1 **Emission characteristics and dynamic source apportionment of speciated organics in**

2 **particulate matters in a Hong Kong residence**

3 Yunxi Huo, Hai Guo*, Xiaopu Lyu, Dawen Yao,

4 Air Quality Studies, Department of Civil and Environmental Engineering, Hong Kong

5 Polytechnic University, Hong Kong

6 * Correspondence to Hai Guo (hai.guo@polyu.edu.hk) and Xiaopu Lyu

7 (xiaopu.lyu@polyu.edu.hk)

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9 **Abstract**: A growing number of studies warn of the adverse health effects of indoor particulate

10 matters (PM). However, little is known about the molecular compositions and sources of PM-

11 bound organics, a critical group of species with highest concentration and complexity in indoor

12 PM. In this study, we measured the chemical compositions of PM with a focus on speciated

13 organics in a Hong Kong residence, where prescribed activities were performed with normal

14 frequency and intensity. The PM-bound organic matters (OM) showed higher concentrations

15 indoors (21.1 ± 6.6 μg/m^3) than outdoors (10.3 ± 0.7 μg/m^3) ($p<0.05$), but the indoor

16 concentrations were lower (8.2 ± 0.1 μg/m^3) during the undisturbed periods, i.e., periods when

17 there was no activity or the PM-bound OM levels were not influenced by activities (e.g.,

18 laundry). A large number of organic molecules were identified to be of indoor origins.

19 Chemically comprehensive emission profiles of speciated organics were obtained for 5 indoor

20 activities in the residence. Source apportionment based on speciated organics measured at

21 hourly resolution revealed that cigarette smoking and cooking were the largest contributors to

22 PM-bound OM among all the prescribed activities during the disturbed periods. In contrast,

23 outdoor air infiltration dominated the sources of indoor PM-bound OM during the undisturbed

24 periods, with an average contribution of 77.1%. This study enhances knowledge on emissions

25 and sources of speciated organics in indoor PM and may benefit research on health risks of

26 indoor air.

27 **Keywords:** Indoor air quality; Particulate matters; Organic compounds; Emission profile;

28 Source apportionment

29

30 **Practical Implications:**

31 • Organics dominated the compositions of indoor PM, accounting for higher mass

32 fraction than that in outdoor counterpart.

33 • Some indoor activities could increase the concentrations of PM-bound organics by up

34 to hundreds of folds, implying health risks of acute exposure.

- Emission profiles of speciated organics determined for high-emission activities in real residence will be of great value in guiding source apportionment of indoor PM with respect to mass concentration and toxicity.

- Improving indoor air quality cannot be independent of tackling ambient air pollution, due to the overwhelming contributions of outdoor air infiltration to PM-bound organics during undistributed periods.

1. Introduction

People spend approximately 80%–90% of their time indoors (Klepeis et al., 2001). The premature deaths associated with indoor air pollution continuously increase, especially in developing countries (Smith and Mehta, 2003). Fine particulate matter (PM$_{2.5}$) has been identified as a major health killer causing respiratory and cardiovascular diseases (Li et al., 2003; Pope et al., 1995). Organics account for significant fractions of PM$_{2.5}$ indoors (Rivas et al., 2014) and lead to adverse health outcomes as indicated by epidemiological and toxicological studies (WHO, 2013; Breitner et al., 2011). Moreover, the toxicities of particulate organics differ among species and are not necessarily mass-dependent (Jin et al., 2019). Molecular information helps to identify the sources and chemical evolution of PM-bound organics (Lyu et al., 2021; Zeng et al., 2020), which eventually enhances the traceability of indoor air toxicity. Therefore, studies on speciated organics provide a fundamental insight on indoor air quality.

Indoor sources including occupant-related activities generate a large number of organics spanning a wide range of volatility, with considerable fractions in PM (Wu et al., 2018; Wei et al., 2016). While studies shed some light on the composition of indoor PM-bound organics, the conventional filter-based and long-time-span sampling failed to recognize the emissions of individual sources from convolved chemical information (Lai et al., 2020a; Loupa et al., 2016). Chamber experiments are capable of measuring and even quantifying emissions from individual indoor sources (Manoukian et al., 2013; Lee et al., 2006, 2004). However, many factors in real indoor environments, such as ventilation conditions and concentrations of oxidants which may influence the diffusion and chemical aging of air pollutants, are difficult to be reproduced in chambers (Morrison et al., 2019; Vu and Harrison, 2019). In addition, with the filter-based measurements or chamber experiments, it is almost impossible to examine the chemical evolution of organics in real indoor environments, due to the low time resolution or incomparable settings. Aerosol Mass Spectrometry (AMS) partly overcomes the deficiencies of traditional filter-based method and is increasingly used in analysis of indoor PM-bound organics in recent years (Avery et al., 2019; Johnson et al., 2017). Nevertheless, it measures total and fragmented organics instead of specific molecules, which impedes identification of definite sources and related toxicity.

Thermal-desorption Aerosol Gas-chromatograph (TAG) coupled with mass spectrometry is an emerging technique tailored for in-situ analysis of speciated organics in PM, which has been extensively applied in atmospheric chemistry studies (Isaacman et al., 2014; Williams et al., 2006). In brief, it automates the processes of sample collection, transfer and detection and

incorporates online derivatization and calibration with internal standards, enabling quantitative analysis of a wide range of both polar and non-polar organics (Lyu et al., 2020). Recently, TAG has been applied to study the indoor PM-bound organics (Lunderberg et al., 2020, 2019; Kristensen et al., 2019), whose sources, evolution and dynamic behaviors under changing conditions (e.g., temperature, ventilation and outdoor air pollution) were investigated with designed experiments. For example, the first deployment of TAG in a California residence revealed continuous emissions of semi-volatile organic compounds (SVOCs) from static indoor sources, while the abundances of SVOCs were almost unaffected by the number of occupants (Kristensen et al., 2019). Based on the fast response data measured by TAG, Lunderberg et al. (2019) indicated that indoor air temperature and particle loading were key factors that influenced the gas-particle partitioning of diethylhexyl phthalate indoors. Overall, TAG renders as a powerful tool in studying indoor PM-bound organics.

Hong Kong, one of the most densely populated metropolises in the world, is known for its small living space per capital. The lifestyle in Hong Kong is a fusion of Eastern and Western ones. Typical examples are the supply of both Chinese and Western foods in many restaurants. With a subtropical climate, Hong Kong is warm and humid throughout the year, despite relatively lower temperature and moisture in cool seasons, leading to widespread and long-duration use of air conditioning systems. Outdoor air pollution resulting from local sources (e.g., vehicle exhausts) and regional transport in cool seasons is also significant (Yim et al., 2009; Chan and Chung, 2003). Previous studies in Hong Kong investigated the concentrations, emissions and sources of criteria air pollutants and volatile organic compounds indoors, covering the prevalent housing types and common residential activities (Guo, 2011; Guo et al., 2009; Lee et al., 2002). A handful of studies have focused on speciated PM-bound organics (Chen et al., 2020; Deng et al., 2016; Lui et al., 2016). However, the reported species were confined to several categories of organics with known health risks, such as polycyclic aromatic hydrocarbons (PAHs) and carbonyls. Moreover, the analysis was limited by the low time resolution of the data associated with long-time-span sampling methods.

In this study, we measured total and speciated PM-bound organics for ~1 month in a Hong Kong residence, where prescribed activities were carried out. The application of TAG and Aerosol Mass Spectrometer (AMS) significantly improved the resolution of the measurements in time and chemical information (distinguishable at molecular level), compared to previous studies. An earlier paper gave an overview of the measurements, together with air pollutants in gas phase (Lyu et al., 2021). With a special focus on PM-bound speciated organics, here we discussed their high levels indoors and emissions from prescribed activities. Furthermore, the dynamic source contributions of indoor activities to PM-bound OM were quantified.

2. Methods

2.1. Sampling campaign and data processing

An intensive sampling campaign was conducted in a rented residential apartment where the instruments were deployed, and the prescribed activities were carried out. The apartment was

in a residential area of Kowloon, Hong Kong, with a saleable area of 25.4 m^2. It consisted of 1 living room, 2 bedrooms, 1 washroom and 1 kitchen. Close to the apartment were a highway, several streets, dozens of restaurants, many funeral service workshops, two funeral parlors, and a waste recycling station. Figure S1 shows the location and layout of the apartment.

The air changes per hour (ACH) were measured in 4 scenarios which were prevalent throughout the sampling campaign. It turned out that the ACH was relatively stable (1.1-1.2 hr^{-1}) except when the range hood was turned on (2.9 hr^{-1}) (Table S1). All the instruments were placed in the living room, and collected indoor and outdoor samples alternatively every other hour. Pre-cleaned copper tubing with a length of 2-3 m was used as the sampling lines to reduce the loss of charged particles. To mimic real situations in ordinary Hong Kong homes, we designed and carried out indoor activities that are common in Hong Kong, such as cooking, incense burning, candle burning, cigarette smoking, cleaning and makeup. Incenses and candles are usually burned in ritual activities in South China including Hong Kong, when people worship ancestors, gods and ghosts at home. Additional details about the sampling campaign are provided in Text S1 and were elaborated in the earlier overview paper (Lyu et al., 2021).

TAG and AMS were employed to chemically characterize the PM-bound organics. Figure 1 presents several examples of the total ion chromatogram (TIC) obtained from TAG in this campaign. The numerous TIC peaks implied a complex mixture of hundreds to thousands of species detected. Based on the retention time and mass spectra, 85 compounds with relatively large peak areas, whose volatilities were equivalent to those of C$_{16}$-C$_{36}$ n-alkanes, were identified in 461 valid samples. Further, the concentrations were quantified for 55 out of the 85 species using authentic or surrogate standards (Table S2), and the changes in instrument sensitivity over time were corrected using internal standards (IS). Details are given in Text S2. The composition of non-refractory submicron particulate matters, including sulfate, nitrate, ammonium, chloride and organics (total and fragmented), was analyzed by the AMS. The TAG and AMS data had a time response of 1 hour and 4 minutes, respectively. To facilitate discussion, the highly time-resolved AMS data were converted to hourly averages where necessary. The operation procedures and calibrations of TAG and AMS are described in Texts S2-S3, while more information about the instruments is available in Lyu et al. (2020).

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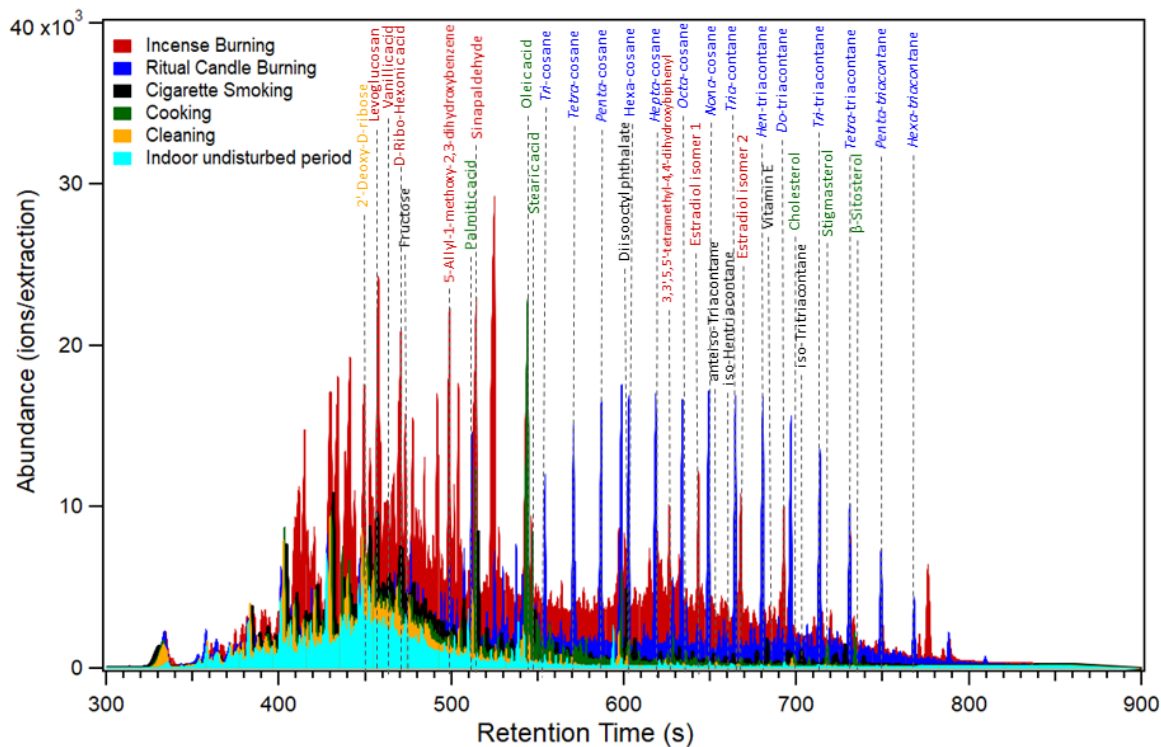
Figure 1. Example of TIC for samples collected during disturbed and undisturbed periods. Some
representative species emitted from indoor activities are labeled.

2.2. Source apportionment model and configurations

Positive matrix factorization (PMF) analysis was performed to determine the source
contributions to PM-bound organic matters (OM) indoors. The measured concentrations of
individual/lumped organics in $PM_{2.5}$ (particulate matters with aerodynamic diameters not larger
than 2.5 μm) and some non-refractory (NR) compositions of PM_1 (analogous to $PM_{2.5}$ but the
cut size is 1 μm) were input into the model. Linoleic acid, which was not quantified, was also
input to better isolate the sources of cooking emissions. For this special case, the IS-scaled
abundances were used as substitutes of absolute concentrations. The concentration matrix was
composed of 13 individual or lumped species in 181 samples, 89 and 92 out of which were
collected during disturbed and undisturbed periods, respectively. The discrepancy in particle
size the data bounded was due to the fact that AMS had very low collection efficiencies for
particles larger than 1 μm (Text S1). As discussed in Text S1, we assumed that the organic
compositions of PM_1 were comparable to those of $PM_{2.5}$, so that the $PM_{2.5}$-bound organic
tracers could be used to apportion the OMs in PM_1 (termed as PM_1-OM hereinafter). Similar
assumptions were adopted in previous studies (Williams et al., 2010).

The uncertainties were set following the principles introduced in Text S4. Due to intensive
emissions of some activities, extremely high concentrations (or IS-scaled abundances) were
occasionally observed for some species. The model was unable to reproduce some of the
extreme values, especially for the data points whose scaled residuals were larger than 4. To
prevent the model from treating them as outliers, a much lower error fraction (1% for these

169 values vs. 10% for others) was used when we calculated the uncertainties. The mathematical
170 effect of this treatment was to add weights onto the extremely high values in PMF iteration
171 process (Wang et al., 2017). More information about the PMF modelling is available in Text
172 S4.

173

3. Results and Discussion

3.1. High levels of PM-bound organics indoors

176 The chemical compositions of indoor and outdoor NR-PM$_1$ measured by AMS are shown in
177 Figure 2a. Noticeably, PM$_1$-OM dominated the compositions for both indoor and outdoor
178 samples, accounting for 82.7±0.3% and 57.6±0.05% of NR-PM$_1$, respectively. The higher
179 fraction of organics in indoor NR-PM$_1$ was in line with the results of previous studies (Perrino
180 et al., 2016; Cao et al., 2012). On one hand, the formation of secondary inorganic aerosol (SIA)
181 species, including sulfate, nitrate and ammonium, is generally more efficient in outdoor
182 atmosphere (Talbot et al., 2016; Lunden et al., 2003). On the other hand, large amounts of
183 condensable organics are emitted indoors (Kristensen et al., 2019). Here, the average
184 concentration of indoor PM$_1$-OM (21.1 ± 6.6 μg/m^3) was approximately twice as high as that
185 outdoors (10.3 ± 0.7 μg/m^3) ($p<0.05$), in contrast to the lower SIA concentrations indoors
186 (4.4±0.2 μg/m^3 vs. 7.6±0.3μg/m^3).

187 Further, we found that the higher levels of PM$_1$-OM in the apartment were primarily caused by
188 the emissions of occupant-related activities. As presented in Figure 2b, the concentrations of
189 indoor PM$_1$-OM during undisturbed periods were comparable to or even lower than the outdoor
190 concentrations. The diurnal cycles on one day when there was no occupant in the apartment
191 indicated that the indoor and outdoor PM$_1$-OM followed the same pattern (Figure S2), which
192 was an obvious sign of outdoor air infiltration. However, during disturbed periods, the indoor
193 PM$_1$-OM levels were substantially elevated by some activities. For example, cigarette smoking
194 at 14:00 on 1 Dec. led to the soaring of PM$_1$-OM up to 454.1 μg/m^3. We adopt the metrics of
195 net increment (NIC) to stand for the concentration increase, which was calculated in the same
196 way as that introduced in Lyu et al (2021). On average, the NIC of PM$_1$-OM in the samples
197 influenced by cigarette smoking, incense burning, cooking and ritual candle burning was 115 ±
198 85.6, 93.0 ± 84.8, 38.0 ± 20.3 and 28.1 ± 20.8 μg/m^3, respectively. The variations of NIC for
199 same activities were mainly attributable to the differences in activity strengths, e.g., simple
200 breakfast vs. big dinner. It is interesting to note that the concentrations of PM$_1$-OM in the
201 disturbed scenario decreased significantly at 10:00, 16:00 and 20:00, compared to the much
202 higher concentrations 2 hours earlier. Although it seemed to be explained by the dominance of
203 low-emission activities (emission here specifically referred to emissions of PM-bound organics;
204 same for the high-emission activities elsewhere) in these hours, such obvious reductions in the
205 concentrations within 2 hours indicated quick removal of PM$_1$-OM from the indoor air.

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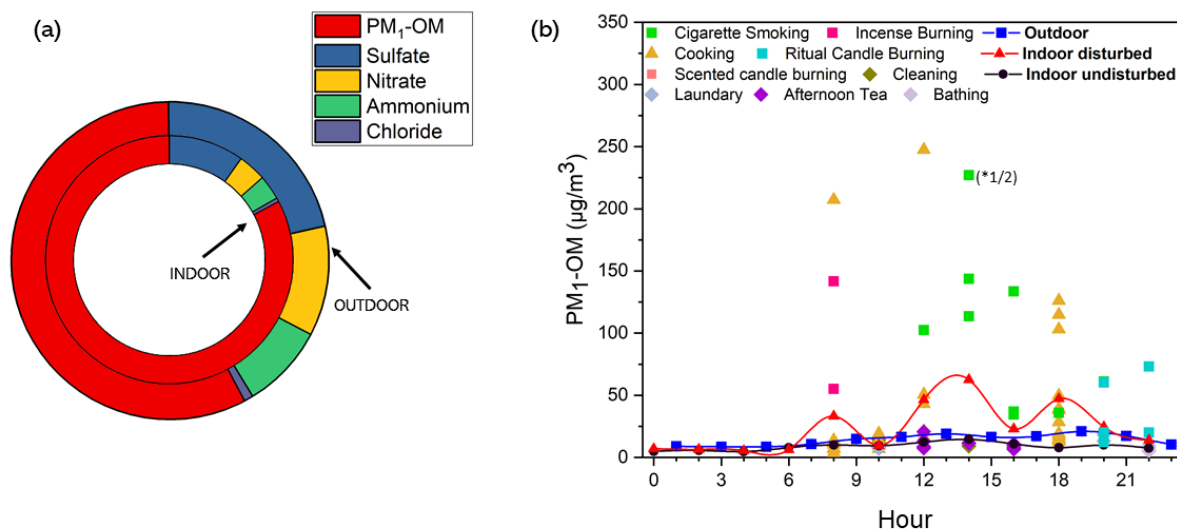
Figure 2. Compositions of indoor and outdoor NR-PM$_1$ (a); and average diurnal cycles of PM$_1$-OM in different scenarios and PM$_1$-OM levels in the presence of individual indoor activity (b).

Figure 3 shows the I/O ratios of speciated organics in PM$_{2.5}$ and NR-PM$_1$ compositions. Although a previous study (Lyu et al., 2021) overviewed the I/O ratios of air pollutants including PM-bound organics collected in this sampling campaign, here the discussion on individual rather than lumped species with more metrics (not only mean values) gave a broader insight into the origins of specific compounds of interest. The species were classified into 3 groups: Group A with both the median and mean I/O ratios lower than 1; Group B with both the median and mean I/O ratios higher than 1; and Group C with the median I/O ratio lower than 1 while the mean higher than 1.

Group A comprised the SIA components, most of the secondary organic aerosol (SOA) species, some biomass burning tracers and PAHs. The indoor concentrations of these species were dominated by outdoor emissions and chemistry. It was not surprising that these SIA and SOA species had the I/O ratios lower than 1, because the generally higher oxidative capacity of ambient air is more favorable for their production. The higher outdoor concentrations of biomass burning tracers, i.e., levoglucosan and mannosan, were not discovered by examining the overall I/O ratio of saccharides (2.57 ± 2.11) in Lyu et al. (2021). Note that both species were detected in the emissions of some indoor activities, e.g., incense burning and cigarette smoking (section 3.2). The even higher outdoor concentrations were likely associated with the intensive funeral services nearby which combusted the worship supplies made of paper and bamboo. However, the occupant-related activities indeed resulted in higher indoor concentrations for levoglucosan and mannosan sometimes (Figure S3), hence a few data points with I/O ratios higher than 1 in Figure 3. This phenomenon was observed for most of the Group A species, including PAHs and even the species that are generally thought to be secondarily formed.

Group B contained iso-nonacosane, anteiso-tritriacontane, cholesterol, β-sitosterol and 9-oxononanoic acid, which were mainly of indoor origins. As discussed in section 3.2 below, the indoor cholesterol was primarily derived from cooking emissions, while β-sitosterol was detected in the emissions of multiple activities (incense burning, cigarette smoking and cooking). Cigarette smoking could bring the mean I/O ratios for iso-nonacosane and anteiso-tritriacontane (tracers of tobacco combustion) to above 1. However, the median values of higher than 1 also resulted from the low concentrations of both species outdoors, because only 9 indoor samples were influenced by smoking. As an aging product of cooking emissions, 9-oxononanoic acid exhibited consistently higher levels indoors driven by heterogeneous processes (Lyu et al., 2021).

Approximately 56% of the total number of species were in Group C, for which the median and mean values of I/O ratios were stratified with 1 as the threshold value. The median of less than 1 indicated the dominance of outdoor emissions and chemistry in the sources of these species indoors for most samples, especially for those collected during undisturbed periods. However, the few samples contaminated by intensive emissions of occupant-related activities played determinant roles in increasing the mean values of I/O ratios to above 1. All in all, the I/O ratios demonstrated high levels of a large number of speciated organics in indoor $PM_{2.5}$, as a result of the emissions of occupant-related activities and subsequent reactions (e.g., heterogeneous production of 9-oxononanoic acid).
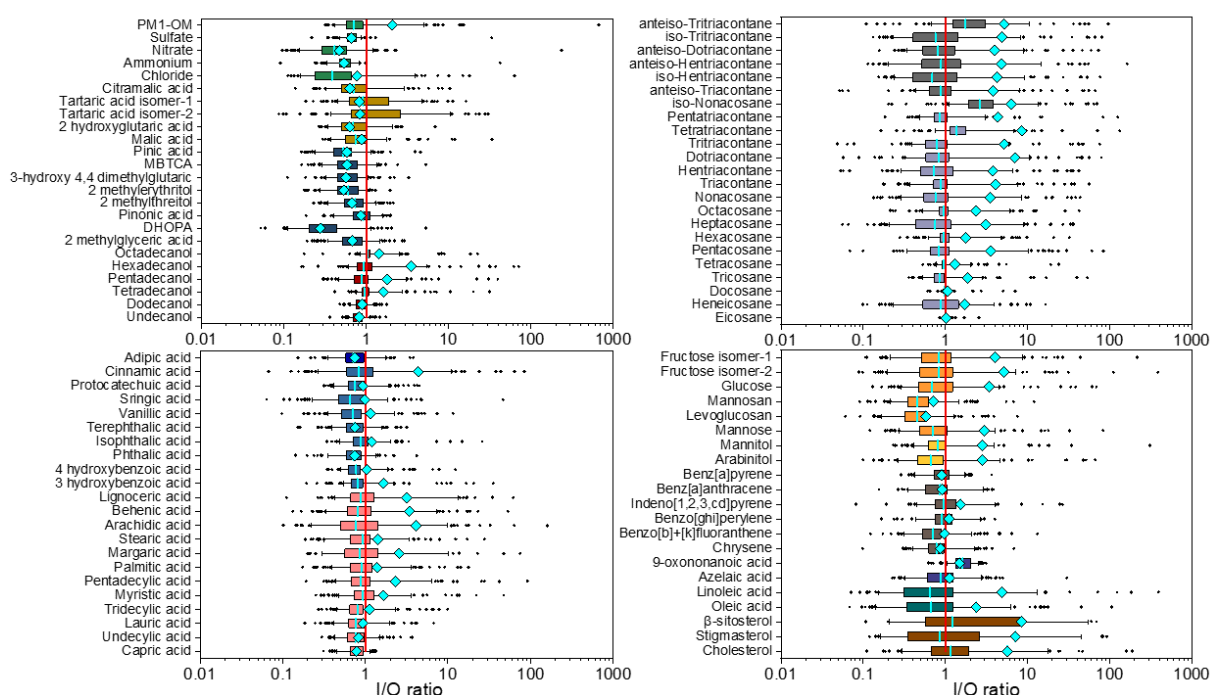
Figure 3. Box and whisker plots of I/O ratios for speciated organics and NR-PM_1 compositions. The far right and far left of the box are 75th and 25th percentiles, respectively, and the line in the center of the box represents the median. The tip of the right (left) whisker denotes 95th (5th) percentile. Blue diamonds

stand for mean values of I/O ratios, and the black dots are for values beyond the range of 5th–95th percentiles.

3.2. Emission characteristics and profiles

To further understand the emissions of speciated PM-bound organics, we calculated the NICs and enhancement ratios (ERs) for the organic species in $PM_{2.5}$ during individual indoor activities, as summarized in Table S3. The ERs were calculated as the ratios of average concentrations (or IS-scaled abundances for unquantified species) in presence of activity to those in absence of activity at the same time slots (Lyu et al., 2021). The total NIC of quantified speciated organics followed the same order as NIC of PM_1-OM, i.e., cigarette smoking (1776 ng/m^3) > incense burning (1131 ng/m^3) > cooking (463 ng/m^3) > ritual candle burning (396 ng/m^3). Marginal increases were observed for the concentrations of PM-bound organics during scented candle burning, for which the NICs were hardly computable. The other activities did not lead to significant changes in the concentrations (or IS-scaled abundances) of the speciated organics. Therefore, we only obtained the emission profiles of speciated organics for the high-emission activities, which are expressed as the fractions of compound-specific NICs in total NIC in Figure 4. The acquisition of the emission profiles was based on the assumption that all the organic species underwent the same rates of physical and chemical decays within <1 hour after they were emitted.

Cigarette smoking was characterized by high emissions of fructose isomers (no. 54 & no. 55 in Figure 4 and Table S2, same for the no. elsewhere). It was reported that saccharides, including fructose, were commonly emitted from burning of plant materials or fragments (Jansen et al., 2014; Pacini et al., 2000). The concentrations of odd *n*-alkanes in the carbon range of C_{27}-C_{33}, vanillic acid, levoglucosan, arabinitol and mannitol were also enhanced during smoking. Iso-/anteiso-alkanes are usually regarded as tracers of cigarette burning because of their widespread presence in tobacco leaf waxes (Kavouras et al., 1998). In this study, they were identified from the TAG chromatograms but were not quantified due to the lack of standards. Indeed, the IS-scaled abundances of the C_{27}-C_{33} iso-/anteiso-alkanes were remarkably enhanced during smoking, with the ERs of 46-195 (Table S3). The other unquantified organics with high ERs included stigmasterol, glucose, β-sitosterol and mannose, some of which have been introduced in Lyu et al. (2021). Interestingly, a well-shaped peak of vitamin E was identified from the TAG chromatograms disturbed by smoking, which was not found during undisturbed periods and thus the ER was not computable. It is unclear whether the vitamin E was an additive specific to the cigarette used in this study or ubiquitously present in cigarettes.

With the application of AMS, previous studies (Li et al., 2012; Ji et al., 2010) detected the ion fragments of hydrocarbons, PAHs, and lignin-related compounds emitted from incense burning. However, the emissions of speciated organics were inadequately characterized, though a handful of studies (e.g., Lui et al., 2016) focused on the species of health relevance, such as

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carbonyls, PAHs and oxygenated PAHs. Here, we found that vanillic acid (no. 41) and syringic acid (no. 43), two typical markers of lignin pyrolysis (Hyder and Jonsson, 2012; Simoneit et al., 1993), were the most distinguishable species in incense burning emissions, accounting for 58.1% and 17.9% of the total NIC, respectively. In spite of the lower NICs, levoglucosan, arabinitol and oleic acid also experienced significant rise in concentrations during incense burning. Besides, β-sitosterol, stigmasterol and linoleic acid, which were not quantified, had the ER of 122, 105 and 90, respectively. The ERs were on the same magnitude as that for vanillic acid (108), indicating strong emissions of both plant sterols and linoleic acid (abundant in many plants, e.g., safflower, sunflower and corn). It is noteworthy that incense burning also brought about the highest NICs and ERs for PAHs among all the activities (Table S3), indicating potential impacts of this ritual activity on health, which was demonstrated before (Zhou et al., 2015).

Chinese cooking involves various ingredients and methods. As a result, cooking emissions contain a wide range of PM-bound organics, such as saturated and unsaturated fatty acids, sterols, saccharides, and carbonyls (Zhao et al., 2007; Robinson et al., 2006; Schauer et al., 2002), and the emission profiles are not necessarily consistent. Among the 31 cooking activities carried out in this campaign, it was realistic to separate two types of emission profiles (Figures 4c-4d). Oleic acid (no. 46), palmitic acid (no. 44), arabinitol (no. 50) and fructose isomers (no. 54 & no. 55) were the typical tracers in both profiles. The difference was that cholesterol (no. 47) constituted a considerable fraction (10.5%) of the total NIC of quantified organics in one profile (Figure 4c), while it was almost negligible in the other one (Figure 4d). The former with higher fraction of cholesterol represented emissions of PM-bound organics from cooking fishes and/or eggs, when elevated NICs were observed for cholesterol. In addition, some unquantified species were identified with high ERs during cooking, such as linoleic acid, arachidic acid and β-sitosterol (Table S3). It is noteworthy that the IS-scaled abundances of linoleic acid were greatly enhanced during pan frying and deep frying, while frying vegetables caused slight enhancements of cinnamic acid. The representative species identified here were broadly in line with those reported in literature (Abdullahi et al., 2013; To et al., 2000). Here, discovering the different emission characteristics depending upon cooking ingredients and methods will contribute to more comprehensive understanding of cooking emissions.

The emissions of ritual candle burning were recognizable due to the moderate to high NICs of C_{22}-C_{35} n-alkanes (no. 16 – no. 29), accounting for ~30% of the total NIC (Figure 4e). The NICs of individual n-alkanes correlated well with each other among the repeated 6 activities (R^2>0.8). In comparison with the emission factors of n-alkanes for burning of paraffin candles tested in a sealed chamber (Fine et al., 1999), the relative emission strengths were comparable for C_{22}-C_{28} n-alkanes between the two studies (Table S4). However, our measurements indicated higher proportions of C_{29}-C_{35} in the total emission of n-alkanes, which might be associated with the smoldering of the bamboo stick inside the candle. Coincidently, the concentrations of some biomass burning tracers, such as vanillic acid (no. 41) and levoglucosan

334 (no. 53), were enhanced. Besides, palmitic acid, oleic acid, arabinitol and fructose isomers also
335 occupied large shares in the total NIC, likely resulting from the combination of emissions from
336 the burning of paraffin and bamboo stick.

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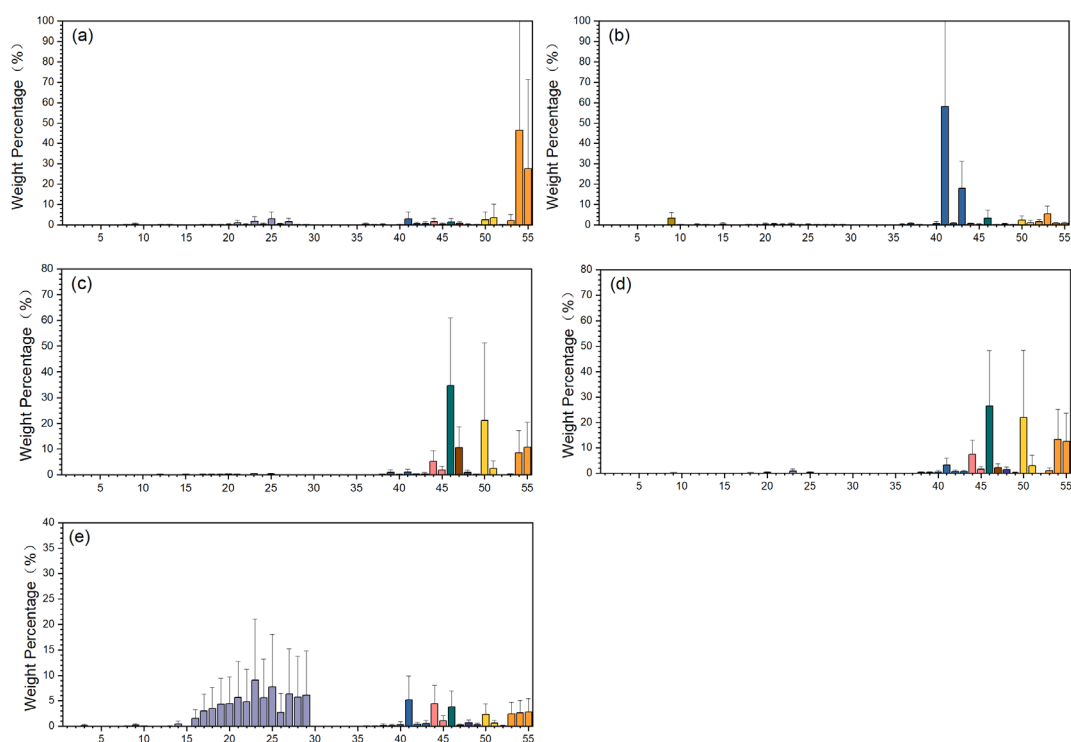
Figure 4. Emission profiles of $PM_{2.5}$ organics of incense burning (a), ritual candle burning (b), cigarette smoking (c), cooking-1 (d), and cooking-2 (e). Error bars represent 95% confidence intervals. Numbers on the horizontal axes correspond to specific organic species that are listed in Table S2.

341 It is inevitable that any emission profile is dependent upon the number and species of
342 compounds included. In this study, though some compounds with high ERs were not compiled
343 into the profiles due to unavailability of the NICs, the emission profiles composed of 55 organic
344 compounds are chemically comprehensive enough to guide source apportionments. Moreover,
345 we emphasized the unquantified species with high ERs in the above discussion. These species
346 can also be references for source identification. Value of the emission profiles and additional
347 representative species is demonstrated in the following section.

348

3.3. Contributions to PM_1-OM of high-emission activities

350 As stated in section 2.2, PMF was utilized for source apportionment of indoor PM_1-OM. To
351 determine the optimal number of factors, we preliminarily examined the ratio of $Q/Qexp$, an
352 index reflecting modelling residuals. Usually, the least number of factors is the one above which
353 the $Q/Qexp$ ratio decreases less dramatically with the increase of factor number (Wang et al.,
354 2017). Figure S4 presents the $Q/Qexp$ ratio against the factor number. Obviously, the decline

355 of *Q/Qexp* became mild after the number of factors increased to 7. Therefore, the model was
356 run with the factor numbers of 7 and above. Eventually, a 7-factor solution was adopted,
357 because at least 1 factor could not be explained in the tests with more factors, based on the
358 known emission profiles or the real occurrences of the prescribed activities. Statistics of linear
359 regressions between the observed and PMF-modelled concentrations (or IS-scaled abundances
360 for linoleic acid) indicated that the observed values were well reproduced by the model (Table
361 S5).

362 Figure S5 shows the chemical profiles of the 7 factors. Based on the emission characteristics of
363 prescribed activities discussed in section 3.2, the sources of cigarette smoking, incense burning
364 and ritual candle burning were liable to be identified. The discrepancies in the profiles between
365 the measurements and PMF solution were mainly due to the fact that the PMF-resolved profile
366 of a specific source was influenced by other factors. For example, vanillic acid, oleic acid and
367 fructose isomers accounted for large proportions in the observed emission profile of ritual
368 candle burning (Figure 4e), while the much higher emissions of these species from other sources
369 (e.g., incense burning, cooking and smoking) made them almost negligible in the PMF-resolved
370 profile. Further, we looked into the other 4 factors and defined 3 of them as cooking emissions,
371 in view of the exclusively high loadings of oleic acid, cholesterol and linoleic acid, respectively.
372 The separation of cooking emissions into 3 factors was likely a result of different cooking
373 ingredients and/or methods, as clarified above. At last, with the dominance of SIA components,
374 factor 7 represented outdoor air infiltration. Because of the weak or even zero emissions, the
375 other activities, such as makeup, cleaning, shower and laundry, were not identified as sources
376 of PM_1-OM. However, it does not mean they had little impact on indoor air quality. The
377 emissions of volatile organics caused by some of these activities have been discussed in Lyu et
378 al. (2021).

379 Benefited from the fast response data generated from AMS and TAG, we were able to get the
380 dynamic contributions of individual sources to PM_1-OM, as plotted in Figure 5a. All the spikes
381 coincided with the real occurrences of high-emission activities, again suggesting significant
382 impacts of occupant-related activities on the loadings of PM-bound organics in indoor air.
383 Figure 5b summarizes the average source contributions during the disturbed and undisturbed
384 periods. In the presence of high-emission activities, cigarette smoking and cooking led the
385 contributions to PM_1-OM, accounting for 31.4% and 31.2%, respectively. The relatively lower
386 contribution of incense burning to PM_1-OM (12.7%) was mainly due to the fact that it was only
387 performed twice. Ritual candle burning made a contribution of 5.9%, and the rest (18.9%) was
388 attributable to outdoor air infiltration. During the undisturbed periods, it was striking that
389 outdoor air infiltration was responsible for 77.1% of PM_1-OM, while only 13.0%, 7.1% and
390 2.8% of PM_1-OM was attributable to the residues of emissions from incense burning, cooking
391 and ritual candle burning, respectively. In fact, the mass concentrations of PM_1-OM built up by
392 outdoor air infiltration were comparable during the disturbed (5.53 μg/m³) and undisturbed
393 periods (5.97 μg/m³), while those assigned to all the other sources decreased noticeably during

the undisturbed periods. Despite this, incense burning still contributed 1.01 μg/m^3 to PM$_1$-OM, which was ~27% of that during disturbed periods (3.72 μg/m^3). On one hand, this implied the re-suspension of residual organics from indoor reservoirs (e.g., walls) where the incense burning emissions might be deposited. Similar effects of this process on some specific organics indoors have been revealed by previous studies (Lunderberg et al., 2020; Wang et al., 2020). On the other hand, it was likely that incense burning identified during undisturbed periods represented some outdoor sources which were independent from the variations of SIA. Rationality of the latter speculation came from the intensive ritual activities in the area where the apartment was located.

It should be noted that the source contributions determined from this single campaign with designed indoor activities do not necessarily reflect the average or universal situations in Hong Kong, though the activities were carried out with general intensities and frequencies. Bearing the uncertainties in mind, we still would like to remind of the great contributions of cigarette smoking and cooking to PM-bound organics indoors. Besides, the intensive emissions from incense burning could also be a serious problem, e.g., causing acute health effects. Outdoor air infiltration played a dominant role in influencing indoor air quality in absence of high-emission activities. Note that the ACH was 1.1-1.2 hr^{-1} for this apartment in most of the time, lower than that for many other indoor spaces with mechanical ventilation (Lai et al., 2020b). Therefore, this finding sends a clear message that indoor air quality can benefit from tackling ambient air pollution.
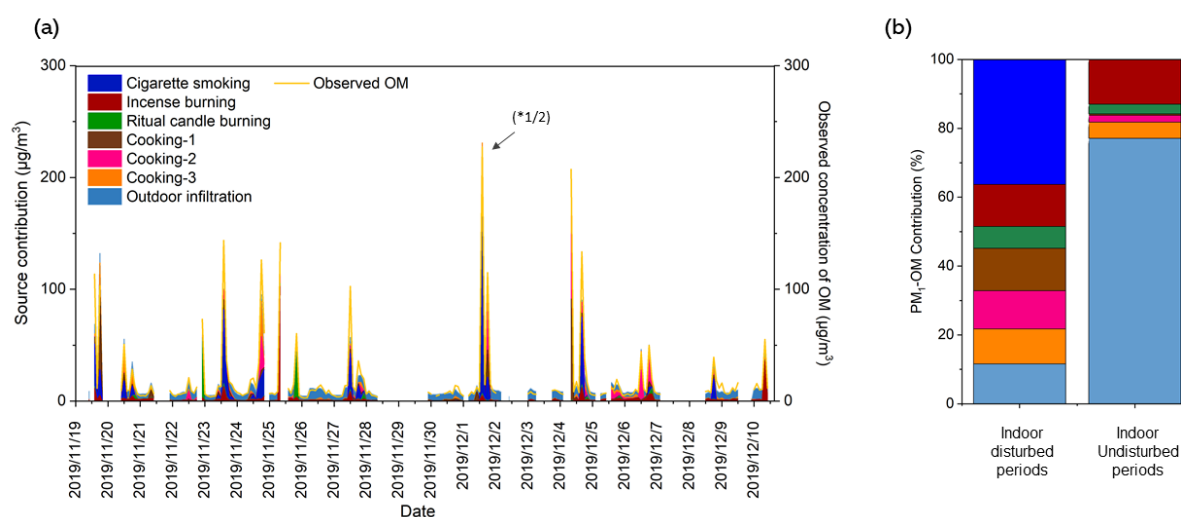
Figure 5. Time series of PM$_1$-OM attributable to individual sources (a) and average source contributions during disturbed and undisturbed periods (b). Missing data are mainly caused by instrument maintenance.

13

4. Conclusions

Condensable organics are ubiquitous in almost all types of indoor environments, considerable fractions of which are partitioned to fine PM. Indoor PM-bound organics have great complexities in chemical compositions and health effects. This study was conducted in a compact flat with subdivided units in Hong Kong, representative of flats for low- and middle-income families in the city. Even people cook, smoke and burn worship supplies at home, indoor air pollution has not attracted sufficient attention compared to other hazards in such flats (e.g., hygiene and fire safety). With the introduction of novel analysis techniques initially developed for atmospheric chemistry study, we measured the total and speciated PM-bound organics in the flat at higher-than-before resolutions in both time and chemical information. Higher indoor (than outdoor) levels were observed for the total PM-bound organics and large number of speciated organic compounds in $PM_{2.5}$. Such phenomenon was mainly caused by the emissions of occupant-related activities. Cigarette smoking, cooking, incense burning and ritual candle burning were the top four activities with respect to the emissions among all the activities we carried out. We made the chemically comprehensive emission profiles of speciated organics available for these activities. Recognizable tracers were identified, including but not limited to fructose isomers for cigarette smoking, vanillic acid for incense burning, oleic acid, cholesterol and linoleic acid for cooking with different ingredients or methods, and C_{22}-C_{35} alkanes for ritual candle burning. The fast response data allowed us to determine the dynamic source contributions to PM_1-OM. While cigarette smoking and cooking were the main contributors during disturbed periods, an overwhelming proportion of indoor PM_1-OM was derived from outdoor air infiltration in absence of high-emission indoor activities. This study enhances current knowledge on emissions and sources of PM-bound speciated organics indoors, based on which the policy implications are evident. On one hand, occupant-related activities must be properly managed, especially in old tenement buildings where the high-emission activities are carried out without any caution. On the other hand, control of outdoor air pollution will benefit indoor air quality, and effective filtration should be applied in regions with severe outdoor air pollution.

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