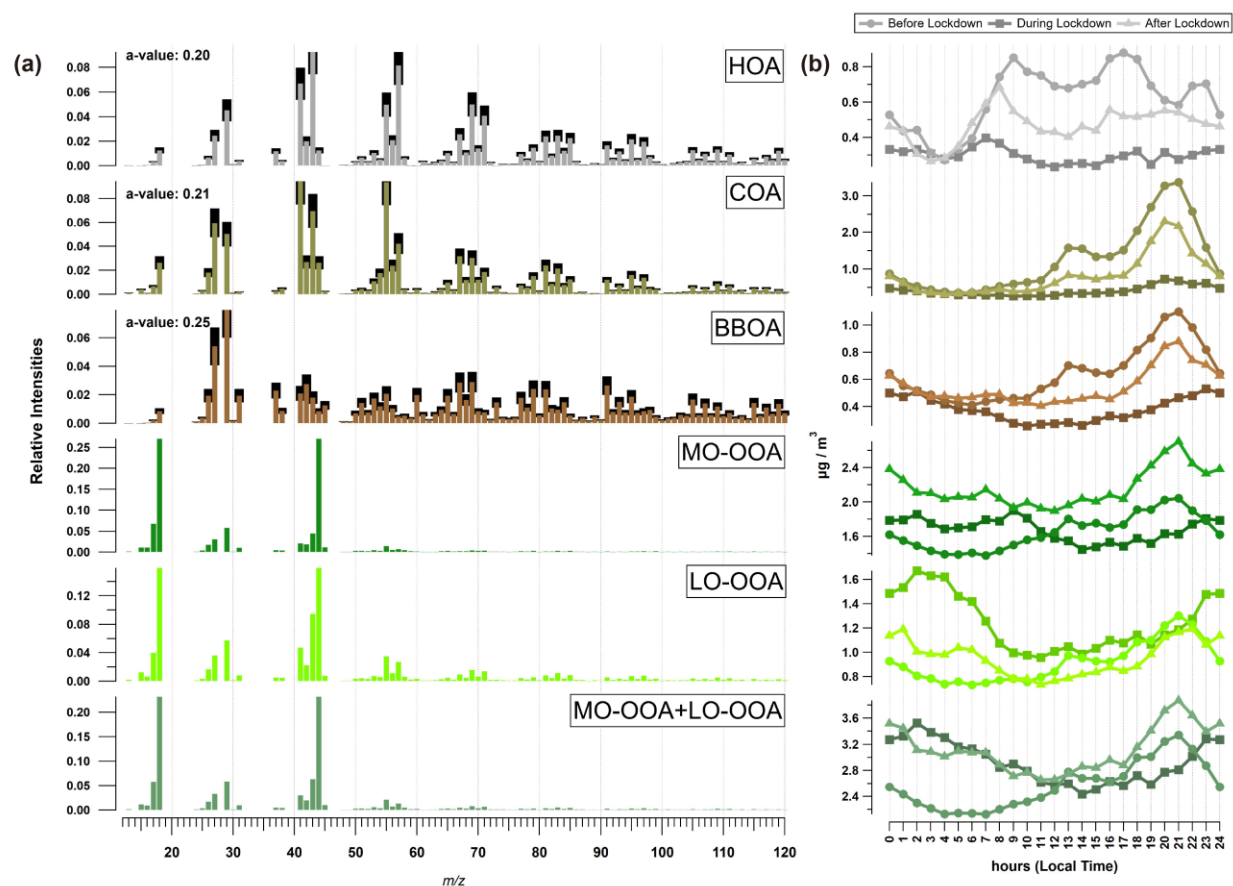


Figure 3 Yearly averaged profiles (a) and diurnal cycles (b) of resolved factors from the rolling PMF analysis at the MY site. Time is expressed in local time.

The right side of 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 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).

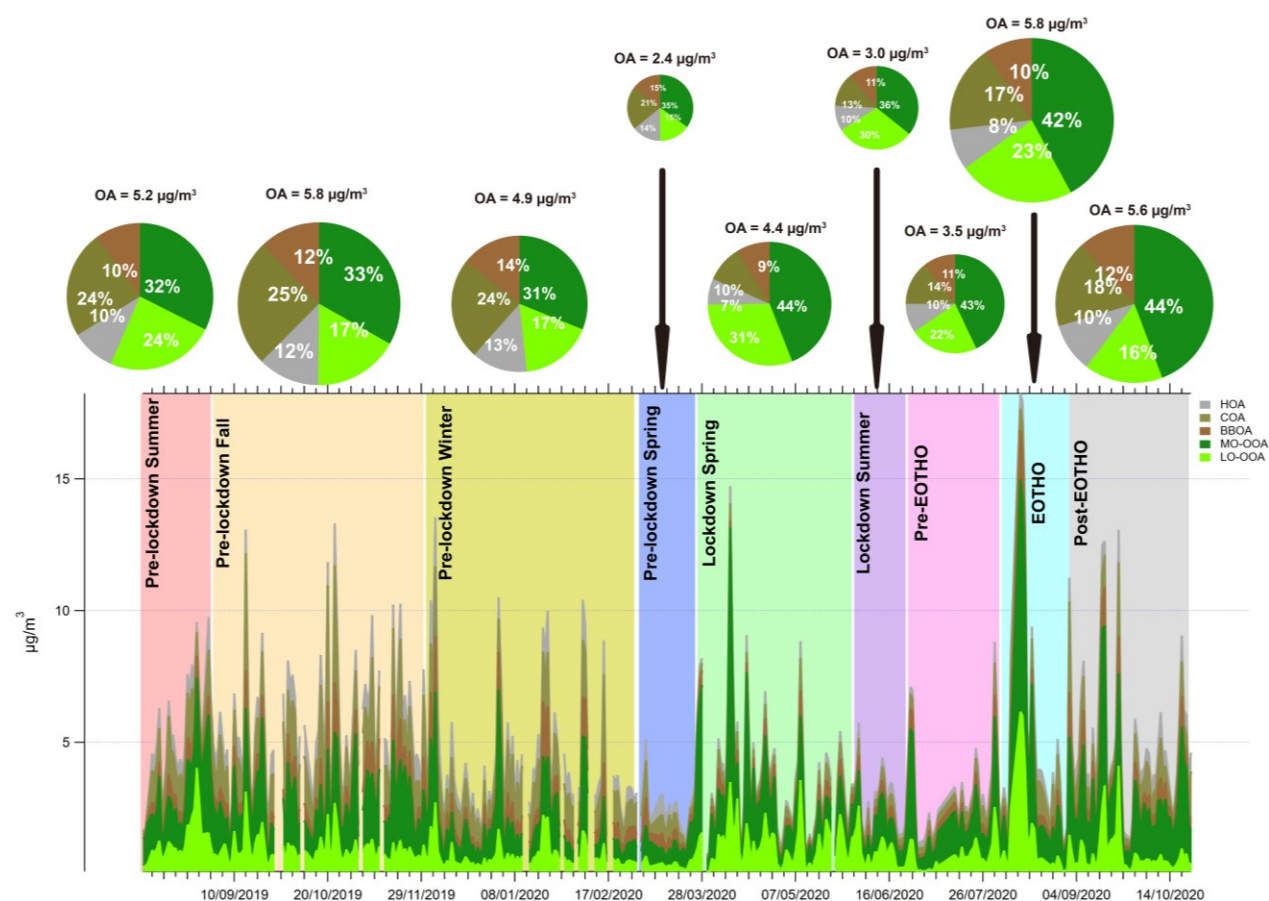


Figure 4 Average mass concentrations for OA sources at MY during different periods from Aug 2019 to Oct 2020

3.3.2 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 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.

3.3.3 Lockdown

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 b). HOA retained distinct morning and evening rush hour peaks but at lower mass concentrations during lockdown (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 5). This is likely due to widespread hybrid working and the remaining oversea travel restrictions suppressing 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 $\mu\text{g}/\text{m}^3$) (Figure 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 accompanied by increased SO_4 (+211%), NH_4 (+132%) and NO_3 (+237%) as shown in Fig. S9, despite the higher temperature could favour partitioning these species into the gas phase.

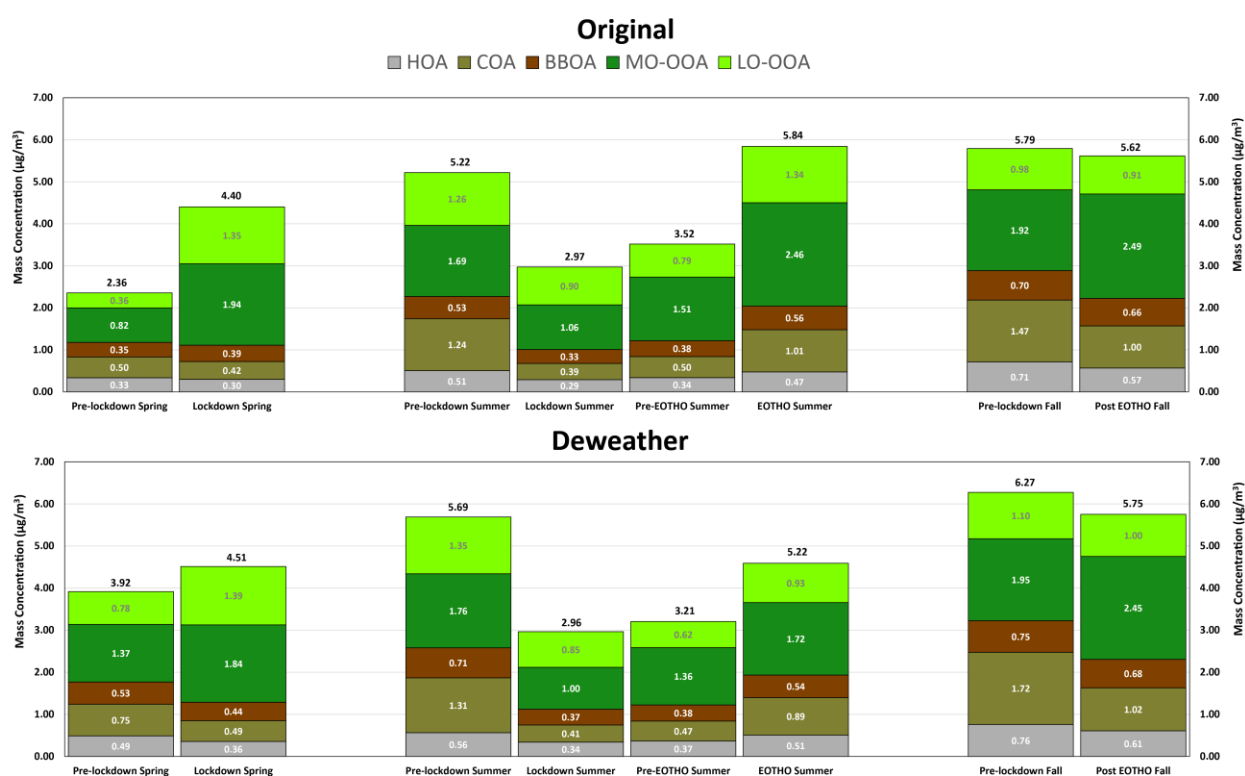


Figure 5 The impacts on OA sources during different periods compared with business-as-usual cases with and without deweathering analysis.

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 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 air masses (Fig. S5).

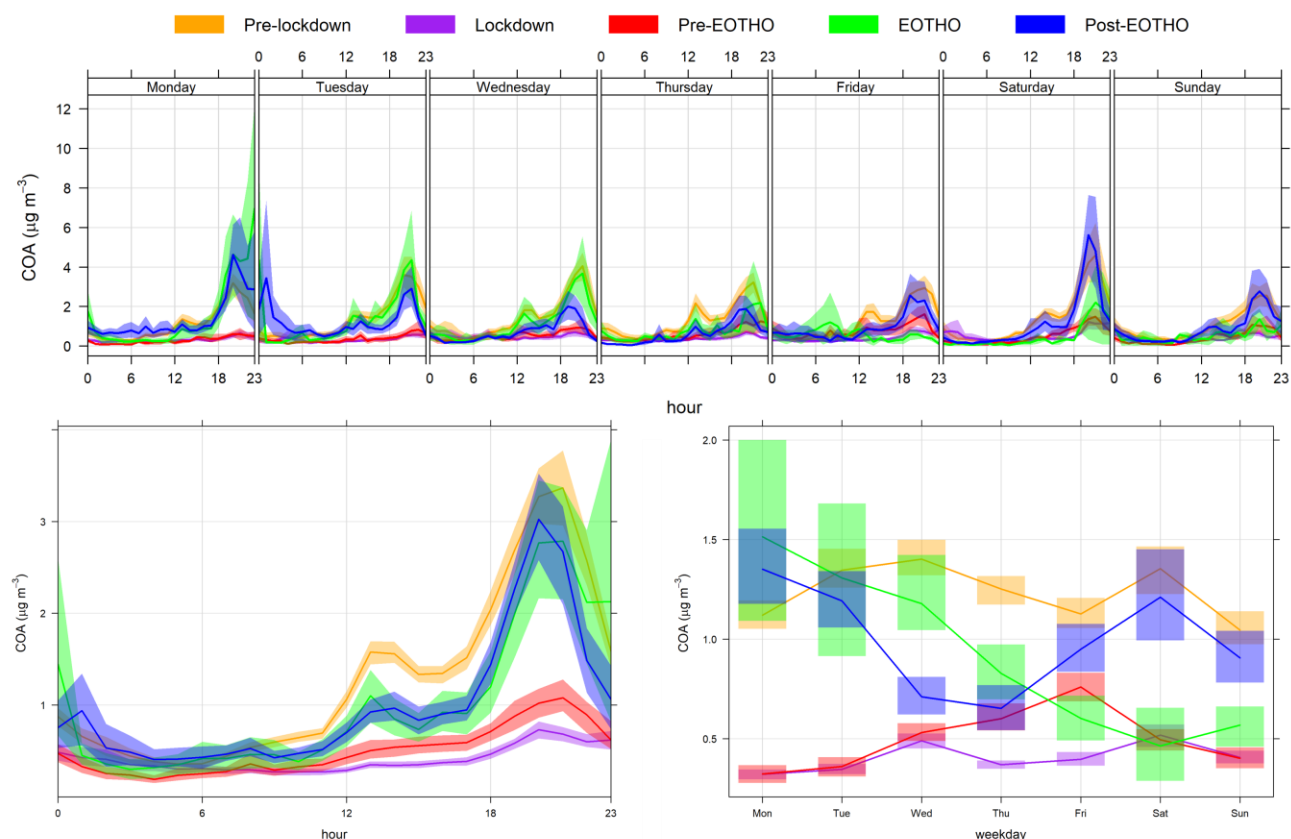
3.3.4 Pre-EOTH

During pre-EOTH (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-OA increased by 45% and LO-OA decreased by 13%, respectively as the relatively higher temperature and solar radiation favoured the evaporation of LO-OA and production of MO-OA from LO-OA and POA. As shown in Figure 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, lower HOA (-33%), BC (-37%) due to reduced vehicle mileage resulted, and COA (-59%) due to the reduced commercial cooking activity. As BBOA is co-emitted with COA during of cooking activities, BBOA also decreased slightly from 0.53 to 0.38 $\mu\text{g}/\text{m}^3$ (-28%, Figure 5).

3.3.5 Eat Out To Help Out (EOTH)

EOTH 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-EOTH Summer) from Jun 24th to Aug 2nd, 2020 (0.5 to 1.0 $\mu\text{g}/\text{m}^3$, Figure 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 EOTH, however, it did not reach pre-

384 COVID concentrations (Figure 5 and Figure 6) as its level was lower on each weekday except for
 385 Mon.



387 *Figure 6 . COA diurnal plots for each weekday, diurnal plots, and weekly plots at different periods in relation with COVID-*
 388 *related policies*

389 EOTHO only operated from Mon to Wed, and this was clear in the diurnal plots (Figure 6 and Fig.
 390 S12) with larger COA concentrations from Mon to Wed, in contrast with larger concentrations
 391 over the weekend (Fri to Sun) before EOTHO (Jun 24th–Aug 2nd, 2020). Interestingly, even after
 392 the EOTHO policy ceased (Sep 1st–Oct 22nd, 2020), COA levels remained elevated on Mon and
 393 Tue but a much higher level during the weekend was observed. This suggests that EOTHO had an
 394 influence on the consumer behaviour even after the lockdown. It is also worth noting that the high
 395 concentrations of COA and BBOA (Figure 6 and Fig. S11) on Monday night were caused by the
 396 last day of EOTHO policy coinciding with a UK public holiday on Aug 31st.

Also, during EOTH0, MO-OOA and LO-OOA increased by 63% and 70% respectively compared to post-lockdown concentrations before EOTH0 and correlated with increased NH_4 , NO_3 and SO_4 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 (EOTH0) 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 EOTH0 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.

Code/Data availability

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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