

1 **Response to the Reviewer 1:**

2 *General Comments*

3 *Liang et al. (manuscript) describes the experimental formation of SOA and nitrate through*
4 *chemical aging induced in an oxidation flow reactor (OFR). The authors injected combustion air*
5 *from incense burning into the OFR at high RH, which were rapidly aged by controlling UV light*
6 *and O₃ to mimic UV-aged, O₃-aged, and OH-aged scenarios. The authors used a single-particle*
7 *AMS (SPAMS) to obtain the chemical composition of the particles and the Gothenburg PAM OFR*
8 *(Go:PAM) as the reaction vessel.*

9 *The authors use the adaptive resonance theory method (ART-2a) algorithm to perform*
10 *cluster/categorization analysis with the mass spectra (Zhao et al., 2008). The authors conclude*
11 *that the OH-aged case generates more secondary nitrate than the O₃-aged case based on the*
12 *higher relative peak area (RPA). The enhanced secondary nitrate formation is attributed to higher*
13 *uptake of nitrogen-containing species.*

14 *The manuscript overall lacks quantitative information, and I am confused about the OFR*
15 *configuration. The authors provide some [NO_x] information in Figure S10 and in the text, but a*
16 *NO_x instrument is not shown in the OFR set up in Figure S1. Moreover, the flow rates entering*
17 *and exiting the OFR in Figure S1 already match, so adding a NO_x monitor would cause a flow*
18 *imbalance.*

19 *Moreover, the methodology and instrument details are lacking for replication, and additional*
20 *explanations are needed to connect the chemistry in these OFR conditions to those of the*
21 *atmosphere. For instance, the manuscript is missing the Go:PAM temperature and experiment*
22 *residence times. The generated particle number concentrations from the WCPC, the particle mass*
23 *collected on the filters for IC analysis, and the mass of incense used are not available. I do not*
24 *have a clear picture of how much aerosol entered and exited the OFR.*

25 *The experiments and discussions fall within the scope of Atmos. Chem. Phys., and the content is*
26 *topical to the atmospheric chemistry community. However, the manuscript is currently*
27 *underprepared for publication, and there are technical issues that need resolution. Given the*
28 *importance of understanding how chemical aging impacts aerosol evolution, I request the authors*
29 *perform major revisions and resubmit the manuscript.*

30

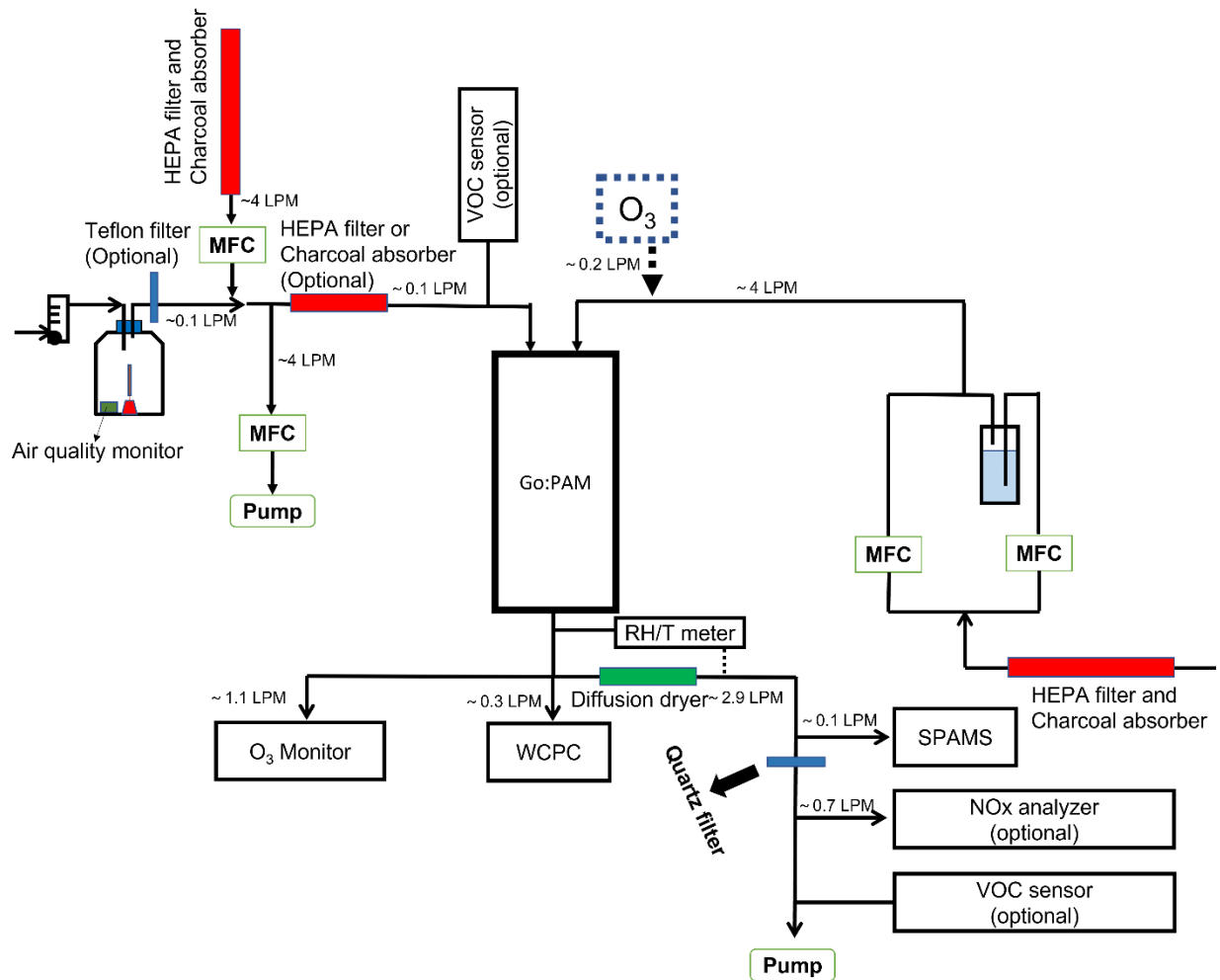
31 **Author's Response:** Thank you for the constructive comments. Many of the comments/questions
32 are addressed by the addition and clarification of experimental conditions and result discussions.
33 These comments have helped improve the manuscript and strengthen the analysis of the results,
34 but they do not affect the key conclusions in the manuscript.

35 SPAMS cannot give quantitative mass concentration information of the aerosol samples,
36 especially the organics. The Aerodyne AMS is generally accepted as quantitative. However, the
37 ionization efficiency is often assumed constant, irrespective of the chemical compositions. Hence,
38 we feel that the inability of accurate mass concentration determination is not necessarily a unique

39 shortcoming of SPAMS. Rather, it has less exposure in the scientific community to yield an
 40 accepted ionization efficiency for all its analytes. Nevertheless, it is a unique and useful tool to
 41 clearly identify the mixing states of particles, which is the emphasis of this paper. For instance, it
 42 clearly shows that nitrate preferentially formed in -ON type particles, and dicarboxylates formation
 43 in K- type particles prevailed over OC- type particles, though the detailed speciation analysis of
 44 the organic compounds is limited by fragmentation. We clarified this limitation of the technique
 45 in the manuscript.

46 We apologized for the confusion from the Go:PAM schematic (Figure S1). It has been corrected
 47 to provide additional experimental information. The characterization of the Go:PAM was reported
 48 in Watne et al., 2018 and this system has been used in a number of publications in aerosol aging
 49 (Le Breton et al., 2019; Watne et al., 2018; Wang et al., 2022; Yu et al., 2022; Zhang et al., 2020;
 50 Li et al., 2019; Liu et al., 2019; Xu et al., 2022; Zhou et al., 2021a; Zhou et al., 2021b; Tsiligiannis
 51 et al., 2019).

52



53

54 **Figure S1.** The schematic of the experimental set-up. The NO_x analyzer and the VOC sensor were
55 used only in the experiment for determining the gas removal efficiency and NO_x decay under OH
56 exposure.

57 We have also added the temperature, residence time, and WCPC particle counts to the
58 methodology description as shown below. The particles were collected on filters for IC analysis,
59 and the mass of incense used was useful to estimate the emission factor (EF) of incense burning
60 particles, although mass concentration was not the focus of this manuscript.

61 Line 77-78: The RH and the temperature at the exit of the Go:PAM were monitored by an RH and
62 temperature sensor (M170, Vaisala, Finland). All the experiments were conducted at 80 ± 0.6 %
63 RH and 22 ± 1.7 °C. The residence time in the Go:PAM was ~100 s.

64 Line 86-89: We also collected particles on 47 mm quartz filters (PALL, USA) at the exhaust of
65 the Go:PAM reactor for offline mass and chemical analysis. The number of particles collected on
66 the filters was estimated by the total WCPC counts during the sampling period. The particle
67 number concentration from the WCPC was 6100 ± 2400 # cm⁻³ and the estimated number of
68 collected particles was around 10^8 #.

69

70 *Specific Comments*

71 *1. The SPAMS calibration is not outlined, and details on the ART-2a solution is inadequate.*

72

73 *Details on the SPAMS operation would help assess the data quality. The Aerodyne soot particle*
74 *AMS requires laser alignments for consistent measurements (Avery et al., 2020); does the SPAMS*
75 *in this manuscript need a similar calibration? Particle transmission through the aerodynamic lens*
76 *is size-dependent (Huang et al., 2013). How would size-dependent particle detection influence the*
77 *data interpretation?*

78 *The absolute peak area (APA), relative peak area RPA, and number fraction (NF) are frequently*
79 *invoked in the data interpretation. The authors use APA and RPA as analogues to concentration*
80 *(or fraction of total aerosol). However, I suspect that depends on how efficiently different species*
81 *are ionized by the pulsed Nd:YAG laser, and I would like to know if adjustments have been made*
82 *to the RPA based on the ionization efficiency. What is the ionization efficiency (IE) of species*
83 *mentioned in the manuscript, and is IE consistent across species?*

84 *As for the ART-2a solution, Zhao et al. (2008) and Huang et al. (2013) note that there is no general*
85 *rule for the vigilance factor, and that a comparative approach (like re-grouping or comparing*
86 *with other clustering algorithms) may be needed. I also note there is no PAH category, despite*
87 *particulate PAH found in previous incense combustion studies (Ji et al., 2010) and a PAH*
88 *contribution being found in a similar mass spectrometer with ART-2a (Passig et al., 2022). Can*
89 *the authors provide more detail and justification for their ART-2a solution?*

90

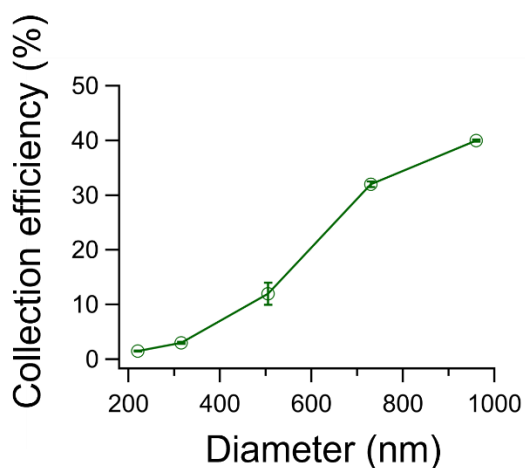
91

92 **Author's Response:**

93 The SPAMS we used operates with a principle similar to the ATOFMS, but not Aerodyne SP-
94 AMS. The laser alignment was calibrated by atomizing Polystyrene Latex (PSL) particles with
95 sizes ranging from 200-1000 nm.

96 The collection efficiency of SPAMS increased with size from ~1% at 200 nm to ~40% at 960 nm
97 (as shown below). Therefore, the secondary aerosol formation in small particles may have been
98 underestimated.

99



100

101 **Figure S13.** Relation between the particle collection efficiency of Polystyrene Latex particles and
102 diameter in SPAMS.

103 The matrix effect makes the determination of ionization efficiency (IE) difficult for SPAMS,
104 which is also an issue in other single-particle mass spectrometers (e.g., A-ToF-MS, LAAP-MS).
105 Therefore, unlike the aerodyne AMS, SPAMS is not a quantitative instrument, but is unique for
106 single-particle mixing state characterization, which is the focus of this work. One should note that
107 the Aerodyne AMS is not free from matrix effect either and hence offline chemical composition
108 measurements were usually conducted for comparison. APA and RPA were used as semi-
109 quantitative indicators of the abundance and concentration of chemical species (Zhou et al., 2021b;
110 Cheng et al., 2018). We have changed the title to ‘Secondary Aerosol Formation in Incense
111 Burning Particles by Ozonolysis and Photochemical Oxidation via Single Particle Mixing State
112 Analysis’ to emphasize the focus of this paper.

113

114 There are various algorithms for clustering single particles, but all are based on spectral similarities
115 (Zhao et al., 2008; Rebotier et al., 2007). ART-2a is the sole algorithm incorporated into the
116 commercial SPAMS system, and it is also the most common one for analyzing single-particle data.
117 Although there is no general rule for the vigilance factor in ART-2a, Zhao et al. (2008) reported
118 that both a relatively small vigilance factor (e.g., 0.5 or 0.6) and a relatively high vigilance factor

119 (e.g., 0.8) show very similar clustering accuracies ($\pm 5\%$). Nevertheless, we have added the
120 vigilance factor and further clarifications to the manuscript:

121 Line 226-229: The collection efficiency of SPAMS increased from ~1% at 200 nm to ~40% at 960
122 nm (Figure S13). Therefore, secondary aerosol formation in small particles may have been
123 underestimated. However, this would not affect the conclusion of the results that nitrate formed
124 in incense burning particles upon O₃+Dark and O₃+UV aging.

125 Line 113-116: In the ART-2a analysis, we used a vigilance factor of 0.85, and more than 98% of
126 the particles were analyzed. Note that there is no general rule for the vigilance factor in ART-2a.
127 Zhao et al. (2008) reported that both a small vigilance factor (e.g., 0.5 or 0.6) and a relatively high
128 vigilance factor (e.g., 0.8) show very similar clustering accuracies ($\pm \sim 5\%$) (Zhao et al., 2008).

129

130 We have searched for the signals of particulate PAHs. Passig et al. (2022) identified PAH-
131 containing particles by the presence of at least four peaks on the m/z channels 178, 189 (fragment
132 of alkylated phenanthrenes), 202, 220, 228, and 252. We did not find such peaks in any of our
133 samples, although they were detected in an AMS study (Ji et al., 2010). We have added this
134 information to the manuscript:

135 Line 126-130: Despite that particulate polyaromatic hydrocarbons (PAHs) were found in a
136 previous incense combustion study (Ji et al., 2010) and a recent study of ambient particles based
137 on a single-particle mass spectrometer with ART-2a (Passig et al., 2022), none of the fresh incense
138 burning particles in our experiments contained the PAHs peaks (m/z = 178, 189 (fragment of
139 alkylated phenanthrenes), 202, 220, 228, and 252). Regardless of the presence of PAHs or not, our
140 conclusion on nitrate formation does not depend on the detection of specific chemicals such as
141 PAHs.

142

143 *2. OFR characterization and operation details are missing.*

144 *Offline OHexp calibrations may be inaccurate when OH reactive species suppressed OH.*
145 *Basically, OH suppression is when the external OH reactivity (extOHR) entering the OFR is high*
146 *enough to titrate the OH, which results in OH-aging being suppressed. In such scenarios, offline*
147 *OHexp calibrations become unreliable, possibly by orders of magnitude (see Section 3.1.4 of Peng*
148 *and Jimenez, 2020). Peng and Jimenez (2020) also notes that OFR254 is susceptible to OH*
149 *suppression at low O₃ injections.*

150 *Operational information of the OFR would be valuable for replication and should be mentioned*
151 *in the supplementary. Watne (2018) describes the Go:PAM as being made of quartz; have there*
152 *been efforts to constrain electrostatic particle wall loss (Cao et al, 2020)? How would gas wall*
153 *loss (Palm et al., 2016) affect the results reported, or is gas wall loss negligible? What cleaning*
154 *procedure was taken to minimize carryover effects between experiments?*

155

156 **Author's Response:**

157 Watne et al. suggested that the penetration of the particles is close to 100% for particles larger than
158 100 nm. Hence the wall loss is negligible for the 0.2-2 um particles that SPAMS measures. Besides,
159 a control experiment measuring the total VOCs at the entrance and the exhaust of the Go:PAM
160 suggested that the gas wall loss was also minor ($6 \pm 4\%$). We have added this analysis to the
161 manuscript:

162 Line 80-83: Watne et al. suggested that the penetration of the particles is close to 100% for particles
163 larger than 100 nm. Hence the wall loss is negligible for the 0.2-2 um particles that SPAMS
164 measures. Besides, a control experiment measuring the total VOCs at the entrance and the exhaust
165 of the Go:PAM suggested that the gas wall loss was also minor ($6 \pm 4\%$).

166 Also, we have added details on the OHexp calibration with SO₂. The estimation of extOHR
167 requires VOC analysis and is not available in our study. We have also added this limitation to the
168 text.

169 **Text S1.** Estimation of the OH exposure.

170 SO₂ was used to calculate the OH exposure in the Go:PAM. The UVC lamps were turned on to
171 warm up for ~30 min and turned off. Then, O₃ (300, 800, 1500 ppb) and SO₂ (~200 ppb) were
172 introduced to the Go:PAM with the UVC lamps turned off until its initial concentration remained
173 constant at steady-state conditions, which typically took around 5 min. The [SO₂] was recorded as
174 [SO₂]_{Initial}. After that, the UVC lamps were turned on until the final [SO₂] stabilized and was
175 recorded as [SO₂]_{Final}. The time scale for the stabilization of [SO₂] was around 4 min. The OH
176 exposure at each condition is calculated using Eq. (A1):

177
$$OH\ exposure = \frac{1}{k_{OH,SO_2}} \times -\ln\left(\frac{[SO_2]_{Final}}{[SO_2]_{Initial}}\right) \quad (A1)$$

178 where $k_{OH,SO_2} = 9 \times 10^{-13} \text{ cm}^3 \text{ molec}^{-1}$ is the bimolecular rate constant between OH and SO₂ (Davis
179 et al., 1979). The equation above is the result of integrating the differential rate equation for SO₂
180 and assuming pseudo-first order kinetics. The estimation of external OH reactivity (i.e., the OH
181 reactivity with VOCs) requires VOC analysis and is not available in our study. Therefore, the OH
182 exposure shown in this study may have been underestimated.

183

184 *3. Kinetic modeling may be needed for interpretation.*

185 *The authors' argument on secondary nitrate formation, either heterogeneously or in the gas phase*
186 *is limited by the lack of quantification HNO₃, HONO, NO_x, NO_y etc. The difference in condensed*
187 *nitrate between the O₃ and OH-aged cases may be due to differences in HNO₃/HONO/NO_x uptake*
188 *as the authors allege. A kinetic calculation showing that the formation of these species under the*
189 *difference OFR conditions are comparable would be more demonstrative.*

190 *Moreover, gas-phase organic nitrate formation, either through VOC+NO₃ or RO₂+NO (Ziemann*
191 *and Atkinson, 2012) and condensation should be considered. Kinetic modeling may be needed to*

192 *connect the experimental aging conditions in the Go:PAM to those of the atmosphere (Peng and*
193 *Jimenez, 2020).*

194 **Author's Response:** Kinetic modeling will be useful in quantitatively evaluating the multiphase
195 kinetics during aging incense burning particles. However, this is beyond the scope of this
196 manuscript. Hence our data interpretation focuses on the formation of nitrate via mixing state
197 analysis of the resulted particles, but not on the detailed mechanisms. It will require another study
198 to conduct detailed modeling with new experimental data. We understand the limitation of this
199 study that the SPAMS is not quantitative and hence have not attempted to do kinetic modeling.

200

201 *Line 30: I recall incense burning is found in other cultures and am unsure if the practice is*
202 *“especially” common in Asian and African religious rituals. I suggest either providing a reference*
203 *for that point or removing “especially” in this sentence.*

204 **Author's Response:** Agree, but the use of “especially” has not excluded other possibilities.

205

206 *Line 35: The incense burning references cited here mention that there is variation in the particle*
207 *emission factor (EF) across incense varieties. How does the particle EF in these experiments*
208 *compare with those previous works? Were the combustion conditions comparable to those*
209 *previous works?*

210 **Author's Response:** The EF of the incense burning particles can vary by the type of incense and
211 burning conditions. The theme of this paper is the aging of incense-burning particles and the
212 formation of the secondary components of particulates, but not the particle EF. Again, our
213 conclusion of nitrate formation in single incense burning particles is not affected by the EF. We
214 have added additional information for the burning conditions:

215 *Line 62-64: The air exchange rate per hour (ACH) is 0.3, comparable to the typical natural*
216 *ventilation conditions (Lee et al., 2004). The relative humidity (RH) and the temperature inside*
217 *the burning bottle were 56 ± 9 % RH and 22 ± 2.7 °C.*

218

219 *Line 62: There is no information on the incense sticks used, like the manufacturer or composition,*
220 *and Liang et al. (2022) used several as shown in their Figure S21. What type of incense was used*
221 *here? How much incense was burned? This information could be valuable for replication studies.*

222 **Author's Response:** We added additional information on the incense sticks to the manuscript:

223 *Line 61-62: In brief, we burnt an incense stick (Figure S2, Kwok Tin Heung, Hong Kong) in a 20*
224 *L glass burning bottle for each experiment.*



225

226 **Figure S2.** The appearance of the incense sticks.

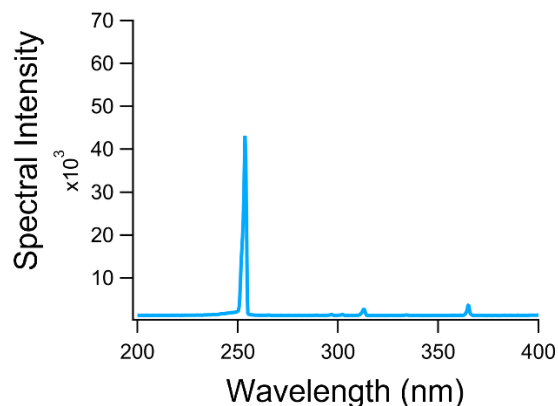
227

228 *Line 64: The methods reference (Liang et al., 2022) states there were four UVA lamps, while here*
229 *the authors say they used “two UVC light tubes.” Please confirm that the Go-PAM set up had*
230 *changed for this manuscript and specify that in the text. Moreover, what lamps were used? Rowe*
231 *et al. (2020) found that 185 and 254 nm photon fluxes would vary across manufacturers, which*
232 *may be re*

233 **Author’s Response:** We apologize for the confusion of referring our previous publication for the
234 schematic. We have now removed the reference to our previous work. We have added detailed
235 information on the UVC lamps used in this study including the spectra of the lamp to the
236 supplementary information.

237 *Line 61: The schematic of the experimental set-up is in Figure S1 and our previous publication*
238 *(Liang et al., 2022).*

239 *Line 65-67: Compressed air (~0.1 L min⁻¹) was used to introduce the diluted incense burning*
240 *particles to the Go:PAM reactor equipped with two UVC light tubes (30W, Philips TUV, $\lambda_{max} =$*
241 *254nm). The spectrum of the lamp was shown in Supplementary information (Figure S3).*



242

243 **Figure S3.** The emission spectrum of the UVC lamps.

244

245 *Line 65: I am confused on how many experiments this manuscript is describing. I see in Figure 2*
246 *that there were 7 involving aging and 1 fresh; were some of these the “control” experiments?*
247 *From this sentence I expect at least 2 types of controls, with either a charcoal absorber or HEPA*
248 *filter. Were the control cases then also aged?*

249 **Author’s Response:** The data from the control experiments are available in the supplementary
250 information, but not Figure 2. These control experiments are aged experiments. We have further
251 clarified the availability of these data.

252 *Line 67-70: In the control experiments, a charcoal absorber or HEPA filter was used to remove*
253 *the gaseous pollutants or particles prior to the introduction to the Go:PAM. The data of these*
254 *control experiments can be found in the supplementary information (Figure S4, S5). All these*
255 *control experiments were aged experiments.*

256

257 *Line 66: How did the authors obtain these removal efficiencies?*

258 **Author’s Response:** We have added the explanation for the removal efficiencies evaluation:

259 *Line 70-73: The removal efficiency of NO_x, VOCs, and particles were ~85%, ~90%, and ~100%,*
260 *respectively, determined by the concentration reduction after applying a HEPA filter or charcoal*
261 *absorber at the exhaust of the burning bottle, using a NO_x analyzer (T200, Teledyne) or a Total*
262 *VOC analyzer (Yuante) (Figure S1).*

263

264 *Line 67: Compressed air or zero air? If the air is coming from a compressor, were efforts made*
265 *to scrub the air of contaminants?*

266 **Author’s Response:** The compressed air was treated by a HEPA filter and a charcoal absorber
267 prior to the experiment system. We have clarified this in the manuscript and the schematic figure.

268 *Line 72-75: A controlled dry-wet mixed carrier flow of compressed air (~4 L min⁻¹) and a flow of*
269 *O₃ (~0.1 L min⁻¹) generated by passing O₂ (99.995%, Linde) to an O₃ generator (Model 610, Jelight*
270 *Company Inc, USA) were introduced into the Go:PAM. The compressed air was treated by a*
271 *HEPA filter and a charcoal absorber prior to the experiment system.*

272

273 *Line 74: The methods reference (Liang et al., 2022) does not mention using a diffusion dryer. At*
274 *~0.1 LPM, what was the residence time in the dryer and is there an estimate of particle loss in the*
275 *dryer? Was the dryer effective in removing H₂O?*

276 **Author's Response:** The RH at the exhaust of the diffusion dryer was ~15%. The residence time
277 of the particles in the dryer was estimated to be 5 s and the particle loss was ~4% according to the
278 CPC measurements. We have added this additional information to the manuscript.

279 Line 83-86: The particles passed through a diffusion dryer before entering the Go:PAM to reduce
280 the matrix effects from water (Neubauer et al., 1998). The RH at the exhaust of the diffusion dryer
281 was ~15%. The residence time of the particles in the dryer was estimated to be 5 s and the particle
282 loss was ~4% according to the CPC measurements.

283

284 *Line 76: What were the estimated number of particles collected?*

285 **Author's Response:** The estimated number of the particle collected has been added.

286 Line 86-89: We also collected particles on 47 mm quartz filters (PALL, USA) at the exhaust of
287 the Go:PAM reactor for offline mass and chemical analysis. The number of particles collected on
288 the filters was estimated by the total WCPC counts during the sampling period. The particle
289 number concentration from the WCPC was $6100 \pm 2400 \text{ # cm}^{-3}$ and the estimated number of
290 collected particles was around 10^8 # .

291

292 *Line 83: Please provide additional details on the OHexp calibration with SO₂, in particular the*
293 *concentrations of SO₂ used and timescales to reach equilibrium. An estimate of extOHR during*
294 *the experiments should be compared with that of the SO₂ calibrations.*

295 **Author's Response:** We have added details on the OHexp calibration with SO₂. The estimation
296 of extOHR requires VOC analysis and is not available in our study. We have added this limitation
297 to the text. As discussed earlier (Line 169-182 in this file), the lack of the extOHR does not affect
298 our conclusion that O₃+UVaging by OH leads to more significant nitrate formation than aging by
299 O₃ alone in incense burning particles.

300

301 *Line 101: See specific comment 1.*

302 **Author's Response:** Please find our response above.

303

304 *Line 113: Explaining the abbreviations would improve readability. For instance, OC and ONEC*
305 *do not appear prior in the text?*

306 **Author's Response:** The explanation of the abbreviations has been added to the manuscript.

307 Line 132-135: ART-2a categorizes fresh incense burning particles into K-ON, K-ONEC, K-Cl,
308 and OC-ON. EC, Cl and OC are abbreviations of elemental carbon, chloride and organic carbon,
309 respectively. Briefly, the "K" and "OC" before the hyphen indicate the characteristics of the

310 positive spectra, while "ON", "ONEC" and "Cl" after the hyphen indicate the characteristics of the
311 negative spectra.

312

313 *Line 147-149: How does the charcoal absorber remove NO_x without removing VOC? How would*
314 *the removal of VOC affect the interpretation here?*

315 **Author's Response:** The charcoal absorber removes both NO_x and VOC. However, the VOC
316 contents were not expected to be important in the observed nitrate RPA reduction, as the amount
317 of nitrogen-containing VOCs was minor in the incense burning plume, according to the literature
318 (Manoukian et al., 2013).

319 *Line 166-170: However, control experiments using a charcoal absorber to remove NO_x only show*
320 *~20% decrease in RPA of total nitrate in O₃ aged particles (Figure S4), indicating that NO₂*
321 *hydrolysis and nitration may not be the main contributor to the nitrate formation. The charcoal*
322 *absorber removes both NO_x and VOC. However, it was not expected to be important in the*
323 *observed nitrate RPA reduction, as the content of nitrogen-containing VOCs was minor in the*
324 *incense burning plume, according to the literature (Manoukian et al., 2013).*

325

326 *Line 156: How would the loss of SVOC/LVOC in the HEPA filter (Shilling, 1997) affect the*
327 *conclusion of the control experiment?*

328 **Author's Response:** The addition of the charcoal to remove VOCs at the exhaust only caused ~6 %
329 variation of the NF of -ON type particles and nitrate-containing particles (i.e., -ONN, -N). Hence,
330 we considered the loss of SVOC/LVOC unimportant to our conclusion. We have added this to the
331 manuscript:

332 *Line 177-183: A control experiment with a HEPA filter before the Go:PAM showed no detectable*
333 *particles by SPAMS. Thus, we assume the total SPAMS-detectable particle number was constant*
334 *before and after aging, and O₃-aging may have preferentially converted some -ON type particles*
335 *to nitrate-containing particles (i.e., -ONN, -N). It has been reported that the HEPA filter would*
336 *cause the loss of semi-volatile VOC (SVOC) or less-volatile VOC (LVOC) (Schilling, 1997).*
337 *However, the addition of the charcoal to remove VOCs at the exhaust only caused ~6 % reduction*
338 *of the NF of -ON type particles and nitrate-containing particles (i.e., -ONN, -N), suggesting the*
339 *roles of SVOC and LVOC were minor to our conclusion.*

340

341 *Line 182: How did the authors arrive at the “~90 % reduction of [NO_x]”? Was this a separate*
342 *test? If so, please add a quick summary of how that test was performed.*

343 **Author's Response:** Yes, it is a separate test. We have clarified in the caption of the schematic.

344 **Figure S1.** The schematic of the experimental set-up. The NO_x analyzer and the VOC sensor were
345 only used in the experiment determining the gas removal efficiency and NO_x decay under OH
346 exposure.

347 *Line 183: Was a NO_x monitor available? If so, please provide the monitor's location in Figure S1*
348 *and specify in the text. Also, please explain how the flow rates entering and exiting the Go:PAM*
349 *would be reconciled.*

350 **Author's Response:** We have revised the experimental schematic.

351
352 *Line 191-193: Do OH and O₃ oxidation form similar functional groups? Are those functionalities*
353 *evenly represented in these general markers? The RPA increase of SOA markers in OH over O₃-*
354 *aging may be skewed if these markers overrepresent one oxidation case over the other.*

355 **Author's Response:** These marker fragments may not be directly related to the functional groups.
356 Rather, they are just an indicator of the abundance of oxygen in the particulate matters. We have
357 clarified this in the manuscript.

358 *Line 214-217: Using the common general markers of oxidized/aged organics in single-particle*
359 *mass spectrometric studies of -16[O], -17[OH], +42[C₂H₂O], and +43[C₂H₃O] as examples (Taiwo*
360 *et al., 2014; Denkenberger et al., 2007; Qin et al., 2006), their RPA increase in O₃-UV-aged*
361 *particles are 18, 10, 3, and 17 times higher than in O₃-aged particles. This significant enhancement*
362 *of RPA suggests that OH aging produced more oxidized and functionalized products than O₃ aging.*

363
364 *Line 214: Do larger and smaller particles have similar surface properties, at least with regards*
365 *to nitrate uptake?*

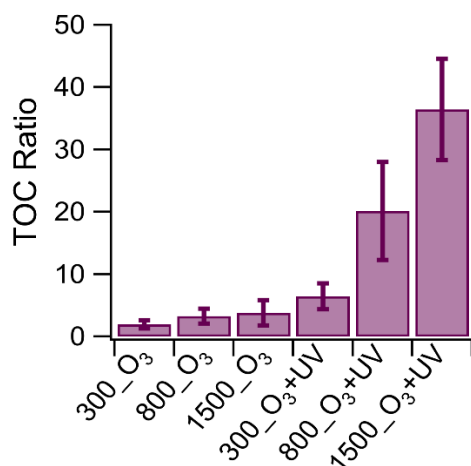
366 **Author's Response:** It has been reported that larger organic and inorganic mixed particles could
367 undergo liquid-liquid phase separation (LLPS) more easily than smaller ones (Kucinski et al.,
368 2019). The organics would mainly locate at the outer layers of the particles, whereas the inorganic
369 components reside as a core. The hydrophobic organic shell may have retarded the uptake of
370 HNO₃/HNO₂/NO_x to form nitrate. Profiling the surface composition and properties of the particles
371 is beyond the scope of this paper. We have added this possibility to the manuscript for future works.

372 *Line 241-247: However, the RPA shows an opposite trend, which can be interpreted as lower*
373 *nitrate concentration in larger particles. Larger particles have larger surfaces but smaller surface-*
374 *to-volume ratios, which lead to a larger absolute amount of nitrate formed but a lower relative*
375 *concentration of particulate nitrate (Figure 2c, e). It has been reported that the larger organic and*
376 *inorganic mixed particles could undergo liquid-liquid phase separation (LLPS) more easily than*
377 *smaller ones (Kucinski et al., 2019). It is possible that the organics are mainly located at the outer*
378 *layers of the particles, whereas the inorganic components reside in the core. The hydrophobic*
379 *organic shell may have retarded the uptake of HNO₃/HNO₂/NO_x to form nitrate.*

380
381 *Line 222: I suspect SOA formation is not "potential," but rather inevitable under the*
382 *aforementioned OFR conditions, so I suggest removing "potential" from the section heading. Also,*
383 *are oxalate and malonate universal and proportional indicators of SOA? That is, do different SOA*

384 precursors form these indicators evenly under different oxidants (O₃/OH)? I am concerned that
385 there are specific chemical conditions where these species are enhanced without a proportional
386 enhancement of SOA, which may skew the NF.

387 **Author's Response:** We have spent a lot of time interpreting the question. It is likely the SOA
388 characteristics and precursors are different under O₃ and OH. Hence, we are conservative and use
389 the term “potential”, even though oxalate and malonate are commonly found highly oxidized SOA.
390 The total organic content (TOC) ratio of aged to fresh particles extract, which indicates the
391 formation of SOA, was higher upon OH oxidation than O₃ oxidation (Figure S16). We have added
392 clarifications to the manuscript:



393
394 **Figure S16.** The total organic content (TOC) ratios of aged to fresh particles extract.

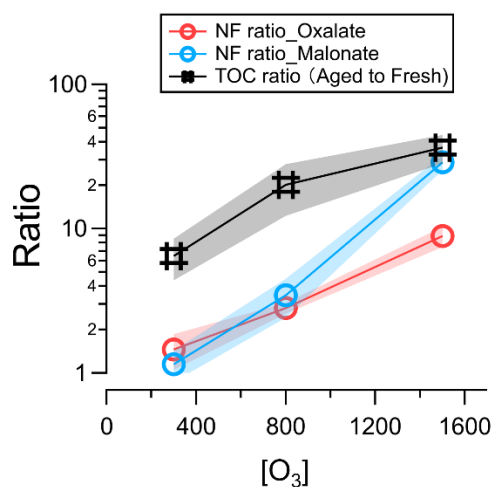
395 Line 89-92: The filter sample was extracted by deionized (DI) water for analyzing water-soluble
396 ions (e.g., nitrate, formate, potassium) by Ion chromatography (IC) using the same protocol
397 reported in our previous work (Liang et al., 2022). The total organic content (TOC) of the water-
398 extract was analyzed by a TOC analyzer (Shimadzu TOC-L).

399 Line 268-271: In contrast, no oxalate and malonate were observed during ozonolysis, irrespective
400 of [O₃]. It is likely the SOA characteristics and precursors are different under O₃ and OH. Hence,
401 we are conservative and use the term “potential”, even though oxalate and malonate are commonly
402 found highly oxidized SOA. The total organic content (TOC) ratio of aged to fresh particles extract
403 was higher upon OH oxidation than O₃ oxidation (Figure S16).

404
405 Line 229: Is NF of oxalate and malonate proportional with SOA concentrations? As it stands, “30
406 and 9 folds” increases of these tracers sounds like SOA increased by that much.

407 **Author's Response:** While the TOC ratio at different O₃+UV, which indicates the formation of
408 SOA, shows an overall trend similar to the NF ratios of oxalate and malonate (Figure S15), the
409 TOC ratio was 1.2-7.1 folds higher than the NF ratios of oxalate and malonate. However, this
410 could be due to the formation of many other species as well as the matrix effects. We hesitate to

411 speculate too much here. The SOA formation may have been underestimated due to the matrix
412 effect. SOA formation during aging of incense burning plume should be further assessed by other
413 quantitative online instruments in future works. We have added this clarification to the manuscript:



414
415 **Figure S15.** The NF ratios (aged to fresh) of oxalate and malonate, and TOC ratios as a function
416 of [O₃] in O₃+UV aging experiments.

417
418 Line 261-268: Compared to 300 ppb O₃ and UV, the NF ratios of malonate and oxalate were 30
419 and 9 folds higher, at 1500 ppb O₃ and UV, respectively. This trend is different from the
420 independence of nitrate formation on OH exposure, probably because the formation of SOA was
421 slower than nitrate via multiphase uptake. These NFs are lower estimates due to the possible
422 degradation by photolysis of Fe-decarboxylate complexes to CO₂ (Gen et al., 2021). While the
423 TOC ratio at different O₃+UV, which indicates the formation of SOA, shows an overall trend
424 similar to the NF ratios of oxalate and malonate (Figure S15), the TOC ratio was 1.2-7.1 folds
425 higher than the NF ratios of oxalate and malonate. However, this could be due to the formation of
426 many other species as well as the matrix effects. SOA formation during aging of incense burning
427 plume should be further assessed by other quantitative online instruments in future work.

428
429 *Technical Comments*

430 *Line 47: Awkward grammar in "For instance ... nitrate."*

431 **Author's Response:** We have revised the sentence as below:

432 Line 48-49: For instance, ozone and OH oxidations of NO_x by ozone and OH radicals were
433 considered primary sources of particulate nitrate (Seinfeld et al., 2008; Liang et al., 2021; Gen et
434 al., 2022).

435

436 *Line 325: Please check the citation styles; they are inconsistent.*

437 **Author's Response:** We have updated the citation styles according to the Endnote tool provided
438 by the website.

439

440 *Lines 409/412: Same reference cited twice?*

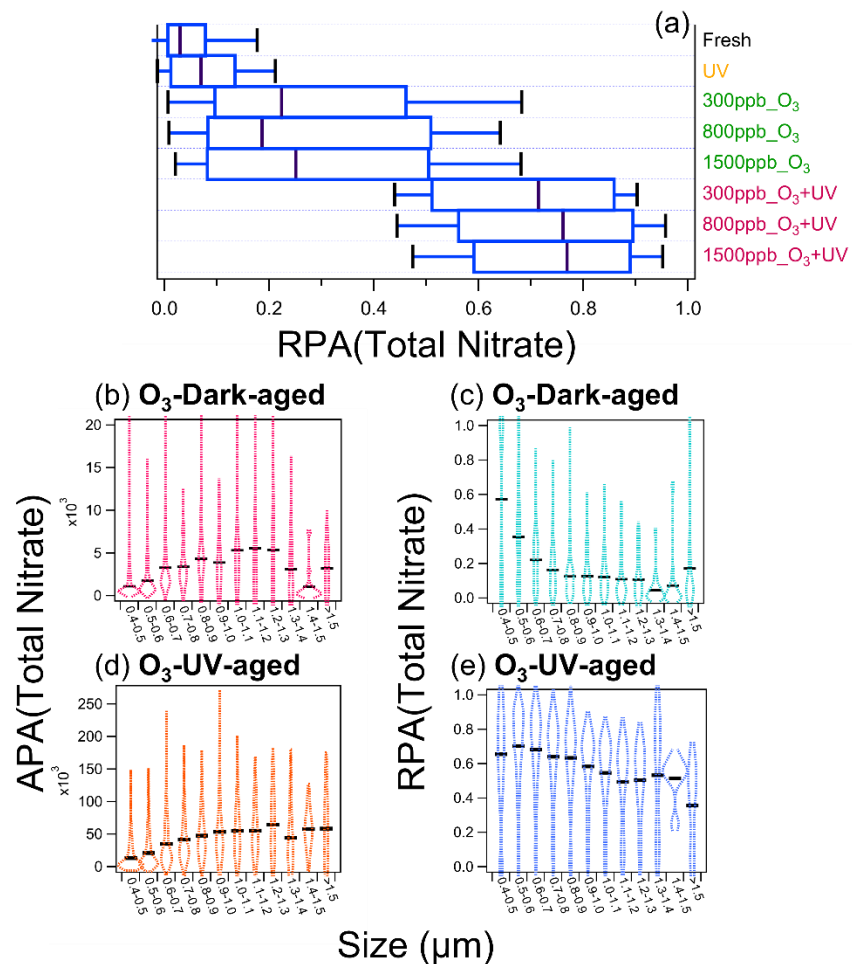
441 **Author's Response:** We have removed one of it.

442

443 *Figure 2: The data appears almost randomly scattered in the lower panels, which may be due to*
444 *points heavily overlapping with each other. The authors may want to replace the box and*
445 *whiskers/scatterplot with a violin plot for easier visualization.*

446 **Author's Response:** Thank you for the suggestions, we have replaced the box-whisker plot with
447 a violin plot.

448



449

450 **Figure 2.** (a) The whisker-box plot of total nitrate RPA of fresh and aged particles. The violin
451 plots of (b, d) APA and (c, e) RPA of total nitrate in O₃- and OH-aged particles as a function of
452 size (unit: μm) of particles aged at 1500 ppb O₃ (+ UV). The medians are shown as the lines, and
453 the kernel densities represent the probability density of the data at different values.

454

455 *Figure S1: Please display where the charcoal absorber, HEPA filter, and NO_x instrument would*
456 *have been placed.*

457 **Author's Response:** The Figure S1 has been updated.

458

459 **References**

460 Cheng, C., Huang, Z., Chan, C. K., Chu, Y., Li, M., Zhang, T., Ou, Y., Chen, D., Cheng, P., and
461 Li, L.: Characteristics and mixing state of amine-containing particles at a rural site in the Pearl
462 River Delta, China, *Atmospheric Chemistry and Physics*, 18, 9147-9159, 2018.

463 Davis, D., Ravishankara, A., and Fischer, S.: SO₂ oxidation via the hydroxyl radical: atmospheric
464 fate of HSO_x radicals, *Geophysical Research Letters*, 6, 113-116, 1979.

465 Denkenberger, K. A., Moffet, R. C., Holecek, J. C., Rebotier, T. P., and Prather, K. A.: Real-time,
466 single-particle measurements of oligomers in aged ambient aerosol particles, *Environmental*
467 *Science & Technology*, 41, 5439-5446, 2007.

468 Gen, M., Zhang, R., and Chan, C. K.: Nitrite/Nitrous Acid Generation from the Reaction of Nitrate
469 and Fe(II) Promoted by Photolysis of Iron–Organic Complexes, *Environmental Science &*
470 *Technology*, 55, 15715-15723, 10.1021/acs.est.1c05641, 2021.

471 Gen, M., Liang, Z., Zhang, R., Go Mabato, B. R., and Chan, C. K.: Particulate nitrate photolysis
472 in the atmosphere, *Environmental Science: Atmospheres*, 10.1039/D1EA00087J, 2022.

473 Ji, X., Le Bihan, O., Ramalho, O., Mandin, C., D'Anna, B., Martinon, L., Nicolas, M., Bard, D.,
474 and Pairon, J.-C.: Characterization of particles emitted by incense burning in an experimental
475 house, *Indoor Air*, 20, 147-158, <https://doi.org/10.1111/j.1600-0668.2009.00634.x>, 2010.

476 Kucinski, T. M., Dawson, J. N., and Freedman, M. A.: Size-Dependent Liquid–Liquid Phase
477 Separation in Atmospherically Relevant Complex Systems, *The Journal of Physical Chemistry*
478 *Letters*, 10, 6915-6920, 2019.

479 Le Breton, M., Psichoudaki, M., Hallquist, M., Watne, Å., Lutz, A., and Hallquist, Å.: Application
480 of a FIGAERO ToF CIMS for on-line characterization of real-world fresh and aged particle
481 emissions from buses, *Aerosol Science and Technology*, 53, 244-259, 2019.

482 Lee, S.-C. and Wang, B.: Characteristics of emissions of air pollutants from burning of incense in
483 a large environmental chamber, *Atmospheric Environment*, 38, 941-951,
484 <https://doi.org/10.1016/j.atmosenv.2003.11.002>, 2004.

485 Li, J., Liu, Q., Li, Y., Liu, T., Huang, D., Zheng, J., Zhu, W., Hu, M., Wu, Y., and Lou, S.:
486 Characterization of aerosol aging potentials at suburban sites in northern and southern China
487 utilizing a potential aerosol mass (Go: PAM) reactor and an aerosol mass spectrometer, *Journal of*
488 *Geophysical Research: Atmospheres*, 124, 5629-5649, 2019.

489 Liang, Z., Zhang, R., Gen, M., Chu, Y., and Chan, C. K.: Nitrate Photolysis in Mixed Sucrose–
490 Nitrate–Sulfate Particles at Different Relative Humidities, *The Journal of Physical Chemistry A*,
491 125, 3739-3747, 10.1021/acs.jpca.1c00669, 2021.

492 Liang, Z., Zhou, L., Infante Cuevas, R. A., Li, X., Cheng, C., Li, M., Tang, R., Zhang, R., Lee, P.
493 K. H., Lai, A. C. K., and Chan, C. K.: Sulfate Formation in Incense Burning Particles: A Single-
494 Particle Mass Spectrometric Study, *Environmental Science & Technology Letters*, 9, 718-725,
495 10.1021/acs.estlett.2c00492, 2022.

496 Liu, T., Zhou, L., Liu, Q., Lee, B. P., Yao, D., Lu, H., Lyu, X., Guo, H., and Chan, C. K.: Secondary
497 Organic Aerosol Formation from Urban Roadside Air in Hong Kong, *Environ Sci Technol*, 53,
498 3001-3009, 10.1021/acs.est.8b06587, 2019.

499 Manoukian, A., Quivet, E., Temime-Roussel, B., Nicolas, M., Maupetit, F., and Wortham, H.:
500 Emission characteristics of air pollutants from incense and candle burning in indoor atmospheres,
501 *Environmental Science and Pollution Research*, 20, 4659-4670, 10.1007/s11356-012-1394-y,
502 2013.

503 Neubauer, K. R., Johnston, M. V., and Wexler, A. S.: Humidity effects on the mass spectra of
504 single aerosol particles, *Atmospheric Environment*, 32, 2521-2529, 1998.

505 Passig, J., Schade, J., Irsig, R., Kröger-Badge, T., Czech, H., Adam, T., Fallgren, H., Moldanova,
506 J., Sklorz, M., and Streibel, T.: Single-particle characterization of polycyclic aromatic
507 hydrocarbons in background air in northern Europe, *Atmospheric Chemistry and Physics*, 22,
508 1495-1514, 2022.

509 Qin, X. and Prather, K. A.: Impact of biomass emissions on particle chemistry during the California
510 Regional Particulate Air Quality Study, *International Journal of Mass Spectrometry*, 258, 142-150,
511 <https://doi.org/10.1016/j.ijms.2006.09.004>, 2006.

512 Rebotier, T. P. and Prather, K. A.: Aerosol time-of-flight mass spectrometry data analysis: A
513 benchmark of clustering algorithms, *Analytica Chimica Acta*, 585, 38-54,
514 <https://doi.org/10.1016/j.aca.2006.12.009>, 2007.

515 Schilling, J. B.: Extraction of semivolatile organic compounds from high-efficiency particulate air
516 (HEPA) filters by supercritical carbon dioxide, Argonne National Lab., Analytical Chemistry Lab.,
517 IL (United States), 1997.

518 Seinfeld, J. and Pandis, S.: *Atmospheric chemistry and physics*. 1997, New York, 2008.

519 Taiwo, A. M., Harrison, R. M., Beddows, D. C., and Shi, Z.: Source apportionment of single
520 particles sampled at the industrially polluted town of Port Talbot, United Kingdom by ATOFMS,
521 *Atmospheric Environment*, 97, 155-165, 2014.

522 Tsiligiannis, E., Hammes, J., Salvador, C. M., Mentel, T. F., and Hallquist, M.: Effect of NO_x on
523 1, 3, 5-trimethylbenzene (TMB) oxidation product distribution and particle formation,
524 *Atmospheric chemistry and physics*, 19, 15073-15086, 2019.

525 Wang, H., Guo, S., Wu, Z., Qiao, K., Tang, R., Yu, Y., Xu, W., Zhu, W., Zeng, L., and Huang, X.:
526 Secondary organic aerosol formation from straw burning using an oxidation flow reactor, *Journal*
527 *of Environmental Sciences*, 114, 249-258, 2022.

528 Watne, Å. K., Psychoudaki, M., Ljungström, E., Le Breton, M., Hallquist, M., Jerksjö, M., Fallgren,
529 H., Jutterström, S., and Hallquist, Å. M.: Fresh and oxidized emissions from in-use transit buses
530 running on diesel, biodiesel, and CNG, *Environmental science & technology*, 52, 7720-7728, 2018.

531 Xu, W., Li, Z., Lambe, A. T., Li, J., Liu, T., Du, A., Zhang, Z., Zhou, W., and Sun, Y.: Secondary
532 organic aerosol formation and aging from ambient air in an oxidation flow reactor during
533 wintertime in Beijing, China, *Environmental Research*, 209, 112751, 2022.

534 Yu, Y., Guo, S., Wang, H., Shen, R., Zhu, W., Tan, R., Song, K., Zhang, Z., Li, S., and Chen, Y.:
535 Importance of semivolatile/intermediate-volatility organic compounds to secondary organic
536 aerosol formation from Chinese domestic cooking emissions, *Environmental Science &*
537 *Technology Letters*, 9, 507-512, 2022.

538 Zhang, Z., Zhu, W., Hu, M., Wang, H., Chen, Z., Shen, R., Yu, Y., Tan, R., and Guo, S.: Secondary
539 organic aerosol from typical Chinese domestic cooking emissions, *Environmental Science &*
540 *Technology Letters*, 8, 24-31, 2020.

541 Zhao, W., Hopke, P. K., and Prather, K. A.: Comparison of two cluster analysis methods using
542 single particle mass spectra, *Atmospheric Environment*, 42, 881-892,
543 <https://doi.org/10.1016/j.atmosenv.2007.10.024>, 2008.

544 Zhou, L., Liu, T., Yao, D., Guo, H., Cheng, C., and Chan, C. K.: Primary emissions and secondary
545 production of organic aerosols from heated animal fats, *Science of the Total Environment*, 794,
546 148638, 2021a.

547 Zhou, L., Salvador, C. M., Priestley, M., Hallquist, M., Liu, Q., Chan, C. K., and Hallquist, Å. M.:
548 Emissions and Secondary Formation of Air Pollutants from Modern Heavy-Duty Trucks in Real-
549 World Traffic—Chemical Characteristics Using On-Line Mass Spectrometry, *Environmental*
550 *science & technology*, 55, 14515-14525, 2021b.

551