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

3

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16 Abstract

17 Particulate matter (PM) poses both health and climate risks. Understanding pollution sources is
18 therefore crucial for effective mitigation. Positive Matrix Factorization (PMF) of Aerosol
19 Chemical Speciation Monitor (ACSM) data is a powerful tool to quantify organic aerosol (OA)
20 sources. A year-long study of ACSM data from London's Marylebone Road monitoring station
21 during the COVID-19 pandemic provides insights into the impact of lockdown and the Eat Out To
22 Help Out (EOTHO) scheme, which offered support to the hospitality during the pandemic, on PM
23 composition and OA sources. Five OA sources were identified including hydrocarbon-like OA
24 (HOA, traffic-related, 11% to OA), cooking OA (COA, 20%), biomass burning OA (BBOA, 12%),
25 more-oxidized oxygenated OA (MO-OOA, 38%), and less-oxidized oxygenated OA (LO-OOA,
26 21%). Lockdown significantly reduced HOA (-52%), COA (-67%), and BBOA (-41%) compared
27 to their pre-COVID levels, while EOTHO increased COA (+38%) significantly compared to the
28 post-lockdown period. However, MO-OOA and LO-OOA were less affected, as these primarily
29 originated from long-range transport. This research has highlighted the importance of commercial
30 cooking as a significant source of OA (20%) and PM_{10} (9%) in urban areas. The co-emission of
31 BBOA with COA observed in Central London demonstrates a similar diurnal cycle and response
32 to the EOTHO policy, indicating that cooking activities might be currently underestimated and
33 contribute to urban BBOA. Therefore, more effort is required to quantify this source and develop
34 targeted abatement policies to mitigate emissions as currently limited regulation is in force.

35



36 1 Introduction

37 Atmospheric particulate matter (PM) are tiny particles suspended in the air, which can not only
38 impact the climate directly and indirectly (IPCC, 2021; Seinfeld et al., 2006), but also cause
39 adverse health effects to human (Kelly and Fussell, 2012; World Health Organization, 2021). PM
40 consist of various constituents, including inorganic species (metals, minerals, black carbon, nitrate,
41 sulphate, etc.) and organic species (complex mixture of thousands of compounds). European
42 Environment Agency has reported that 99% of urban population in Europe are still exposed to
43 polluted air with annual PM_{2.5} (PM with aerodynamic diameter smaller than 2.5 μm)
44 concentrations exceeding the WHO air quality guideline, 5 μg/m³ (Europe's air quality status 2024,
45 2024; World Health Organization, 2021). As the most health-relevant air pollutant, PM_{2.5} has
46 shown strong associations with cardiovascular and respiratory related mortalities and hospital
47 admissions (Dominici et al., 2006; Wei et al., 2022, 2024). Several studies have demonstrated that
48 different constituents/sources contribute to health effects differently with varying toxicities (Kelly
49 and Fussell, 2012). Therefore, targeting the specific composition/sources of PM that are most
50 health-relevant could be the most cost-effective way to mitigate its adverse health effects.

51 Source apportionment is a common but powerful approach to identifying and quantifying the
52 emission sources and atmospheric constituents of PM based on measurements. As the sources of
53 inorganic species (black carbon, ammonium, nitrate, chloride, sulphate, etc.) are relatively well-
54 studied, most of the studies are focused on deconvoluting the sources of organic aerosol (OA),
55 which contains thousands of compounds. Positive matrix factorization (PMF) is one of the receptor
56 models that is widely utilized in the field to conduct source apportionment analysis. Typically, an
57 Aerodyne aerosol mass spectrometer (AMS, Aerodyne Ltd., USA, Jayne et al., 2000) is used to
58 measure the time series of both inorganic and organic species of non-refractory PM, in which,



59 organic mass spectra are used for PMF analysis. However, operating an AMS is labour-intense
60 and expensive. In contrast, the aerosol chemical speciation monitor (ACSM, Aerodyne, Ltd.,
61 Fröhlich et al., 2013; Ng et al., 2011) has been designed for long-term monitoring purposes with
62 less maintenance and lower capital cost, which has gained popularity across Europe (Chebaicheb
63 et al., 2024; Chen et al., 2022) and the U.S. (<https://ascent.research.gatech.edu/>). Chen et al. (2022)
64 demonstrated a robust protocol to conduct advanced PMF analysis on long-term ACSM datasets,
65 which delivers high-quality and consistent source apportionment results. This study follows this
66 standardized protocol to resolve the OA sources in London by implementing advanced PMF
67 techniques.

68 Coronavirus disease 19 (COVID-19) started to spread rapidly worldwide since the first case was
69 identified in Wuhan, China late in 2019. Many countries implemented measures to contain COVID
70 cases, which significantly restricted social and economic activities. In the UK, starting from the
71 end of Mar 26th, 2020, people were ordered to stay at home and all non-essential businesses were
72 closed, including pubs, cafes and restaurants. Non-essential shops were allowed to open on Jun
73 15th, and the first national lockdown came to an end Jun 23rd, 2020. However, pubs, restaurants,
74 and cafes were only allowed to open from July 4th, 2020. Subsequently, the Eat Out to Help Out
75 (EOTHO) Scheme was designed to help the hospitality industry; offering a 50% meal discount up
76 to a maximum of £10 and operated Monday to Wednesday during from Aug 3rd to Aug 31st, 2020;
77 <https://www.gov.uk/guidance/get-a-discount-with-the-eat-out-to-help-out-scheme>.

78 The UK recorded a 2.5% drop in Gross Domestic Product (GDP) in the first quarter of 2020, partly
79 as people reduced their own activity prior to the legally enforced lockdown measures introduced
80 on Mar 26th. This accelerated to a 19.8% fall in GDP in April to June 2020 and household spending
81 fell by over 20% over this period, the largest quarterly contraction on record, which was driven by



82 falls in spending on restaurants, hotels, transport, and recreation (GDP and events in history: how
83 the COVID-19 pandemic shocked the UK economy, 2024).

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



105 policies had on chemical composition and OA sources. To address these issues, we used highly
106 time resolved measurements from an air quality supersite located in the Central London from 2019
107 to 2020, and advanced source apportionment approaches to quantify the influence of the first
108 lockdown and EOTHO scheme on the PM composition and OA sources. This provides unique
109 insight into PM sources and composition in a global mega city.

110 2 Methodology

111 2.1 Air quality monitoring supersite in central London

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

118 2.2 Instrumentations

119 Quadrupole ACSM (Q-ACSM, Aerodyne, Ltd., Ng et al. (2011)) provides 30-min mass loadings
120 of chemical species within non-refractory submicron aerosol (NR-PM₁), including NH₄, NO₃, SO₄,
121 Cl, and OA. Sampled particles are focused into a narrow beam using the aerodynamic lens and
122 impacted on a filament surface at 600 °C, where the NR-PM₁ is vaporised and ionised instantly by
123 an electron impact source (70eV). These ions are detected by the RGA quadrupole mass
124 spectroscopy to provide a mass spectrum of NR-PM₁ up to a mass-to-charge ratio (m/z) of 148 Th.
125 The mass concentration of different chemical species are calculated using the fragmentation table



126 developed by Allan et al. (2004), updated for CI following suggestions provided by Tobler et al.
127 (2020), and a (Canagaratna et al., 2007; Matthew et al., 2008) composition-dependent collection
128 efficiency (CDCE) correction suggested by Middlebrook et al. (2012) by following the ACTRIS
129 standard operation procedure ([https://www.actris-ecac.eu/pmc-non-refractory-organics-and-](https://www.actris-ecac.eu/pmc-non-refractory-organics-and-inorganics)
130 [inorganics](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
131 (Aerosol Magee Scientific, Ltd.) and PM₁ measurements using FIDAS (Palas, GmbH), we
132 conducted the mass closure for fine particles measurements. The sum of NR-PM₁ and BC (in PM_{2.5})
133 reproduces PM₁ concentrations well, with a slope of 1.13 and an R² of 0.73 (Fig. S1).

134 *2.3 Sampling periods and COVID-related policies*

135 PM₁ chemical composition from Aug 1st, 2019 to Oct 22nd, 2020, was analysed as this covered the
136 first lockdown period (Mar 26th–23 Jun 23rd, 2020) and the EOTHO Scheme (Mon-Wed during
137 from Aug 3rd to Aug 31st, 2020, Table 1). In order to isolate the seasonal effects on the PM chemical
138 composition and OA sources from the COVID-related policies, we further split the data based on
139 seasons (Table 1).

140



141

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

142 *2.4 Source apportionment*

143 Positive matrix factorization (PMF) has been widely deployed in source apportionment of PM
144 components including OA from ACSM/AMS datasets collected worldwide (Chebaicheb et al.,
145 2024; Chen et al., 2022; Jimenez et al., 2009; Ng et al., 2011b; Zhang et al., 2011). The PMF
146 algorithm on environmental monitoring data was initially introduced by Paatero and Tapper, (1994)
147 as follows:

$$x_{ij} = \sum_{k=1}^p g_{ik} \times f_{kj} + e_{ij} \quad (1)$$

148 where x_{ij} is the measurement matrix (here, the time series of organic mass spectra from the ACSM
149 at i^{th} time and j^{th} m/z), g_{ik} is the mass concentration at i^{th} time in k^{th} factor, f_{kj} is the relative



150 intensity of j^{th} m/z for k^{th} factor, and e_{ij} stands for the residuals for j^{th} m/z at i^{th} time, p is the number
151 of factors. The PMF model iteratively minimises the Q value using the least-squares algorithm as:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{\sigma_{ij}} \right)^2 \quad (2)$$

152 where n is the number of data points, m is the total number of m/z , and σ_{ij} is the measurement
153 uncertainty estimated before the PMF analysis at i^{th} time for j^{th} m/z .

154 However, PMF suffers from rotational ambiguity (Paatero et al., 2002), which provides non-
155 unique solutions (i.e., similar Q value with different time series and factor profiles). These
156 solutions typically will not be equally environmentally reasonable, even with similar Q values.
157 The multilinear engine ME-2 (Paatero, 1999; Paatero and Hopke, 2009) is a robust approach to
158 reduce the rotational ambiguity and can direct PMF towards environmentally reasonable solutions
159 (both factor profiles and time series).

160 Here, PMF was implemented using the Source Finder v9.5.1.3 (Datalystica Ltd., Switzerland,
161 Canonaco et al. 2013) with the ME-2 solver. The latter imposes *a priori* information on the factor
162 solutions and/or time series. The a -value (ranging from 0 to 1) represents the upper limit of the
163 relative deviation for a factor profile (f_j) or time series (g_i) from the chosen *a priori* input profile
164 (F_j) or time series (G_i) during the iterative least-square minimization (Equation 2), as shown in
165 Equations 3a and 3b (Canonaco et al., 2013):

$$f_j = F_j \pm a \cdot F_j \quad (3a)$$

$$g_i = G_i \pm a \cdot G_i \quad (3b)$$



166 PMF analysis is usually performed using the whole dataset, assuming that the OA source profiles
167 are static over the entire period, which can lead to high errors when it comes to long-term datasets
168 with non-negligible temporal variabilities of OA chemical fingerprints. Canonaco et al. (2015)
169 showed a considerable seasonal variability of oxygenated organic aerosol (OOA) factor profiles,
170 especially between winter and summer in a dataset in Switzerland. Parworth et al. (2015) first
171 introduced the concept of rolling PMF by shortening the analysis period to a smaller time window
172 (e.g., 14 days) and then rolling over the whole dataset with a certain step (i.e., 1 day). This
173 technique was further refined and implemented into SoFi by Canonaco et al. (2021), which allows
174 the PMF model to adapt the temporal variabilities of the source profiles (e.g., biogenic versus
175 biomass burning influences on OOA factors), which usually provides well-separated OA factors.

176 Bootstrapping (Efron, 1979) analysis will randomly select part of the PMF input matrix and
177 duplicates itself to recreate a matrix with the same dimension as the original PMF input matrix.
178 The statistical and rotational uncertainties of the PMF results will then be evaluated by bootstrap
179 and the random α -value approach with at least 50 repeats per rolling window (Canonaco et al.,
180 2021; Chen et al., 2021). The standardized protocol of rolling PMF as presented in Chen et al.
181 (2022) was used to ensure high-quality and comparable sources of OA were retrieved in London.

182 Specifically, PMF was first done on four different seasons as suggested in Chen et al. (2022) to
183 determine the optimum number of factors. A total of 5 OA factors were identified: hydrocarbon-
184 like OA (HOA), cooking-like OA (COA), biomass burning OA (BBOA), more-oxidized OOA
185 (MO-OOA) and less-oxidized OOA (LO-OOA). In addition, site-specific factor profiles were
186 derived for HOA, COA, and BBOA through a seasonal bootstrap PMF analysis for winter (Dec,
187 Jan, and Feb) and used as constraints as suggested in Chen et al. (2022) and Via et al. (2022).
188 However, the MY site is surrounded by many restaurants with prevalent cooking emissions. Thus,



189 the chemical fingerprint for both HOA and COA might not be fully separated. Therefore, we
190 constrained the trend of NO_x time series, BBOA and COA profiles from a previous winter
191 bootstrap solution collected in London North Kensington (2015-2018, Chen et al., 2022) to retrieve
192 environmentally reasonable results with five factors, so-called base case solution. Then, a
193 bootstrap resampling analysis with 100 iterations and five factors was conducted by constraining
194 the factor profiles of HOA, COA, and BBOA from the base case with random a-value from 0.1-
195 0.5 with step of 0.1. It results in stable factor profiles of these three primary sources as shown in
196 Figure S2, which shows good agreements with published reference profiles (Chen et al., 2022;
197 Crippa et al., 2013).

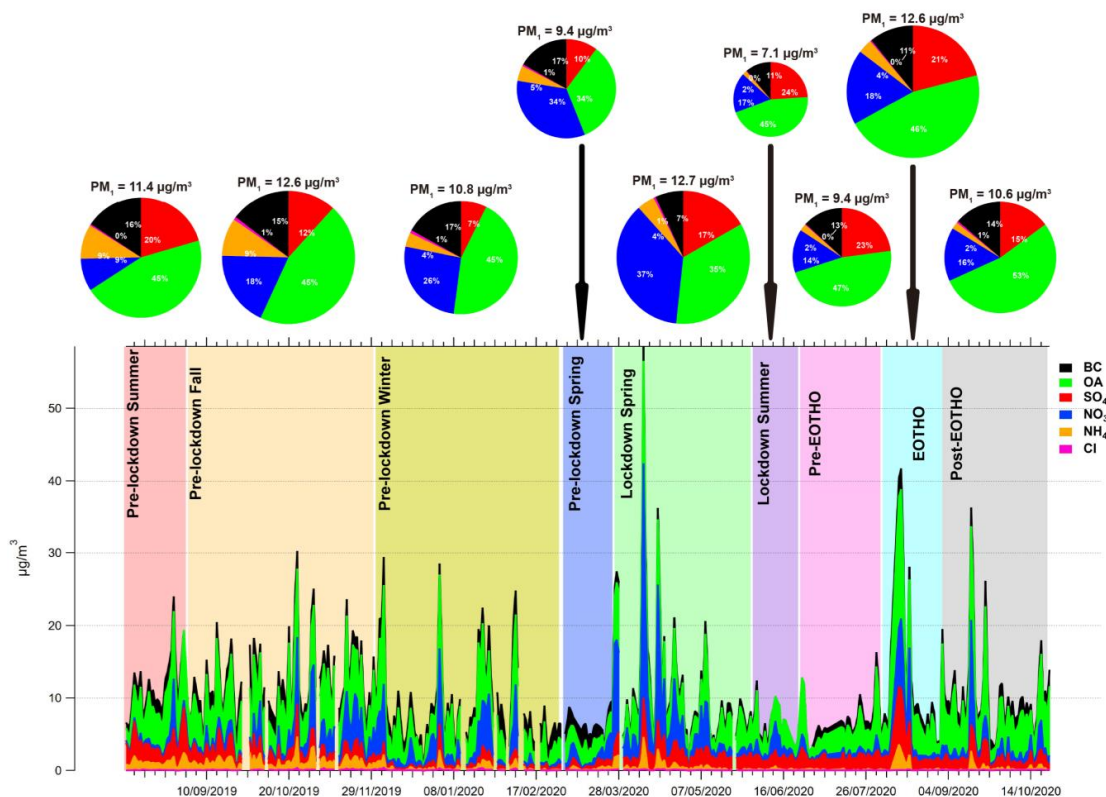
198 By constraining primary factor profiles of HOA, COA, BBOA in Figure S2 (averaged bootstrap
199 results) and two additional unconstrained factors with bootstrap resampling and the random a-
200 value option (0.1-0.5, step of 0.1), rolling PMF is conducted with a time window of 14 days and a
201 step of 1 day. A criteria list including selections based on both time series and factor profiles as
202 shown in Table S1 was applied as per Chen et al. (2022). With the help of t-test in temporal-based
203 criteria (1-3), we can minimize subjective judgements in determining the environmentally
204 reasonable results. Eventually, 3,166 runs (14.1%) of the PMF runs were selected to average as
205 the final results with 4.9 % unmodelled data points, which is comparable with other rolling PMF
206 analyses (Chen et al., 2022).



207 3 Results and Discussions

208 3.1 Chemical composition of submicron PM for different periods around the 209 COVID-19 Lockdown

210 The average PM₁ mass concentration at MY site was 11 µg/m³ for the study period with 44% OA,
211 21% NO₃, 15% SO₄, 16% BC, 5% NH₄, and 0.6% Cl. The distribution of the chemical composition
212 on PM₁ varied depending on the season and variation was associated with the lockdown and
213 EOTO policies (Figure 1). PM₁ increased by 34% in lockdown spring (Mar 26th–May 31st, 2020)
214 compared to pre-lockdown spring (Mar 1st–Mar 25th, 2020), as well as NO₃ and NH₄, the later
215 most likely originated from enhanced agricultural emissions in spring from the UK and wider
216 continental Europe (Aksoyoglu et al., 2020). It was further confirmed, through back trajectory
217 analysis, that elevated PM₁ events (Mar 25th–Mar 28th, Apr 8th–Apr 10th, and Apr 15th–Apr 17th),
218 where the result of airmasses passing over northern continental Europe (Figure S3). NO₃
219 concentration reduced in summer 2019 and 2020 as expected compared to spring or fall seasons
220 due to the volatility of NH₄NO₃, while SO₄ concentrations increased in summer due to enhanced
221 photochemistry. During the lockdown in spring SO₄ concentrations remained high, which was
222 associated with long-range transport.



223

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

226 BC concentrations during the spring lockdown (Mar 26th–May 31st 2020) reduced from 1.59 to
 227 0.87 $\mu g/m^3$ (-45%) compared to the pre-lockdown level in spring (Mar 1st–Mar 25th), due to the
 228 significant reduction in traffic during the first lockdown (Transport for London, 2020). It is worth
 229 noting that the BC concentration had already reduced by 13% in pre-lockdown spring (Mar 1st–
 230 Mar 25th) compared to the pre-lockdown winter. This is likely due to vehicle mileage reducing as
 231 the UK government implemented travel restrictions and advised people to work from home on
 232 Mar 16th, 2020 (Transport for London, 2020). BC increased to 1.24 $\mu g/m^3$ (+57%) after the
 233 lockdown and before the EOTHO (Jun 24th–Aug 2nd, 2020, pre-EOTHO in Figure 1) as people
 234 returned to work and travel. However, BC concentrations remained 31% lower than the pre-



235 lockdown summer (Aug 1st–Aug 31st, 2019) concentration of 1.8 $\mu\text{g}/\text{m}^3$, which suggests that the
236 traffic emissions reduced considerably as the fewer economic activities even after the ease of the
237 first lockdown (e.g., suggestions of hybrid working mode, restricted international travel, reduced
238 tourism, limited access to entertainments). BC also increased to 1.4 $\mu\text{g}/\text{m}^3$ (+10%) during the
239 EOTHO scheme (Aug 3rd–Aug 31st, 2020). This was not only because of increased traffic emission
240 during this period, but may also result from cooking activities (e.g, barbecuing or wood-fired
241 cooking styles) in central London. BC concentrations increased on Mon-Tue compared to post-
242 lockdown but before EOTHO (Jun 24th–Aug 2nd, 2020) (Figure S4).

243 3.2 *OA sources in Central London*

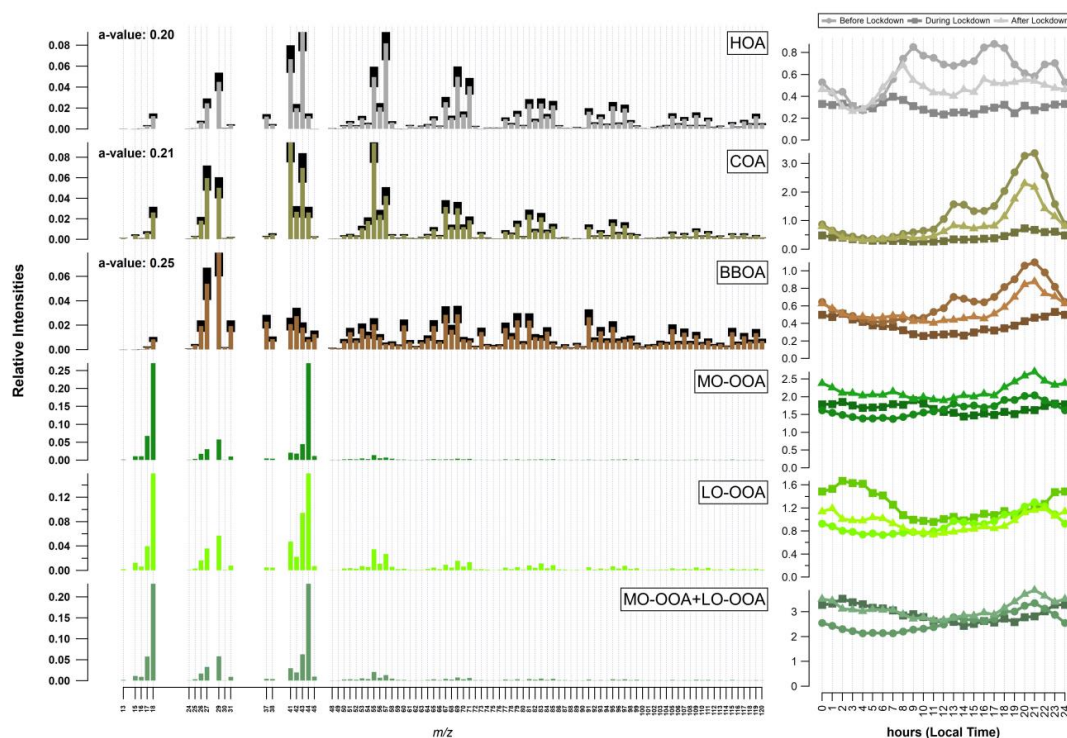
244 As mentioned above, the rolling PMF analysis resolved 5 factor solutions, including HOA, COA,
245 BBOA, MO-OOA, and LO-OOA as shown in Figure 2 and Figure 3. The left panel of Figure 2
246 shows the yearly averaged factor profiles of resolved PMF factors and total OOA calculated as the
247 sum of LO-OOA and MO-OOA. All factors show good agreements with previous studies in terms
248 of key *m/z*.

249 3.2.1 Time series of OA factors

250 Figure 2 shows both time series (30-min time resolution) and diurnal cycles for each OA factor.
251 The mean concentrations of HOA, COA, BBOA, MO-OOA, LO-OOA, and OOA were 0.50 ± 0.1
252 $\mu\text{g}/\text{m}^3$, $0.93 \pm 0.14 \mu\text{g}/\text{m}^3$, $0.55 \pm 0.11 \mu\text{g}/\text{m}^3$, $1.81 \pm 0.41 \mu\text{g}/\text{m}^3$, $1.00 \pm 0.44 \mu\text{g}/\text{m}^3$, and $2.80 \pm$
253 $0.70 \mu\text{g}/\text{m}^3$, respectively, and contributed to OA (PM_{10}) with the fractions of 11% (5% to PM_{10}), 20%
254 (9% to PM_{10}), 12% (5% to PM_{10}), 38% (17% to PM_{10}), 21% (9% to PM_{10}), and 59% (26% to PM_{10}).
255 The concentration of all OA factors shows strong time variations over the year as shown on the
256 left panel of the Figure 2. OA factors also showed strong seasonality besides the effects from



257 COVID-related policies (Figure 3). POA concentrations were generally lower in the warmer
 258 seasons than in winter as lower temperature favours particle formation via condensation and
 259 dilution and dispersion are reduced due to the lower boundary layer. The OOA factor
 260 concentrations were larger the warmer seasons due to enhanced photochemistry at higher
 261 temperature, stronger solar radiation and increased VOC emissions. The seasonality observed here
 262 in central London agreed with the other sites across Europe (Chen et al., 2022).



263

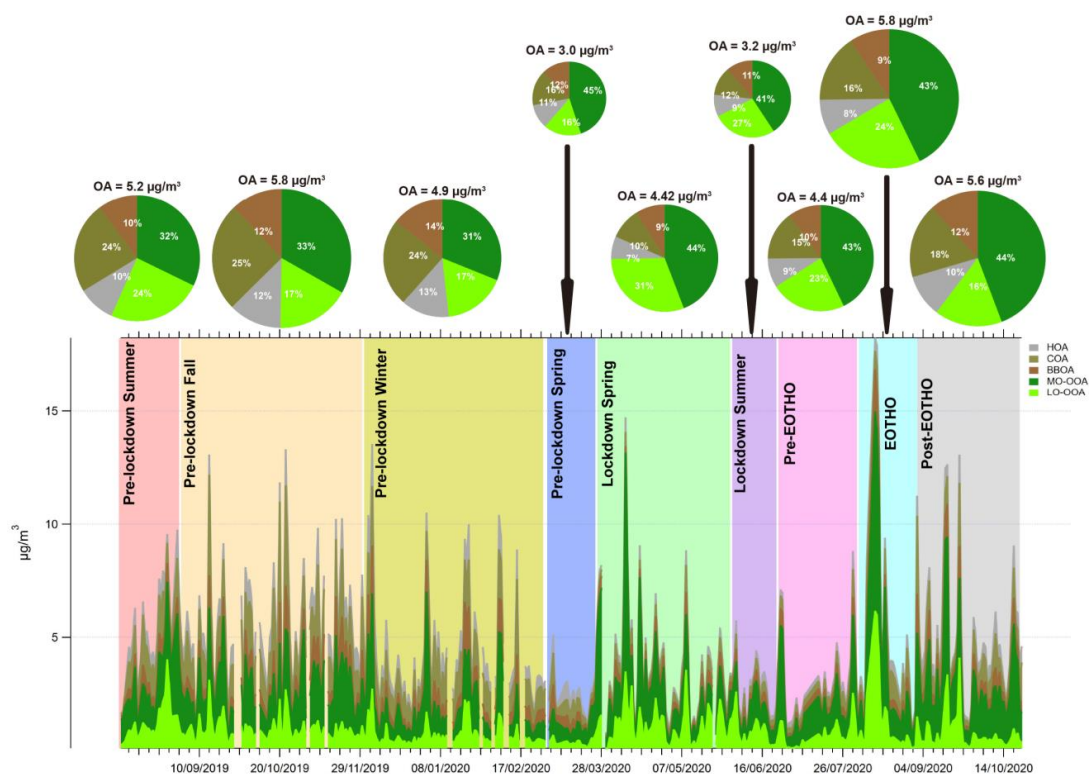
264 *Figure 2 Yearly averaged profiles (left) and diurnal cycles (right) of resolved factors from the rolling PMF analysis at the MY site.*
 265 *Time is expressed in local time.*

266 3.2.2 Diurnal Cycles for OA factors

267 The right side of Figure 2 shows the diurnal cycles before, during, and after the lockdown. POA
 268 factors showed distinct diurnal variations, in which HOA showed morning and evening rush hour
 269 peaks, COA showed distinct lunchtime and evening peaks, and BBOA showed a similar pattern



270 as COA before and after the lockdown. This indicates that the part of what is measured as BBOA
 271 in central London is most likely co-emitted from cooking activity, most likely from barbecuing
 272 style restaurants in the area. Mohr et al. (2009) showed that meat-cooking can slightly elevate m/z
 273 60, which is an important ion in the BBOA factor profile. OOA factors showed much less diurnal
 274 variation compared to POA factors in all periods, this is in agreement with the other 22 European
 275 sites reported in Chen et al. (2022). The MO-OOA showed a smaller diurnal variation compared
 276 to LO-OOA.

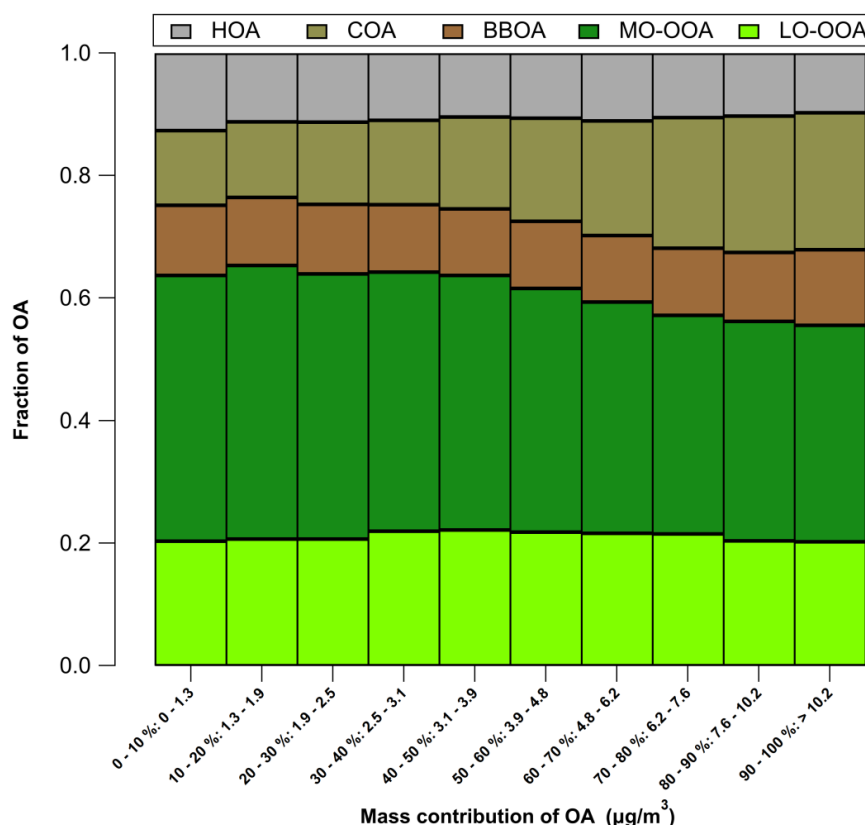


277
 278 *Figure 3 Average mass concentrations for OA sources at MY during different periods from Aug 2019 to Oct 2020*

279 The diurnal variation of COA and BBOA during lockdown lost the distinctive lunch peak as shown
 280 in the pre-lockdown; and the evening peak reduced its intensity (Figure 2). HOA retained distinct
 281 morning and evening rush hour peaks but at lower mass concentrations during lockdown (Figure



282 2). After the first lockdown, the distinct lunch and evening peaks in diurnal patterns of COA and
 283 BBOA reappeared as the open-up of nearby restaurants. The morning and evening rush hour peaks
 284 for HOA enhanced considerably as the ease of the travel restrictions after the first lockdown.
 285 However, POA concentrations did not reach pre-COVID levels. This is likely due to widespread
 286 hybrid working and the remaining oversea travel restrictions suppressing tourism, which reduced
 287 traffic activity and restaurants visits. Conversely, OOA concentrations were slightly higher than
 288 pre-lockdown levels. These were related to long-range transport, with relatively high mass
 289 concentrations of MO-OOA and LO-OOA during the lockdown (Figure S3).



290

291
292

Figure 4 Contributions to total OA from the different identified OA sources at different OA concentrations. Total OA concentrations were split in 10 equally distributed bins.



293 As shown in Figure 4 The contribution to total OA concentrations from HOA, BBOA, and LO-
294 OOA was consistent at different OA concentrations. However, the contribution of COA increased
295 as total OA concentrations increased (Figure 4), This suggests that cooking emissions in Central
296 London are responsible for elevated OA concentrations.

297 3.2.3 Impact of lockdown on OA Concentrations

298 OA concentration decreased by 34% in pre-lockdown spring compared to pre-lockdown winter
299 (Dec 1st, 2019–Feb 28th, 2020) due to seasonality and the impact of lockdown. OOA concentrations
300 were relatively unaffected with some variability before, during, and after the lockdown. due to
301 long-range transportation of airmasses from the continental Europe as observed for NH₄, NO₃, and
302 SO₄. Primary emissions were significantly lower due to reduced vehicle mileage and other
303 economic activity before the official lockdown measure came into force (Figure 3 and Figure 2
304 (a)) as suggested by the 1st quarter drop in GDP. Atmospheric components related to vehicles
305 (HOA and BC) decreased by 50% and 13% respectively, in early March 2020. COA and BBOA
306 decreased by 60% and 47% respectively. COA, due to fewer restaurant activity, BBOA likely
307 reduced partly due to the warmer weather requiring less domestic space heating, and also due to
308 reduced commercial cooking using charcoal and wood.

309 Compared to the pre-lockdown spring, HOA and COA in the lockdown spring decreased by 8%
310 and 11%, respectively, while BBOA increased marginally by 5% (from 0.37 to 0.39 $\mu\text{g}/\text{m}^3$) (Figure
311 3). MO-OOA and LO-OOA increased by 43% and 169%, respectively due to long-range
312 transportation of airmasses from continental Europe and increased photochemistry compared to
313 the first 25 days in Mar 2020. This was accompanied by increased SO₄ (+119%), NH₄ (+16%) and

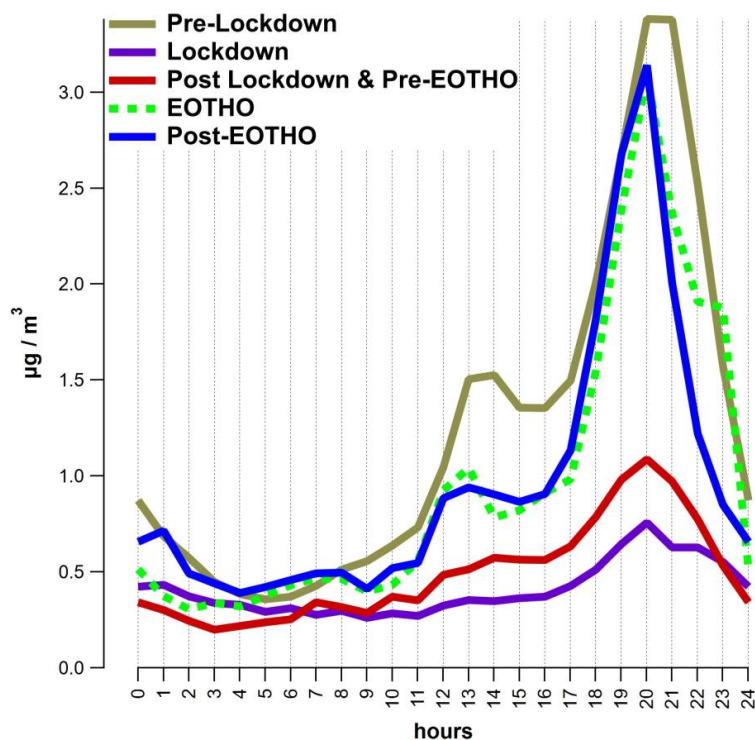


314 NO_3 (+46%), despite the higher temperature could favour partitioning these species into the gas
315 phase.

316 In June 2020, still in lockdown (Jun 1st– Jun 23rd, 2020), POA showed further but marginal
317 decreases (-4%, -8%, and -10% for HOA, COA, and BBOA, respectively, Figure 3) compared to
318 the lockdown spring as the enhanced photochemistry leads to increased formation of OOA from
319 the POA. However, the overall mass concentration of MO-OOA and LO-OOA decreased
320 significantly by 34%, and 37%, respectively as the result of fewer long-range transported airmasses.
321 During pre-EOTHO (Jun 24th–Aug 2nd, 2020), HOA, COA, and BBOA all showed considerably
322 increases of 34%, 69%, and 25%, respectively when compared to lockdown summer period. In
323 which, MO-OOA and LO-OOA also increased by 45% and 18%, respectively as the results of
324 long-range transported airmasses from continental Europe, enhanced biogenic emissions and
325 photochemistry. The POA concentrations were much lower when compared to summer 2019 (Aug
326 1st–Aug 31st, 2019) as travel and economic activities did not return to pre-COVID levels.
327 Specifically, reduced vehicle mileage resulted in lower HOA (-22%), BC (-31%), COA (-46%)
328 due to the reduced commercial cooking activity. As BBOA is co-emitted with COA during of
329 cooking activities, BBOA also decreased slightly from 0.53 to 0.44 $\mu\text{g}/\text{m}^3$ (-17%, Figure 3).

330 3.2.4 Eat out to help out (EOTHO)

331 During EOTHO (Aug 3rd–Aug 31st, 2020), MO-OOA and LO-OOA increased by 31% and 35%
332 respectively compared to post-lockdown concentrations before EOTHO and correlated with
333 increased NO_3 and SO_4 concentrations. This was due to long-ranged transported airmasses and
334 enhanced photochemistry as well as the photooxidation of POAs.



335

336

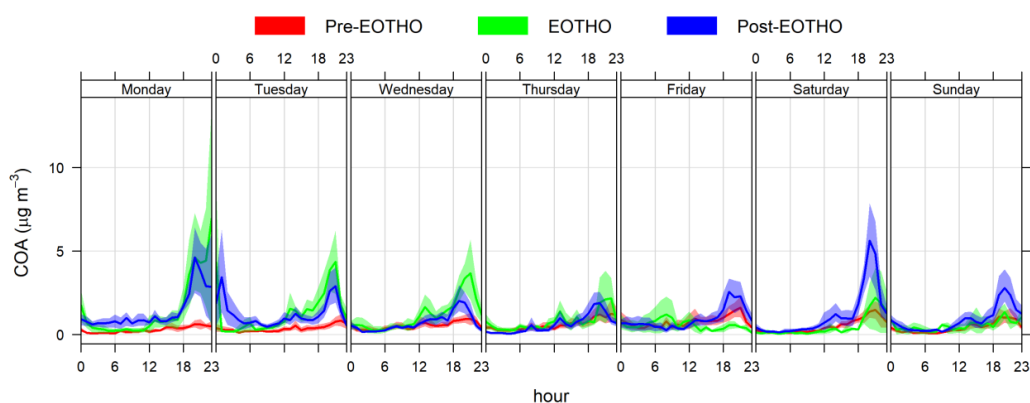
Figure 5 . COA diurnal plots at different periods in relation with COVID-related policies

337 However, EOTHO policy had a significant impact on all POA factors. In particular, the COA
338 concentration increased by 38% compared to the post-lockdown period from Jun 24th to Aug 2nd,
339 2020 (Figure 5). HOA and BBOA concentrations also increased by 22% and 23%, respectively,
340 which suggested the human activities resulting in these emissions recovered slowly after the
341 lockdown. COA was significantly higher due to EOTHO, however, it did not reach pre-COVID
342 concentrations (Figure 5). After the EOTHO policy (Sep 1st–Oct 22nd, 2020), COA concentrations
343 increased by 10% (Figure 5). This may have partially been due to lower temperatures, reduced
344 dispersion and photochemistry in Autumn.

345 EOTHO only operated from Mon to Wed, and this was clear in the diurnal plots (Figure 6) and
346 Figure S6 with larger COA concentrations Mon to Wed, in contrast with larger concentrations over
347 the weekend (Fri to Sun) before EOTHO (Jun 24th–Aug 2nd, 2020). Interestingly, even after the



348 EOTH policy ceased, COA levels remained elevated on Mon and Tue but a much higher level
 349 during the weekend was observed. This suggests that EOTH had an influence on the consumer
 350 behaviour even after the lockdown. It is also worth noting that the high concentrations of COA
 351 and BBOA (Figure S5) on Monday night were caused by the last day of EOTH policy coinciding
 352 with a UK public holiday on Aug 31st.



353

354 *Figure 6 The diurnal cycles for each day of the week in COA concentrations before, during, and after the Eat Out To Help Out*
 355 *(EOTH) policy in post-lockdown period (Jun 24th–Oct 22nd, 2020)*

356 4 Conclusion

357 This study demonstrates the importance of source apportionment studies to better understand how
 358 national and local government policies can impact the PM mixture, and how these effects can be
 359 differentiated from the influences of meteorology and large-scale atmospheric processes. PM
 360 concentrations increased at the beginning of the lockdown (Mar–Apr 2020), coinciding with
 361 reduced economic activities, however by examining the source apportionment (and inorganic PM
 362 composition) the impact of lockdown policies on primary emissions could be quantified. COVID-
 363 related policies were found to have profound but largely unintended impacts on air quality. The
 364 first lockdown significantly reduced POA sources: including HOA by 52%, COA by 67%, and



365 BBOA by 41%. While all these components reduced dramatically during the lockdown, they only
366 gradually increased again and did not reach pre-COVID levels during the duration of this study.
367 Most significantly, while the Eat Out To Help Out (EOTHO) policy was effective in helping the
368 hospitality industry to recover from economic losses during the lockdown, it had unintended
369 impacts on air quality as cooking emissions increased. Clearly detecting this change confirms the
370 presence of COA (20% to OA) as an important source of OA in London, and other cities, and the
371 importance of commercial cooking as a source. Also of note was the impact that EOTHO had on
372 BBOA concentrations, which increased by 23% while this policy was in place. This establishes a
373 clear link between commercial cooking activity and BBOA measured in cities due to the use of
374 charcoal and wood as cooking fuels, as well as potentially emissions from cooking ingredients.
375 Cooking may therefore be underestimated as a source if COA concentrations are considered in
376 isolation, and BBOA is only associated with other sources of solid fuel burning. This emphasises
377 the need to develop policies and technical solutions to mitigate commercial cooking emissions in
378 the urban environment, especially as there are limited regulations on this industry in terms of air
379 pollution. It also demonstrated the importance in continuous monitoring with subsequent source
380 apportionment analysis to better understand the influence of government policies to improve air
381 quality more effectively.

382



383 Code/Data availability

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

388 Author contribution

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392 Formal analysis, Data curation. **Max Priestman:** Methodology, Formal analysis, Data curation.
393 **Anna Font:** Writing – review & editing, Methodology, Formal analysis, Data curation. **David C.**
394 **Green:** Writing – review & editing, Supervision, Project administration, Methodology, Resources,
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396 Competing interests

397 The authors declare that they have no known competing financial interests or personal
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399

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