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- 1 Real-time molecular characterization of air pollutants in a Hong Kong residence: implication of
- 2 indoor source emissions and heterogeneous chemistry
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Abstract: Due to the high health risks associated with indoor air pollutants and long-term exposure, 9 indoor air quality has received increasing attention. In this study, we put emphasis on the molecular 10 composition, source emissions and chemical aging of air pollutants in a residence with designed 11 activities mimicking ordinary Hong Kong homes. More than 150 air pollutants were detected at 12 molecular level, 87 of which were quantified at a time resolution of not less than 1 hour. The indoor 13 to outdoor ratios were higher than 1 for most of the primary air pollutants, due to emissions of 14 indoor activities and indoor backgrounds (especially for aldehydes). In contrast, many secondary 15 air pollutants exhibited higher concentrations in outdoor air. Painting ranked first in aldehyde 16 emissions, which also caused great enhancement of aromatics. Incense burning had the highest 17 emissions of particle-phase organics, with vanillic acid and syringic acid as markers. The other 18 noteworthy fingerprints enabled by online measurements included linoleic acid, cholesterol and 19 20 oleic acid for cooking, 2,5-dimethylfuran, stigmasterol, iso-/anteiso- alkanes and fructose isomers for smoking, C<sub>28</sub>-C<sub>34</sub> even *n*-alkanes for candle burning, and monoterpenes for the use of air 21 freshener, cleaning agents and camphor oil. We showed clear evidence of chemical aging of 22 23 cooking emissions, giving a hint of indoor heterogeneous chemistry. This study highlights the 24 value of organic molecules measured at high time resolutions in enhancing our knowledge on indoor air quality. 25

# 26 **Practical Implications**

- Online mass spectrometry technology enabled definite and fast-response detection of a large number of air pollutants in a residence with implications on source emissions and indoor chemistry.
- Incense burning caused intensive emissions of organic matters in both gas and particulate phases.
- Distinct tracers identified for nine individual activities facilitated the understanding of
   emission characteristics of indoor sources.
- Chemical aging of cooking emissions was clearly shown, indicating the importance of
   indoor heterogeneous chemistry.

Keywords: Indoor air quality, volatile organic compounds, particulate matters, organic aerosol,indoor chemistry

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### 39 **1. Introduction**

People spend up to 90% of their time indoors (Spengler and Sexton, 1983). Indoor air pollution causes heart disease, pneumonia, stroke, diabetes and lung cancer, resulting in 1.6 million premature deaths globally in year 2017 (IHME, 2018). Some indoor sources, such as cooking emissions, also play significant roles in influencing ambient air quality (Lee et al., 2015; Wang et al., 2020a).

45 Though no consensus has been reached on which air pollutants are most responsible for the health effects of indoor air pollution, many air pollutants have been confirmed to be harmful to human 46 47 health (Salthammer et al., 2010; Chi et al., 2019), such as the gaseous pollutant formaldehyde and fine particulate matters (PM2.5). Therefore, great efforts have been made to understand the 48 chemical compositions of indoor air pollution as comprehensively as possible (Lee et al., 2002; 49 Weisel et al., 2008; Krugly et al., 2014; Zhao et al., 2019), plus the other characteristics of these 50 51 air pollutants (e.g. size distributions of ultrafine particles) (Morawska et al., 2003). Indoor air 52 pollutants are generated from a wide range of human activities (cooking, cigarette smoking, cleaning, human breath, etc.), furnishings, and building & decoration materials (Samet et al., 1987; 53 54 Habre et al., 2014; Liu et al., 2016). It is well documented that combustion of solid fuels for cooking and heating is the leading source of indoor air pollution with most serious health 55 consequence in developing countries (Ezzati, 2005). Additionally, outdoor air pollution presents 56 to be a universal source of air pollutants indoors due to air exchange (Blondeau et al., 2005; Chen 57 et al., 2012). The chemical evolutions of indoor air pollutants have not been studied as extensively 58 as those in ambient air (Weschler and Carslaw, 2018; Gligorovski and Abbatt, 2018). However, 59 recent studies discovered the higher-than-expected concentrations of hydroxyl radical (OH) in 60 special indoor settings, due primarily to the photolysis of combustion-related nitrous acid (HONO) 61 (Alvarez et al., 2013) and/or terpene-ozone reactions during cleaning (Carslaw et al., 2017). 62 Besides, the high surface-to-volume ratios indoors enhance the possibility and significance of 63 surface reactions (Ault et al., 2020; Wang et al., 2020b), accelerating heterogeneous formation of 64 HONO for example (Gandolfo et al., 2017; Liu et al., 2020). Recently, by applying novel tools for 65 studying atmospheric chemistry to indoor chemistry, researchers have made compelling 66 breakthroughs in revealing indoor dynamic source emissions and ozone chemistry (Kristensen et 67 68 al., 2019; Liu et al., 2019, 2021). Last but not the least, the occurrence and transformation of indoor air pollutants are not independent from the ventilation conditions (Godish and Spengler, 1996). 69

Hong Kong, a home of 7.5 million people with a land area of 1,100 km<sup>2</sup>, is one of the cities with 70 world's smallest living space per capita (<16  $m^2$  per capita for all housing and <5 for subdivided 71 apartments) (NYT, 2019). The city is also known for its high densities of population, buildings 72 and vehicles. Catering is well developed in Hong Kong, forming unique mix of Chinese and 73 western foods. At a roadside site in vicinity of residential areas in this city, cooking emissions 74 were found to be responsible for 38.6% (by mass) of the ambient organic aerosols (Lee et al., 75 2015). Besides, people in South China including Hong Kong succeed the custom of burning 76 candles and/or incenses when worshiping ancestors, ghosts and gods in house at daily to monthly 77 frequency depending on individuals. Other activities, such as smoking, cleaning and laundry, are 78 also common in Hong Kong homes. The outdoor air pollution is mainly manifested as regional 79

photochemical smog and street-level air pollution resulted from vehicle emissions (Wang et al.,
2017; Zhu et al., 2018). Lastly, warm and humid climate prevails in Hong Kong for more than half
of a year, making it unlikely to stay indoors without air conditioning, which is another factor

83 influencing indoor air quality in Hong Kong.

The concentrations, compositions and sources of regular air pollutants, e.g. volatile organic 84 compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and criteria air pollutants, were 85 studied in Hong Kong (Lee et al., 2001, 2002; Guo et al., 2003, 2009; Ai et al., 2015). The health 86 87 implications of indoor air pollution or environmental factors were also investigated (Yu et al., 2004). In particular, Yu et al. (2004) and Li et al. (2007) indicated the airborne transmission of 88 SARS virus in residential building and hospital, and its relationship with ventilation. However, the 89 90 previous studies outline the gaps in understanding indoor air pollution in Hong Kong. Firstly, the molecular compositions of condensed organics, a dominant constituent of indoor particular matters 91 (Polidori A et al., 2006), were seldom analyzed except for PAHs (Tong et al., 2019). The 92 measurements lacked either high time resolution or chemical completeness. Secondly, it was 93 difficult to recognize the emission characteristics of individual indoor sources due to the 94 95 cumulative sampling methods. Lastly, we have little knowledge on indoor chemical evolutions in Hong Kong. 96

97 In this context, we performed a real-time molecular characterization of indoor air pollutants in a 98 residence with designed activities mimicking ordinary Hong Kong homes, using a set of online 99 instruments. This study provides an overview of the sampling campaign, focusing on the 100 concentrations and origins of air pollutants measured in the residence, emission characteristics of 101 indoor activities, and the chemical evolution of cooking emissions. The real-time measurements 102 of indoor air pollutants at the molecular level, especially organics, allowed an unprecedented 103 insight on indoor air quality in Hong Kong homes.

## 104 2. Methodology

As shown in Figure 1, an apartment, consisting of a living room, 2 bedrooms, a kitchen and a 105 bathroom, was rented for the study. The apartment was located on the 7<sup>th</sup> floor of a 22-year-old 106 building with 17 floors in a residential area of Kowloon, Hong Kong. In addition to common 107 sources of air pollutants in cities, e.g. vehicles, restaurants and gas station, two funeral parlors 108 were located ~220 m south. The floor area of the apartment was 25.4 m<sup>2</sup>, a typical space for an 109 ordinary Hong Kong family with 3-4 members. The ventilation system comprised 5 windows, 3 110 air conditioners (ACs), an exhaust fan in the bathroom, and a range hood in the kitchen. The air 111 exchange rate (AER) of the apartment was measured in a few scenarios, based on the decay of 112 carbon dioxide (CO<sub>2</sub>) when there was no occupant in the apartment. To maintain comfort and 113 ensure normal operation of the instruments, at least 1 AC was on all the time. Irrespective of 114 which AC(s) was (were) on and which door(s) within the suite was (were) open, the AER remained 115 stable  $(1.1 - 1.2 \text{ h}^{-1})$ . It did not change much, unless the range hood was on and the kitchen door 116 was open, which brought the AER to  $2.9 \text{ h}^{-1}$ . 117

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Common activities in Hong Kong homes were designed and carried out following the schedules 119 shown in Figure 2. To mimic the real situations, the activities were conducted at time and 120 121 frequencies close to actual since the normal operation of all instruments on 18 November 2019, while the activity frequencies were lower in the earlier phase of the sampling campaign. The 122 intensity of the same activity was not necessarily consistent, e.g. simple breakfast vs. big dinner, 123 124 usually 1 cigarette vs. occasionally 2 cigarettes for smoking, and the burning of scented candle vs. ritual candle. The consequent differences in indoor air quality are discussed where necessary. 125 Natural gas was the main fuel for cooking, with electricity as an alternative when induction cooker 126 was occasionally used. The types of cigarettes, incenses, and air fresheners were consistent 127 throughout the study. In most of the daytime, there were 2 occupants in the apartment. The 128 recorded temperature and relative humidity in the apartment were in the range of 20.0–30.2 °C and 129 22.1%-57.4%, with an average of 25.5 °C and 33.7%, respectively. 130

As shown in Figure 1, all the instruments were deployed in the living room. The indoor and outdoor 131 air was sampled through a  $\sim 2$  m and  $\sim 3$  m pre-cleaned copper tubing (Restek Cat.# 22629, ID: 132 0.19") at the flow rate of 16.7 L m<sup>-1</sup>, respectively. Due to the short residence time (<1 s), low wall 133 loss of charged particles, and relatively low delay on transfer of organic vapors (Deming et al., 134 2019), the copper tubing was suitable for simultaneous gas and particle sampling. However, the 135 partitioning and wall loss could not be completely ruled out. A High Resolution-Time of Flight-136 Aerosol Mass Spectrometer (HR-ToF-AMS, hereinafter referred to as AMS) was used to measure 137 the non-refractory compositions of submicron particulate matters (PM<sub>1</sub>), including organic matters 138 (PM<sub>1</sub>-OM), sulfate, nitrate, ammonium and chloride. The concentrations were recorded every 2 139 minutes. Speciated organic aerosol (OA) markers in PM2.5 were analyzed by a Thermal desorption 140 Aerosol Gas chromatograph-Time of Flight-Mass Spectrometer (TAG-ToF-MS, hereinafter 141 referred to as TAG). We identified 92 OA markers according to their retention time and mass 142 spectra, and quantified the concentrations of 64 species at the time resolution of 1 hour. The 143 operation principles, measurement configurations, calibrations and data quality control of AMS 144 and TAG were elaborated in Lyu et al. (2020). The accuracy and precision for the measurements 145 were within the range of 1.5%–21.2% and 4.1%–33.9%, respectively. 146

The VOCs were detected utilizing a quadruple Proton Transfer Reaction-Mass Spectrometer 147 (PTR-MS, hereinafter referred to as PTR). The procedures for PTR calibration are described in 148 Text S1 and Figure S1. 50 VOC species were identified, 12 of which were quantifiable with the 149 accuracy of 3.0%–19.9% and precision of 1.6%–10.0%. The time resolution of PTR measurement 150 was 1 minute. Besides, 6 inorganic trace gases, including sulfur dioxide (SO<sub>2</sub>), carbon monoxide 151 (CO), nitric oxide (NO), nitrogen dioxide (NO<sub>2</sub>), ozone (O<sub>3</sub>) and carbon dioxide (CO<sub>2</sub>), were 152 monitored by a set of trace gas analyzers at the resolution of 1 minute (Table S1). All the online 153 instruments measured the indoor and outdoor air in turn every other hour, which was achieved 154 through automatic switching of 2 solenoid valves installed on the sampling lines (Figure 1). 155 Exhausts of all the instruments were pumped out of the apartment. Due to malfunctions of some 156 instruments (especially TAG), the periods with valid data for the species measured online were 157 not exactly the same (Table S1). 158

In total, we obtained the mixing ratios (or concentrations) for 87 out of 153 air pollutants detected 159 at the time resolution of 1 minute to 1 hour. While we mainly focus on the quantifiable air 160 161 pollutants, the signal intensities of some air pollutants which were not quantified are also adopted for discussions. The sensitivity of PTR was relatively stable, based on the small changes in K 162 values between every two calibrations (<7%). For TAG data, the signal intensities were the peak 163 164 areas of target compounds scaled by those of corresponding internal standards (ISs), i.e. IS-scaled peak areas. The ISs, comprising 41 deuterated compounds, were used to track and correct the 165 changes in instrument sensitivity (Lyu et al., 2020). Therefore, it is reasonable to use the signal 166 intensities for ratio analyses. 167

To facilitate discussions, the 64 quantifiable OA markers are classified as even *n*-alkanes, odd *n*alkanes, PAHs, phenolic acids, phthalic acids, saccharides, sugar alcohols, fatty acids, oleic acid, dicarboxylic acids (DCA), hydroxyl dicarboxylic acids (OHDCA), Secondary Organic Aerosol (SOA) tracers of isoprene (Isop-SOA-T), monoterpenes SOA tracers (MT-SOA-T), and 2,3-

dihydroxy-4-oxopentanoic acid (DHOPA). The species in every group are listed in Table S2.

## 173 **3. Results and Discussion**

### **3.1. Indoor versus outdoor concentrations**

Figures S2–S5 show the time series of gas-phase and particle-phase air pollutants that were quantified in this study. The average mixing ratios (or concentrations) in 3 scenarios, i.e. with indoor activities, without indoor activity and outdoors, are compared in Table 1.

Most of the air pollutants with predominantly primary sources exhibited higher levels indoors, 178 resulting in the indoor-to-outdoor (I/O) ratios of higher than 1. This did not necessarily mean the 179 more intensive emissions of interior sources but demonstrated the great impacts of interior sources 180 on indoor air quality, due to proximity to emissions and worse dispersion conditions indoors than 181 outdoors. Exceptions were NO, NO2, SO2, chloride, PAHs, phenolic acids and phthalic acids, 182 whose I/O ratios were close to or even lower than 1. Because of intensive emissions indoors, the 183 low I/O ratios indicated significant outside emissions, such as NO, NO2 and PAHs in vehicle 184 exhausts (Guo et al., 2003), and phenolic acids emitted from burning of worship supplies (mainly 185 186 biomass) in nearby funeral parlors (Simoneit et al., 1999). The indoor mixing ratios (or concentrations) of all the primary air pollutants were considerably elevated in presence of activities, 187 which partially accounted for the I/O ratios of higher than 1. In absence of activity, the 188 189 concentrations of the primary particle compositions were lower than those in ambient air. However, it was interesting that the mixing ratios of CO<sub>2</sub> and aldehydes indoors without activity still 190 overwhelmed those outdoors. While the higher CO<sub>2</sub> levels were explained by the breath of 191 occupants, the more abundant aldehydes in the apartment even in absence of activity were likely 192 caused by emissions from furnishings and building & decoration materials (Baumann et al., 2000, 193 Uhde and Salthammer, 2007). 194

195 In contrast, the I/O ratios were lower than 1 for a majority of secondary air pollutants, excluding

- 196 those having primary emissions as alternative sources (e.g. formaldehyde) and oxidation products
- 197 of indoor emissions (e.g. 9-oxononanoic acid produced by ozonolysis of oleic acid, see section
- 198 3.3). O<sub>3</sub> is a typical photochemical air pollutant in ambient air (Lin et al., 1988), which presented

indoors mainly due to air exchange. Sulfate, nitrate and ammonium are well-documented 199 compositions of secondary inorganic aerosols (Huang et al., 2014). Oxidations of anthropogenic 200 201 and biogenic VOCs lead to the formations of DCA, OHDCA, Isop-SOA-T, MT-SOA-T and DHOPA (Blando and Turpin, 2000, Hu et al., 2008), while DCA is also derived from primary 202 203 emissions. According to the PTR data that were not quantified, the signal intensities of some VOC 204 precursors of these SOA markers indoors were higher than those outdoors, such as isoprene (Isop-SOA-T precursor), monoterpenes (MT-SOA-T precursors), and toluene (one of DHOPA 205 precursors). Therefore, the lower mixing ratios (or concentrations) of the secondary air pollutants 206 indoors most likely indicated weaker oxidative capacity of the indoor air than ambient air. 207

### **3.2.** Emission characteristics of indoor activities

As discussed, indoor activities caused obvious enhancements of primary air pollutants. Figure 3 209 210 shows the variations of some selected air pollutants on one day with intensive activities as an 211 example. Increases of NO, NO2 and PM1-OM were observed during cooking noodles in the 212 morning with range hood off. While all of them could be emitted from natural gas combustion (Singer et al., 2017), the increase of PM<sub>1</sub>-OM was also associated with the stir fry of side dishes, 213 214 in the light of rising fractions of saccharides and oleic acid. Differently, making lunch and dinner 215 resulted in much less pronounced increases of these air pollutants, though more dishes were made. A plausible explanation was the effect of the range hood, which was turned on during both cooking 216 periods. We compared the indoor mixing ratios (or concentrations) of some representative air 217 pollutants with and without range hood exhaust during 2 cooking practices, when the range hood 218 was turned on intermittently. As shown in Table S3, the indoor levels of these species were reduced 219 by 4% - 81% with the range hood exhaust, confirming the effectiveness of the range hood in 220 221 removing cooking emissions from the apartment. Besides, the fraction of oleic acid increased markedly during making lunch and dinner, due to the use of larger amount of oil than in making 222 breakfast. 223

The occupant smoked in the apartment at  $\sim 12:20$  and  $\sim 18:00$ , causing tremendous rise in the 224 concentrations of PM<sub>1</sub>-OM and nitrate, as well as the mixing ratios of many VOCs and CO. 225 226 Saccharides experienced the largest enhancements among the OA markers that were quantified, thereby an obvious increase of the proportion in the total concentration of OA makers. A ritual 227 candle was burned during 22:21–22:40 in the kitchen. Correspondingly, we observed notable 228 increases of NO, NO<sub>2</sub>, SO<sub>2</sub>, CO and *n*-alkanes (especially even *n*-alkanes). Spraying air freshener 229 in the morning ( $\sim 10:25$ ) was responsible for the enhancements of formaldehyde and acetaldehyde. 230 Lastly, makeup in the morning and eating fruit (persimmon) & drinking tea in the afternoon did 231 not cause significant changes in the mixing ratios (or concentrations) of air pollutants discussed 232

- 233 here. However, the use of camphor oil as a repellent in the makeup substantially increased the
  - signal intensity of monoterpenes (see section 3.3).
  - Furthermore, we calculated the net increments (NIC) of the 12 quantifiable aldehydes during
  - individual indoor activities, with the formula of NIC =  $C_{w/A} C_{w/A}$ .  $C_{w/A}$  and  $C_{w/A}$  represent
  - 237 the mixing ratios with and without the presence of indoor activities, respectively.  $C_{w/A}$  were the
  - averages over the rising, plateau (if any) and decline periods of the mixing ratios, and the time
  - frames for calculating  $C_{w/oA}$  were the periods before the activities within the same hours the

activities were conducted. Since the sampling switched to outdoor air in odd hours, the decline
periods were not complete in some cases. Therefore, the NICs were somewhat overestimated.
However, we do not think this had a great impact on the fractions of individual aldehydes in the
total NIC of the 12 aldehydes, bearing in mind the discrepancies in decay rates of different
aldehydes.

Figure 4 shows the total NIC of the 12 aldehydes during individual indoor activities and the 245 apportionments. Overall, painting ranked the first in the total NIC, followed by incense burning 246 247 and smoking. Ritual candle burning had the lowest NIC of aldehydes. Butanal contributed 54.6% 248 to the total NIC during painting, which even played more predominant role (82.1%) in the total NIC attributable to cleaning. Pentanal, formaldehyde and butanal were the main aldehydes emitted 249 250 from incense burning (smoking), contributing 40.9% (43.5%), 17.1% (12.1%) and 14.3% (16.6%) to the total NIC, respectively. Emissions from spraying air freshener mainly consisted of 251 formaldehyde, butanal and propanal. Hexanal accounted for 72.2% of the total NIC of the 12 252 aldehydes in emissions of scented candle burning. Pentanal, formaldehyde and hexanal were the 253 main constituents of cooking emissions. While hexanal dominated the total NIC during the use of 254 camphor oil, other aldehydes (such as heptanal, propanal and decanal) also made considerable 255 contributions. The total NIC during ritual candle burning was mainly attributed to formaldehyde 256 (44.6%), propanal (15.4%), decanal (14.1%) and acrolein (10.3%). It should be noted that the pie 257 areas in Figure 4 are proportional to the total emissions of the 12 aldehydes instead of all VOCs. 258 Many VOCs with great enhancements in some activities were not quantified, such as ethanol in 259 spraying air freshener and monoterpenes in cleaning and camphor oil use (see section 3.3). 260

Similarly, the NICs of particle-phase air pollutants were calculated, as shown in Figure 5. 261 Activities that did not cause significant changes in PM1 compositions are not discussed. It is 262 striking that the PM1-OM contributed more than 95% of the total NIC of PM1 during cooking and 263 all the activities involving combustion except scented candle burning. The burning of scented 264 candles only increased PM<sub>1</sub> concentration by  $5.2 \pm 1.7 \,\mu g \, m^{-3}$  on average, which was 6.6–37 times 265 lower than the NICs of PM<sub>1</sub> caused by the other activities. During scented candle burning, 76±0.4% 266 of the NIC of PM<sub>1</sub> was attributed to PM<sub>1</sub>-OM (NIC:  $4.0 \pm 1.7 \ \mu g \ m^{-3}$ ), with the rest contributed by 267 sulfate (~16%) and ammonium (~7%). In contrast, incense burning caused the largest NIC of PM<sub>1</sub>-268 OM (189 ± 45  $\mu$ g m<sup>-3</sup>), followed by smoking (119 ± 15  $\mu$ g m<sup>-3</sup>), cooking (41 ± 7.3  $\mu$ g m<sup>-3</sup>) and 269 ritual candle burning  $(33 \pm 7 \mu \text{g m}^{-3})$ . While continuous emissions were observed during the other 270 combustion activities, scented candle burning mainly led to enhancement of PM1-OM in the 271 ignition and extinction states, with much higher concentrations in the extinction state. This was 272 due to the emissions of larger size particles in the smoldering phase immediately after the candle 273 was extinguished (Manoukian et al., 2013). For ritual candle burning, smoke plumes were 274 discernable even in the flaming phase, which might explain the different emission patterns. 275 Furthermore, we investigated the elemental compositions of PM1-OM detected by AMS in indoor 276 activities. The hydrogen to carbon atomic ratios (H:C) and oxygen to carbon atomic ratios (O:C) 277 are plotted in the right panel of Figure 5. Highest H:C ratios (1.94±0.02) and lowest O:C ratios 278 279  $(0.23\pm0.02)$  were observed during ritual candle burning. The features, especially the proximity of average H:C ratio to that in long acyclic alkanes (~2), indicated strong emissions of high saturation 280 molecules (such as alkanes) from ritual candle burning (Heald et al., 2010). In contrast, incense 281

- burning witnessed the lowest H:C ratios  $(1.76\pm0.01)$  and highest O:C ratios  $(0.42\pm0.01)$ , implying multiple oxygen-containing functional groups in the molecules emitted from incense burning (Heald et al., 2010). These inferences are verified by the enhancement ratios (ERs) of speciated
- OA markers below, while the NICs of them will be presented in a subsequent paper.

#### 286 **3.3. Enhancement ratios**

287 Since many air pollutants (especially VOCs) that were not quantified experienced significant enhancements in signal intensities during some indoor activities, neglecting them might lead to 288 incomplete or even biased understandings on emission characteristics of indoor activities. To make 289 use of the unquantified species, the ERs are calculated for all the detected air pollutants following 290 the formula of ER =  $C_{w/A}/C_{w/A}$ . While  $C_{w/A}$  and  $C_{w/A}$  have been defined for the quantified 291 species, they represent the signal intensities for the unquantified species with and without the 292 indoor activities, respectively. Limited by the time resolution of 1 hour for TAG data, we 293 calculated  $C_{w/oA}$  on the days without activity and in the same hours as  $C_{w/A}$  was calculated. 294

Table 2 lists the top 5 ERs for indoor gaseous and particulate air pollutants. Cooking resulted in 295 high ERs for inorganic trace gases (NO, CO and NO<sub>2</sub>), due to the combustion of natural gas (Singer 296 et al., 2017). Ethanol also had an ER of 1.9, which was caused by the occasional use of alcohol-297 298 containing ingredients, such as fermented rice. Cyclohexane was measured in cooking emissions 299 with the emission rates lower than many other VOCs (e.g. formaldehyde, acetaldehyde and 2butanone) in previous studies (Alves et al., 2015; He et al., 2020). The reason for the high ER of 300 301 cyclohexane in this study is unknown. The particle-phase species with high ERs in cooking 302 included linoleic acid, cholesterol, arabinitol/xylitol,  $\beta$ -sitosterol and oleic acid. Linoleic acid,  $\beta$ sitosterol and oleic acid were most likely derived from the oils (corn oil and peanut oil) and 303 ingredients (Zhao et al., 2007). Cholesterol and arabinitol/xylitol were mainly derived from the 304 ingredients, because they were enhanced when fried eggs & fishes and stir-fried vegetables were 305 made, respectively. The most noteworthy gaseous air pollutant during smoking was 2,5-306 dimethylfuran whose ER was 4.3. 2.5-dimethylfuran was proposed as an excellent marker for 307 smoking in blood and breath samples (Ashley et al., 1996; Castellanos et al., 2019). In particle 308 phase, the iso-/anteiso-alkanes were observed with high ERs during smoking, consistent with 309 previous studies (Cass, 1998). Stigmasterol, which had the highst ER (269), is one of the most 310 abundant plant sterols and has often been detected with great abundance in smoking emissions 311 (Rogge et al., 1994). Simple sugars (e.g. fructose, glucose and sucrose) are important components 312 and common additives in tobacco, presenting at levels of up to 20% in tobacco by mass (Jansen et 313 al., 2014). This explained the notable enhancement of fructose isomers (ER = 162) during smoking. 314 The gaseous air pollutants differentiating incense burning from the other activities were benzene, 315 propene and acetaldehyde. Vanillic acid and syringic acid, pyrolysis products of lignin (Simoneit 316 et al., 1993), were unique markers of incense burning, with the ER of 108 and 80, respectively. 317 While  $C_{28}$ - $C_{34}$  even *n*-alkanes were significantly enhanced during the burning of both scented 318 candle and ritual candle, the former witnessed enhancements of fatty acids (stearic acid and 319 palmitic acid),  $\beta$ -sitosterol and fructose isomers, in contrast to the C<sub>27</sub>-C<sub>35</sub> odd alkanes with 320 different structures for the latter. It should be noted that the higher ERs for C<sub>28</sub>-C<sub>34</sub> even *n*-alkanes 321 were due to the lower  $C_{w/oA}$ . In fact, the NIC of C<sub>27</sub>–C<sub>35</sub> odd *n*-alkanes was comparable to that of 322

323  $C_{28}-C_{34}$  even *n*-alkanes for both candles, consistent with the emissions of paraffin candles (Fine 324 et al., 1999). The high ERs of alkanes in ritual candle burning were roughly consistent with the 325 H:C ratio of 1.94±0.02 detected by AMS. Besides, the gaseous air pollutants with top 5 ERs were 326 also different (except NO) for the burning of two types of candles.

The other indoor activities did not lead to significant increases of particle-phase air pollutants, but 327 the signal intensities of some VOCs were obviously enhanced. For example, the ER of 328 monoterpenes (limonene/ $\alpha$ -pinene/ $\beta$ -pinene) reached 196, 10 and 3.7, when we used camphor oil, 329 330 air freshener and cleaning agents, respectively. Air freshener was also a strong source of ethanol (ER = 129). Styrene, formaldehyde and linalool/geraniol with ERs of 3.0–7.5 were widely detected 331 in air freshener spray (Kim et al., 2015; Steinemann, 2017). Painting caused tremendous 332 333 enhancements of aromatics and acrolein. Studies confirmed considerable emissions of aromatics 334 in paint applications (Yuan et al., 2010). Acrolein, as a combustion by-product of organics (Seaman et al., 2007), however was not identified to be associated with painting. The reason for 335 its enhancement is unknown and should be referenced with caution. 336

### 337 **3.4.** Chemical evolution of cooking emissions

Chemical evolution of indoor air pollutants has significant impacts on the fate and subsequent 338 339 health risk of primary emissions. In this study, the aging of cooking emissions provided some 340 insights into the indoor heterogeneous chemistry. Studies (Vesna et al., 2009; Zeng et al., 2020) indicated that unsaturated fatty acids (e.g. oleic acid and linoleic acid) were prone to be oxidized 341 by O<sub>3</sub> and OH radical via heterogeneous reactions. The oxidation products, i.e. nonanal and 342 nonanoic acid were too volatile to be measured by TAG. Azelaic acid and 9-oxononanoic acid, the 343 other two oxidation products of cooking emissions, were reasonably detected. However, 344 simultaneous enhancements of azelaic acid and oleic acid were observed during cooking, 345 indicating the primary emissions of azelaic acid, which was confirmed by previous studies (He et 346 al., 2004). Therefore, we select 9-oxononanoic acid as a representative of the oxidation products. 347 Since 9-oxononanoic acid was not quantified due to lack of standard, the IS-scaled peak areas were 348 used in the analyses below. 349

Figure 6(a)–(b) shows the diurnal patterns of 9-oxononanoic acid and O<sub>3</sub>. The average IS-scaled 350 peak area of 9-oxononanoic acid indoors  $(0.97\pm0.11)$  was significantly (p<0.01) higher than that 351 outdoors (0.72±0.11). For both indoor and outdoor profiles, bimodal distributions were observed 352 with one peak in the afternoon and the other peak in the evening. The daytime variations of 9-353 oxononanoic acid coincided fairly well with the patterns of O<sub>3</sub>. It is expected that the indoor OH 354 had the peak at noon, when the ambient OH reached highest levels and the HONO photolysis 355 indoors was most intensive driven by the elevated HONO levels during making lunch and strongest 356 sunlight that penetrated windows. Therefore, the afternoon peak of 9-oxononanoic acid was likely 357 explained by the OH-initiated oxidations of cooking emissions and the peak delay relative to OH 358 might be due to the oxidation processes of several generations (Zeng et al., 2020). However, the 359 possible O<sub>3</sub>-involved formation pathways could not be excluded (Vesna et al., 2009). It seemed 360 that O<sub>3</sub> alone was not accountable for the higher levels of 9-oxononanoic acid indoors and in the 361 evening, where and when the O<sub>3</sub> mixing ratios were relatively low. A plausible explanation was 362 that the abundance of precursors was another influencing factor. For example, oleic acid was more 363

abundant in indoor air, and reached its highest concentrations in making dinner, which fueled the production of 9-oxononanoic acid in the evening. Besides, the high levels of indoor 9-oxononanoic acid might be partially explained by OH-initiated oxidations, especially when HONO levels were elevated after cooking. The factors influencing the yields of 9-oxononanoic acid are also worth exploring.

Furthermore, we examined the levels of 9-oxononanoic acid and oleic acid in the indoor samples 369 collected during cooking and two hours later. A marginal increase (p=0.29) was identified for 9-370 371 oxononanoic acid within the 2 hours when oleic acid decreased remarkably (Figure 6(c)). As a result, the ratio of 9-oxononanoic acid/oleic acid went up (Figure 6(d)). At the same time, the 372 decrease of oleic acid/stearic acid ratio (Figure 6(d)) was a sign of chemical aging of cooking 373 emissions, because oleic acid is more reactive than stearic acid (Wang et al., 2020). It is noteworthy 374 that some indoor activities were occasionally conducted 2 hours after cooking, which might 375 interfere with the chemical compositions of TAG samples. However, the conclusion will still hold 376 even if all the samples subject to interference of other activities are filtered out. 377

378 In general, there was no activity with intensive emissions after making dinner till the next morning, except for a few times of candle burning. Therefore, it provided a good opportunity to look into 379 the continuous variations of the fingerprints of cooking emissions and the chemical evolution. As 380 shown in Figures 6(e)-(f), the level of 9-oxononanoic acid gradually increased after cooking, 381 reaching the maximum 4-8 hours later when oleic acid decreased to the lowest level. We noticed 382 that the increase rate of 9-oxononanoic acid within the first 2 hours (9.4% hr<sup>-1</sup>) was much lower 383 than the decrease rate of oleic acid (-45.8% hr<sup>-1</sup>). This might be due to the fact that 9-oxononanoic 384 acid is not the first-generation product of the unsaturated fatty acids (Vesna et al., 2009), hence 385 time difference between the degradation of the initial precursors and production of 9-oxononanoic 386 acid. As expected, the ratio of 9-oxononanoic acid/oleic acid increased within the first 10 hours, 387 and decreased subsequently due to some fresh emissions of oleic acid in making breakfast and 388 decay of 9-oxononanoic acid. A continuous decline was observed for the ratio of oleic acid/stearic 389 acid, till the last 2 hours. Overall, the dynamic characteristics of all the fingerprints analyzed from 390 TAG data clearly indicated the chemical aging of cooking emissions, even at night. 391

## 392 4. Conclusions

Indoor air quality is a big health concern in Hong Kong, where people have to endure long working 393 hours, small living space and a wide range of indoor sources of air pollutants. This study focused 394 on air pollution in Hong Kong homes, one of the most important indoor environments. We 395 deployed a set of online instruments in a residence for 3-4 occupants. Air pollutants in gas and 396 particle phases were chemically characterized at molecular level and improved time resolutions. 397 The origins for most of the primary and secondary air pollutants were unambiguously 398 distinguishable, based on not only the established knowledge but also the stratified I/O ratios with 399 1 as the benchmark. While the presence of indoor activities was an essential reason for the higher 400 concentrations of primary air pollutants indoors, aldehydes had I/O ratios higher than 1 even in 401 absence of any activity. Emission characteristics of 9 indoor activities were thoroughly revealed. 402 403 Painting and incense burning had the highest emissions of aldehydes and PM1-OM, respectively. The molecular fingerprints of individual indoor activities were identified, such as linoleic acid and 404

405 oleic acid in cooking emissions, and 9-oxononanoic acid as one of their oxidation products. Based 406 on the evolutions of relevant fingerprints, the chemical aging of cooking emissions indoors was 407 clearly indicated. The high apartment rent, large sizes of the instruments and intensive labor in 408 transferring, deploying and maintaining the instruments hamper widespread application of the 409 online measurements in more dwellings. However, the indoor sources and chemistry examined are 410 expected to be ubiquitous in this region. Therefore, this study gives unprecedented insights into

- 411 the dynamic profiles and chemical evolutions of air pollutants that may be present in ordinary
- 412 Hong Kong homes, and is also a prototype for more in-depth indoor air quality studies.
- 413

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- 421

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