

1 **Insights into the sources of ultrafine particle numbers at six**  
2 **European urban sites obtained by investigating COVID–19**  
3 **lockdowns. Response to reviewers.**

4 Note: Review comments are displayed in plain text, responses to those comments are displayed in  
5 blue and sections that have been added to the text are coloured *green (and italicised)* We thank the  
6 reviewers for their insightful comments and provide responses below.

7 **Reviewer: 1**

8 **Summary:**

9 The authors applied comprehensive analysis to study the emission sources, generation mechanisms,  
10 and potential sinks of urban particulate matter (PM) through the particle number and mass  
11 concentration data from observation sites in four European cities. By comparing the PM concentration  
12 differences between the lockdown and reference periods, the authors analyzed the impact of COVID-  
13 induced lockdowns on emission intensity and atmospheric physicochemical processes, and  
14 consequently, the changes in PM concentrations. The apportionment of the sources/generation  
15 mechanisms of PM was quantified by positive matrix factorization (PMF) approach. This study found  
16 that nucleation, road traffic, and diffuse urban emissions were the dominant sources to mean PM  
17 number concentration, while formation of secondary inorganic aerosol contributes most to the PM's  
18 mass concentration. Lockdown was found to have varied impacts on the abovementioned PM  
19 sources and mechanisms.

20 **General suggestions:**

21 1. This paper studied the impacts of lockdown on PM concentration and their determinant factors.  
22 Given the lockdown starting/ending date in each country is varied, it would be better to add a figure to  
23 illustrate what principle/index you used here to determine the lockdown time periods for four  
24 countries (i.e., the proxy vehicle mobility data or other ancillary index).

25 COVID lockdowns are defined as the periods where mixing in public spaces is prohibited.  
26 Lockdowns were phased in their introductions and removals differently in each country, and in most  
27 cases were staged. We do not have reliable traffic data at each site, although we agree this would be a  
28 good metric. For each country, we chose the start as the point where legal measures were imposed  
29 (typically closures of schools), and the end where economic or social activity was reintroduced. No  
30 single metric was used, as this would lead to vastly different “lockdown” periods for each country (for  
31 example, Budapest kept schools shut longer than Germany). Using periods of different numbers of  
32 months would then lead us into problems of differing meteorology. For example, in the UK, the start  
33 is defined as when the official government lockdown was introduced, and the end as when non-  
34 essential shops opened, spurring movement of people. We include this in the below discussion

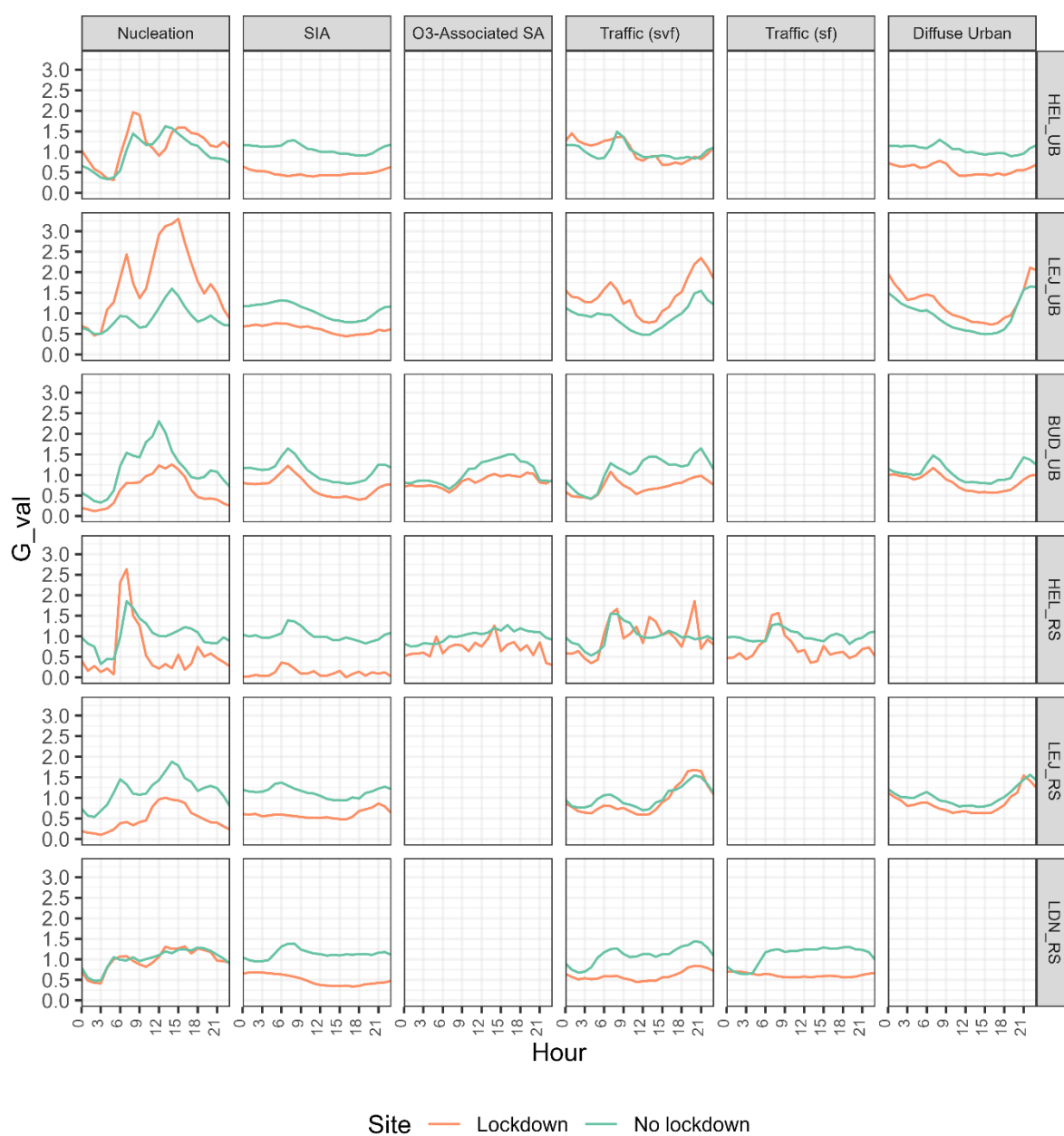
35 *“As the COVID lockdown was different in each country, we chose the start point as where a*  
36 *substantial limitation on human movement was imposed around mid-March, and the end of the period*  
37 *as when some large change was made to human movement in mid-May. Using any single metric*

38 would lead to vastly different sampling periods where differences in meteorology would dominate our  
39 results, rather than differences in emissions.”

40 2. The daily and weekly cycles of each major factor in Fig 3. represent the typical emission/formation  
41 patterns of PM. Does this pattern change during the lockdown period? A comparative figure between  
42 lockdown and reference year on different time-scale cycles may help to illustrate the shift of  
43 anthropogenic activities and its impact on PM concentration.

44 Great suggestion, thank you. We include this figure in the supplementary as Figure S5 and include the  
45 following discussion in the paper (new text in bold)

46 “The response of these factors to lockdown measures were largely consistent, and the diurnal cycle of  
47 each source-related factor is similar for both the lockdown and reference years (Figure S5)”



49 **Figure S5:** Diurnal cycle of G values from the PMF model for each factor for the reference and  
50 lockdown periods.

51 3. Do current PMF factors quantify the contributions of transported or aging non-anthropogenic PM  
52 from upwind regions? Or any of the additional factors can be added to partially explain the  
53 contribution of PM's mid-/long-range transport from non-anthropogenic emissions.

54 In an urban area, and in particular at a roadside, the influence of natural PM will be small relative to  
55 the anthropogenic. As there is no chemical speciation in the MPSS data, but simply size and number  
56 data, it is difficult to isolate factors for naturally occurring aerosols. This is easier when analysing  
57 chemical data, in particular when chemical tracers are used (Yin et al., 2015). However, as biogenic  
58 VOC concentrations are small relative to anthropogenic VOC concentrations, secondary organic  
59 aerosol is mostly dominated by AVOCs. Similarly, anthropogenic nitrogen and sulphur emissions will  
60 dominate the inorganic fractions of secondary aerosol, therefore, natural secondary aerosol likely gets  
61 identified in the PMF model as one of our secondary aerosol factors, and makes a small contribution.  
62 Similarly, natural biomass burning would be indistinguishable from anthropogenic biomass burning  
63 that is also aged, but our sampling sites are far from any natural fires and we believe the influence of  
64 aged biomass burning is low. Sea salt makes a small contribution to the PNSD, and usually requires  
65 some dedicated work to pick apart in PNSD PMF, even at coastal sites (Xu et al., 2024), and  
66 therefore, we believe sea salt is a small contributor. We have no way of quantifying what fraction of  
67 our NPF is natural vs biogenic, but model studies would indicate that most particle formation is  
68 influenced by anthropogenic emissions (Gordon et al., 2017), and particle growth is likely driven by  
69 the products of AVOC oxidation (Li et al., 2022). We acknowledge these uncertainties in the  
70 following lines when discussing secondary aerosol:

71 *“In an urban environment it is reasonable to presume that most SA precursors are anthropogenic, but  
72 an influence of natural SA precursors will also contribute some fraction of total SA.”*

73 When discussing sea salt and dust:

74 *“We do not expect a substantial contribution from dust and sea salt in the particle number size  
75 distribution at diameters <600 nm.”*

76 When discussing NPF:

77 *“NPF in these urban areas is likely driven by sulphuric acid, bases such as dimethylamine, and  
78 OOMs, likely from AVOC oxidation (Lee et al., 2019), meaning our NPF is mostly driven by  
79 anthropogenic emissions”*

80 And when discussing our diffuse urban factors:

81 *“A contribution of aged natural aerosol may also be present in this factor.”*

82 4. The deweathering/detrending technique is needed especially comparing the lockdown effects on  
83 pollutant concentration. The authors have stated that such technique has not been applied here because  
84 some of the PMF factors are associated with meteorological parameters. It would be recommended to  
85 add a similar PMF analysis on the deweathered data to exclude the effects of interannual variations on  
86 PNC or PMC. The factors such as road traffic solid fraction is less biased after conducting detrending  
87 process.

88 We agree that deweathering leads to better insights when applied to pollutant data such as NO<sub>x</sub>, CO,  
89 etc. (Shi et al., 2021). Deweathering mostly accounts for the dispersion of pollutants due to dilution  
90 (heightening of the boundary layer, dilution due to wind speeds etc.). In the case of the PNSD  
91 however, the effects of meteorology are more complex. Many PM components are semivolatile, and  
92 increases in temperature can lead to their evaporation. Similarly, high temperatures inhibit rapid NPF  
93 as they increase the evaporation rate of small clusters. However, high temperatures are often  
94 coincident with intense solar radiation, which will accelerate the formation of sulphuric acid and other  
95 NPF precursors. Emissions of SOA precursors from processes such as evaporation of VOCs from  
96 traffic vehicles is also temperature dependent (Cliff et al., 2023). We therefore argue the dynamics of  
97 the PNSD are too complex to try and apply deweathering to. For this reason, we included the  
98 following text in the manuscript, which we have updated with extra detail.

99 *“In examining temporal changes affecting pollutant concentrations, it is now common practice to*  
100 *remove the influences of changes in weather variables which affect primary pollutant concentrations*  
101 *(Vu et al., 2019; Shi et al., 2021) primarily by accounting for dilution effects. Such a treatment was*  
102 *not applied in this study as weather variables affect both photochemical nucleation, with elevated*  
103 *temperatures accelerating the evaporation rate of clusters, and intense solar radiation accelerating*  
104 *the generation of NPF precursors (Lee et al., 2019; Bousiotis et al., 2021), and the semi-volatile*  
105 *fraction of both traffic and secondary aerosols through evaporation (Charron et al., 2003). Similarly,*  
106 *emissions of organic aerosol precursors are temperature-dependent (Lee et al., 2019). The dynamics*  
107 *of the number size distribution are therefore more complex than can be accounted for by de-*  
108 *weweathering methods, and such results could not be interpreted with confidence. Rather, the trends in*  
109 *meteorological variables are presented in **Figure S8**, and commented upon in the text, where*  
110 *considered relevant.”*

111 As well as all associated discussion surrounding Figure S8

112 **Line-by-line suggestions:**

113 1. Line-107 (Table 1 title): May use the full name of ACTRiS when it first appears.

114 Great suggestion. This has been done.

115 2. Line-228 & 232 (Figure 3 & 4 title): Briefly explain how the G values used here was computed.  
116 What does the magnitude of G values stand for.

117 We now finish the titles of Figure 3 and 4 with the following:

118 *“The G value is the time-series component of the PMF solution (see supplement).”*

119 3. Line-232 (Figure 4 title): The figure indicates the normalized G values. Briefly introduce how does  
120 the normalization was conducted.

121 The title of Figure 4 now contains the following:

122 *“The normalisation is performed so each variable has a maximum value of 1 for easy comparison.”*

123 4. Figure 2 & 3: Recommend to enlarge the figure and adjust the image layout for a better  
124 visualization of the results. The shaded area in Fig. 3 is a bit difficult to distinguish.

125 We have made two edits to the figures. 1) we have made the shaded region in Fig. 3 darker for  
126 legibility; 2) we have printed all images as 600 dpi images for clarity.

127 5. Figure S7, temperature row: Is the magnitude of temperature in this figure the absolute or relative  
128 temperature

129 Thanks for pointing out the error – the units are in fact in degrees centigrade, not Kelvin. They are  
130 absolute, and we have now amended the figure.

131

## 132 **Reviewer: 2**

133 The authors applied PMF to elucidate how particle number concentrations evolve during spring  
134 2020 lockdown period relative to the reference years of 2014-2019. While the topic is overall  
135 interesting, I feel the novelty is very weak in the current version, and a substantial revision is  
136 needed to satisfy the standard of the journal.

137 1. This study heavily relies on PMF method. Is there any uncertainty? The authors seem to  
138 consider the source apportionment based on PMF as a fact, while I think there might be large  
139 uncertainties associated with this method.

140 The main uncertainty with PMF, especially as applied to particle number size distribution  
141 (PNSD) data is the attribution of factors. As we do not have chemical tracer data (Yin et al.,  
142 2015), the assignment of factors is based upon the shape of the size distribution and the diurnal  
143 and annual cycles of each factor. There is a little ambiguity here, as sometimes several sources  
144 blend into one. This is discussed in the paper extensively in section 3.1.1., where new particles  
145 from NPF and traffic blend together into a “nucleation” factor, section 3.1.2 where non-traffic  
146 combustion emissions contribute to the “traffic<sub>(svf)</sub>” factor, section 3.1.4., where “diffuse urban” is  
147 a combination of aged emissions, and section 3.1.5 plus 3.1.6 where the origin of the secondary  
148 aerosol is unknown. This is discussed extensively in section 3.1. Further, there is the problem of  
149 rotational ambiguity, which is currently not discussed in the manuscript. We now include the  
150 following in the discussion of PMF methods:

151 *“Rotational ambiguity arises in PMF as the decomposition of the data into G and F is not unique;*  
152 *there can be multiple pairs of G and F matrices that satisfy the model while producing the same*  
153 *quality of fit. However, PMF inherently limits this ambiguity through the imposition of non-negativity*  
154 *constraints on both G and F. This restriction significantly reduces the degrees of freedom for rotation,*  
155 *as it allows only transformations that maintain non-negative entries, thereby ensuring that all*  
156 *extracted factors and their contributions remain physically interpretable. Despite this, different*  
157 *rotations that meet the non-negativity conditions can still produce valid solutions, potentially leading*  
158 *to different interpretations of the source contributions and profiles. These can be investigated using*  
159 *FPEAK (Paatero and Tapper, 1994), however, in this work the initial solutions were deemed*  
160 *physically meaningful, and changing FPEAK did not improve our results.”*

161 *Paatero, P. and Tapper, U.: Positive matrix factorization: A non-negative factor model with optimal*  
162 *utilization of error estimates of data values, *Environmetrics*, 5(2), 111–126,*  
163 *doi:10.1002/env.3170050203, 1994.*

164 2. The overall comparison between lockdown and reference period seems to be very general and  
165 not surprising. It looks like many statements has been known already.

166 We argue that the new findings in this manuscript satisfy the criteria for novelty of ACP. We  
167 provide the first PMF study of particle number size distributions in Europe during COVID  
168 lockdowns, and we perform this across six sites. The smaller-than-expected reduction in traffic  
169 during COVID lockdowns points to a complex picture of pollutant reduction under traffic  
170 reduction. The increase in traffic emissions and PNCs at the LEJ\_UB site points to a possible  
171 misattribution of “traffic” factors in the literature, as stationary combustion sources may be more  
172 important than previously thought. This is only evident when looking at the data during the  
173 COVID lockdown periods, and we therefore advance the field of PNSD PMF by advocating new  
174 naming conventions. The mixed response of the nucleation factor points to the complexity of the



175 process: primary emissions both accelerate NPF through the emissions of precursors, and slow  
176 NPF through the emission of particles with a high surface area. The same applies to primary  
177 traffic nucleation particles, where their lifetimes increase as total PM surface area declines. The  
178 consistent decline of secondary aerosols is also novel, and can likely be attributed to the reduction  
179 in precursor emissions. Further to this, this study extends previous analyses (Rivas et al., 2020) to  
180 new sites.

181 3. In many sections, there are only a couple of sentences, briefly describing the difference  
182 between lockdown and reference period. These short paragraphs are not acceptable in my  
183 opinion, and they do not raise any scientific finding in depth.

184 We provide data in Tables 3 and 4, Figures 5 and 6, and in an updated Figure S5 below. We do  
185 not provide a detailed discussion of the time series of each factor, as the diurnal cycle is the same  
186 for each factor in the reference and lockdown years. We highlight this fact in the following  
187 sentence:

188 *“The response of these factors to lockdown measures were largely consistent, **and the diurnal cycle***  
189 ***of each source-related factor is similar for both the lockdown and reference years (Figure S5)”***

190 In each section from 3.2.1. to 3.2.5. we provide 2-3 paragraphs of discussion of each factor. This  
191 contains both discussion of the changes in magnitude, and speculation about the possible causes.  
192 As this review does not contain any particular suggestions for what we should be describing, we  
193 have opted to add more detail about the magnitude of change (this is included in bold in the  
194 below sections)

195 *“Among urban background sites, mean PNCs from road traffic<sub>svf</sub> factors were higher at LEJ\_UB,*  
196 *comparable at HEL\_UB, and lower at BUD\_UB during lockdown compared to the equivalent periods*  
197 *in the reference years, **with a mean change of +3.7% across the three sites (Error! Reference source***  
198 ***not found. and Error! Reference source not found.).”***

199 *“At roadside sites, mean PNCs from road traffic<sub>svf</sub> factors were comparable at HEL\_RS, and lower at*  
200 *LEJ\_RS and LDN\_RS during lockdown compared to the equivalent periods in the reference years,*  
201 ***with a mean decrease of -21% (Error! Reference source not found. and Error! Reference source***  
202 ***not found.).”***

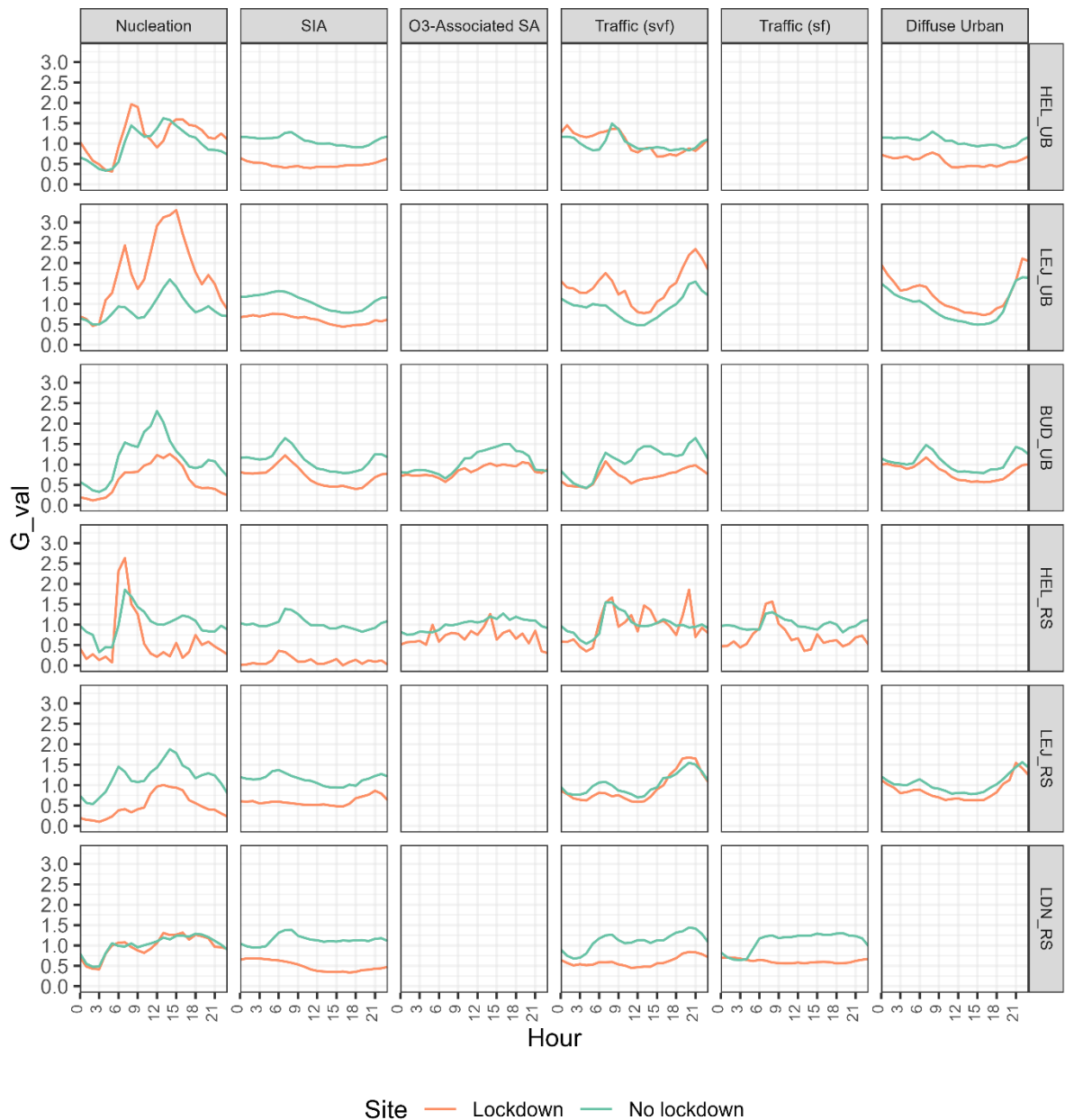
203 *“Among urban background sites, mean PNCs from diffuse urban factors were higher at LEJ\_UB, and*  
204 *lower at HEL\_UB and BUD\_UB, during lockdown compared to the equivalent periods in the*  
205 *reference years, **with a mean decrease of -11% (Error! Reference source not found. and Error!***  
206 ***Reference source not found.). Mean PNCs from the factor were also lower at LEJ\_RS during***  
207 ***lockdown by -16% (Error! Reference source not found. and Error! Reference source not found.).”***

208 *“Among urban background sites, mean PNCs from nucleation factors were higher at HEL\_UB and*  
209 *LEJ\_UB, and lower at BUD\_UB, during lockdown compared to the equivalent periods in the*  
210 *reference years **with a mean increase of 13%”***

211 *“At roadside sites, mean PNCs from nucleation factors were comparable at LDN\_RS, and lower at*  
212 *HEL\_RS and LEJ\_RS, during lockdown compared to the equivalent periods in the reference years,*  
213 ***with a mean decrease of -43%”***

214 *“Mean PNCs from O<sub>3</sub>-associated SA and SIA factors were lower at urban background and roadside*  
215 *sites during lockdown compared to the equivalent periods in the reference years, **with mean***

216 *decreases of -42% in SIA at the urban background sites, and -42% and -60% for O<sub>3</sub>-associated SA*  
 217 *and SIA at the roadside”*



218

219 **Figure S5:** Diurnal cycle of G values from the PMF model for each factor for the reference and  
 220 lockdown periods.

221 4. There are no mechanisms at all besides of the general descriptions. I feel the authors should at  
 222 least try to add some mechanism discussion, not just describe things based on PMF, and cite a  
 223 few papers without any deeper understanding of the changes in PNCs.

224 In our view, we have included extensive mechanistic discussion, and provide sufficient reasoned  
 225 speculation to explain what we observe in the data. We do not provide detailed chemical  
 226 mechanisms, but we have not measured any chemical data, and we cite appropriate literature  
 227 where there is plentiful mechanistic discussion. As this review contains no specific suggestions,



228 we have updated with some extra discussion where we deem it necessary. New text below in  
229 bold. In our discussion of traffic<sub>svf</sub>:

230 *“Lockdown restrictions had immediate and varied impacts on energy use, with increased residential*  
231 *demand due to people being confined to their homes. **This points to a systematic misattribution of***  
232 ***this factor throughout the literature, where this traffic factor contains some contribution from***  
233 ***domestic heating.”***

234 In our discussion of nucleation:

235 ***NPF is inhibited by high particle surface area, which was lower during lockdowns at all sites***  
236 ***except LEJ\_UB (Figure 6) (Du et al., 2022). Primary and delayed nucleation particles are linked to***  
237 ***the number of vehicles on the road, as well as the associated emissions technologies, which can***  
238 ***significantly impact the formation mechanisms and composition of emitted nanocluster aerosol***  
239 ***(Rönkkö et al., 2017). . The mixed response of nucleation factors at urban background sites to***  
240 ***lockdown restrictions likely represents the interplay between these complex variables.***

241 ***Across all urban background sites, the largest increase to nucleation was at LEJ\_UB, 92%***  
242 ***increase, where the greatest increase to traffic factors was also seen (51% increase to traffic<sub>svf</sub>),***  
243 ***while CS also increased (Figure 6). The largest decrease in nucleation was at BUD\_UB, 45%***  
244 ***decrease, where the greaste reduction in traffic factors was also seen (-36% decrease to traffic<sub>svf</sub>),***  
245 ***while CS decreased. Primary nanocluster aerosol will have a lifetime on the order of tens of***  
246 ***minutes, and will need to grow to 10 nm and also undergo dilution while being transported from***  
247 ***the roadside to the urban background measurement stations. Primary and delayed primary particle***  
248 ***emissions will be less important here than at the roadside, and we infer some substantial role of***  
249 ***primary gaseous traffic emissions in accelerating NPF (Brean et al., 2023).***

250 ***“The large decrease to nucleation at the roadside can be attributed either to a decrease in primary***  
251 ***nanocluster aerosol emissions, or to a reduction in NPF precursor emissions (Brean et al., 2023).”***

252 In our discussion of secondary aerosols:

253 ***“...These interactions may help to explain why changes to mean PNCs from SIA factors were***  
254 ***typically more pronounced at roadside sites. Similarly, if O<sub>3</sub>-associated SA is generated through***  
255 ***ozonolysis of VOCs, VOC concentrations will have declined substantially during lockdown periods,***  
256 ***although, O<sub>3</sub> concentrations increased during lockdown periods (Shi et al., 2021). This decline to***  
257 ***secondary aerosol is large, consistent across all sites, and will result in a substantial reduction to***  
258 ***PM mass.”***

259 5. Figure quality is very poor. Most figures are not easy to read and should be redrawn to be  
260 legible.

261 Thank you for highlighting this. We have presented all figures as 600 dpi .jpg images and these  
262 will also be uploaded as vector images with final submission.

## 263 **Minor comments**

264 1. Line 417: The insolation was markedly higher in London during the lockdown period than in  
265 previous years, but the other cities show only a small increase. Any reasons why London has  
266 much higher downward solar radiation over this period? Why not in other cities?

267 Yes. It is similarly high in HEL\_RS. The lockdown period in southern England was characterised  
268 by unusually sunny weather. We can't find any publications discussing this.

269 2. O<sub>3</sub>-associated SA and SIA: What is ozone associated SIA? It is very vague, and the readers  
270 would not know what it is. Is it secondary organic or inorganic aerosols? Is SIA also O<sub>3</sub>  
271 associated?

272 O<sub>3</sub>-associated SA has been called O<sub>3</sub>-rich SA in the previous literature (e.g., Rivas et al., 2020),  
273 which we re-name, as the latter implies the aerosol itself is rich in ozone. We have no way of  
274 knowing whether it is an organic or inorganic aerosol, as we have no measurements of aerosol  
275 chemical composition. This is a commonly found PMF factor in PNSD PMF, where secondary  
276 aerosols are highly associated with high O<sub>3</sub> concentrations. Other papers have inferred that this  
277 may be the condensation of organic ozonolysis products, but we do not want to infer too much  
278 chemistry when we have no chemical data. This is discussed in section 3.1.5. As shown in Figure  
279 4, SIA is not associated with O<sub>3</sub> except when there exists no O<sub>3</sub>-associated SA.

## 280 References

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