# 1 Molecular fingerprints and health risks of home-use incense burning

# 2 smoke

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28 **Abstract:** The burning of incense for home use is a widespread practice that has been shown to have 29 significant negative impacts on human health and air quality. However, there is a lack of 30 understanding regarding its emission profiles and associated health risks. To address this knowledge 31 gap, we utilized a state-of-the-art thermal desorption comprehensive two-dimensional gas 32 chromatography-mass spectrometer (TD-GC×GC-MS) to (semi-)quantify the emission factors (EFs) 33 of 317 volatile compounds and thoroughly investigate the organic profiles of incense burning smoke across a full-volatility range. Results showed that toluene  $(70.8 \pm 35.7 \ \mu g \ g^{-1})$  is the most abundant 34 35 compound in incensing-burning smoke, followed by benzene, furfural, and phenol. Phenol, toluene, 36 furfural, 2-furanmethanol, benzene, and benzyl alcohol are the main contributors to ozone and 37 secondary organic aerosol (SOA) estimation. Intermediate volatility organic compounds (IVOCs) 38 accounted for 19.2% of the total EFs, but 40.0% of the estimated SOA. Additionally, a novel 39 pixel-based method, combined with aroma analysis, revealed that furfural can act as a key tracer of incense burning, and is responsible for the distinctive flavor of incense smoke. High bioaccumulation 40 41 potential (BAP) assessment using pixel-based partition coefficient estimation revealed that 42 acenaphthylene, dibenzofuran, and phthalate esters (PAEs) are chemicals of high-risk concern and 43 warrant further control. Our results highlight the critical importance of investigating home-use 44 incense burning and provide new insights into the health impacts of incense burning smoke by novel 45 approaches.

#### 47 **1 Introduction**

48 Incense burning is a prevalent custom in many cultures, especially in East and Southeast Asia 49 (Chen et al., 2021). In modern times, incense burning for fragrance has become a frequent practice in 50 households (Manoukian et al., 2013), while functional incense burning, such as mosquito coils, is 51 used for specific purposes. Exposure to incense smoke is linked to adverse health effects like eve 52 irritation, carcinogenicity, genotoxicity, and respiratory system damage (Wong et al., 2020; Yang et 53 al., 2007, 2017). Incense is composed of fragrant materials, aromatic woods, herbs, and adhesive 54 powders, usually available in the form of sticks and coils (Wong et al., 2020; Yadav et al., 2022). 55 Incense burning releases multiple pollutants into the air, including particulate matter (PM), carbon 56 monoxide (CO), volatile organic compounds (VOCs), and intermediate volatility/semi-volatile 57 organic compounds (I/SVOCs) (Wong et al., 2020; Yang et al., 2007; Jetter et al., 2002).

58 Current studies mainly focus on the hazardous VOC and SVOC homologs released from 59 incense-burning smoke. For instance, Lee et al. investigated 8 carbonyls and 11 VOCs emitted from 60 incense burning and found that the emission factors (EFs) of traditional incense burning were higher than aromatic incense (Lee and Wang, 2004). Lu et al. detected 230 kinds of VOCs from 61 mosquito-repellent incense burning, elucidating that alkanes, esters, aldehydes, ketones, and 62 63 aromatics are predominant (Lu et al., 2020). Staub et al. measured 6 methoxy phenolics, 10 64 monoterpenoids, and other 21 kinds of SVOCs in the burning smoke of incense sticks, and identified 65 cedrol as an important odor source (Staub et al., 2011). However, most of the studies have focused on VOC compounds, with less attention given to gaseous organics in the full volatility range 66 67 (VOC-IVOC-SVOCs). A full-volatility organic characterization may better evaluate the ozone 68 formation potential (OFP) and SOA formation, as I/SVOCs are potentially important precursors of 69 ozone and secondary organic aerosol (SOA) formation (Zhao et al., 2007; Tang et al., 2021; Guo et 70 al., 2014, 2020). Meanwhile, mapping organics from incense smoke helps to evaluate the potential 71 health risks of toxic compounds.

Comprehensive two-dimensional gas chromatography (GC×GC) is a powerful technique dealing
 with the coelution problem in conventional one-dimensional gas chromatography (1D GC).
 Pollutants from gasoline exhaust, diesel exhaust, and cooking emissions are well separated and

identified (Drozd et al., 2019; Alam et al., 2018; Song et al., 2022a). As much as 50 ~ 98% of the
total response in GC×GC chromatograms could be explained (Huo et al., 2021; Song et al., 2022b).
Previous work identified 324 compounds from incense smoke by coupling solid-phase
microextraction (SPME) with GC×GC, yet chemicals are not quantified (Tran and Marriott, 2007).
Thus, a non-targeted and quantitative assessment of incense burning emissions is currently lacking.

In this work, two types of incense sticks and three kinds of incense coils were burned in a steel chamber. Gaseous pollutants were trapped by Tenax TA desorption tubes and then analyzed by a thermal desorption comprehensive two-dimensional gas chromatography-mass spectrometer (TD-GC×GC-MS). Pixel-based multiway principal component analysis (MPCA) was utilized to identify markers of incense burning. Risk assessment of pollutants from incense burning emissions was then evaluated by pixel-based approaches and high-risk compounds related to incense burning were assessed.

#### 87 2 Methodology

#### 88 2.1 Sampling and instrumentation

89 Incenses were purchased from the market, including 4 common incense sticks, 2 Thailand 90 incense sticks, 1 mosquito coil, and 2 incense coils (Figure S1). Incenses could also be classified by 91 their material, containing 2 aromatic coils, 4 aromatic sticks, 1 mosquito coil, 1 sandalwood stick, 92 and 1 smokeless sandalwood stick (Figure S1). Incenses were burned in a stainless combustion chamber (1 m<sup>3</sup>). After ignition, the burning incense changed from flaming to smoldering. Each kind 93 94 of incense was burned at least twice. Incenses were weighed before and after combustion. 95 Preconditioned Tenax TA desorption tubes (Gerstel 6 mm 97 OD, 4.5 mm ID glass tube) were utilized to trap organics with a sampling flow of  $0.2 \text{ Lmin}^{-1}$ . 96

A comprehensive two-dimensional gas chromatography-quadrupole mass spectrometer (GC×GC-qMS, GC-MS TQ8050, Shimadzu, Japan) coupled with a thermal desorption system (TDS 3 C506, Gerstel, Germany) was used for sample analysis. The desorption temperature was 280 °C. The cooled injection system (CIS) with a Tenax TA liner was held at 20 °C and ramped to 320 °C once injecting the gaseous sample into GC columns. The column combination was SH-Rxi-1ms (1<sup>st</sup>, 30 m × 0.25 mm × 0.25  $\mu$ m) and BPX50 (2<sup>nd</sup>, 2.5 m × 0.1 mm × 0.1  $\mu$ m). The modulation period was 103 6s. See Table S1 and elsewhere (Song et al., 2022a) for more information.

#### 104 2.2 Chemical identification, quantification, and 2D binning

A series of standard mixtures (2 µg mL<sup>-1</sup>, 5 µg mL<sup>-1</sup>, 10 µg mL<sup>-1</sup>, 20 µg mL<sup>-1</sup>, 40 µg mL<sup>-1</sup> in 105 106 CH<sub>2</sub>Cl<sub>2</sub>) was injected into Tenax TA tubes (2 µL). After purging the solvent with nitrogen gas, the 107 standards were thermally desorbed. The standard mixture contains 26 n-alkanes (C7 - C32, CNW 108 Technologies, ANPEL Laboratory Technologies (Shanghai) Inc., China), 16 PAHs, 11 phenolic 109 compounds, 9 alcohols, 4 aldehydes, 8 aromatics, 24 esters, 7 ketones, 5 siloxanes, and 39 other compounds. Gaseous organics are quantified by external calibration curves with most of the  $R^2$ 110 111 between 0.95 and 0.999 (Table S2). Chemicals with the same retention times and mass spectrums 112 were directly qualified and quantified. The unidentified chemicals were qualified by matching their 113 mass spectrum with library spectrums in the National Institute of Standard Technology library (NIST 114 17). Reverse factors of more than 700 were acceptable in this work. As homologs on the 115 two-dimensional chromatogram (contour plot) were eluted with near-equal one-dimensional intervals, 116 chemicals were then qualified by combing the location of the contour plot and the mass spectrums 117 (Song et al., 2023). Compounds without standards were semi-quantified by *n*-alkanes from the same 118 volatility bin (uncertainty 69%) and surrogates from the same chemical class (uncertainty 27%). 119 Instrument detection limits (IDLs) for organics semi-quantified were unknown, as a result, chemicals 120 with negative values calculated by calibration curves were quantified by the volume-to-mass (ng) 121 ratio of the lowest quantification point of standards (Table S2). A total of 317 chemicals were 122 (semi)-quantified, including 10 acids, 34 alcohols, 19 aldehydes, 25 aromatics, 38 esters, 49 ketones, 123 18 *n*-alkanes, 26 nitrogen-containing compounds, and 10 phenols (Table S3).

The compounds identified were sliced into two-dimensional bins (2D bins) (Song et al., 2022a).  $1^{st}$  retention times are linked to the volatility of species (B8 to B31 with decreasing volatility) while  $2^{nd}$  retention times are associated with polarity (P1 to P8 with increasing polarity). Emission factors of compounds in the same 2D bin were aggregated (Table S3).

# 128 2.3 Emission factor (EF), ozone formation potential (OFP), and secondary organic aerosol

- 129 (SOA) estimation
- 130 The emission factor (EF,  $\mu g g^{-1}$ ) was calculated by the following equation:

$$EF = \frac{mV}{ftM} \tag{1}$$

m is the absolute mass of pollutants ( $\mu$ g) captured by Tenax TA tubes. V is the volume of the steel chamber (1 m<sup>3</sup>). The sampling flow and duration of the Tenax TA tube are f (0.0002 m<sup>3</sup> min<sup>-1</sup>) and t (min), respectively. M is the combustion mass (g) of the incense. The sampling volume of Tenax TA tubes (0.003 ~ 0.01 m<sup>3</sup>) was significantly smaller than the total volume of the steel chamber (1 m<sup>3</sup>) and the volume change of the chamber could be neglected.

The ozone formation potential (OFP,  $\mu g g^{-1}$ ) was calculated using equation (2). EF<sub>i</sub> is the 137 emission factor of precursor i ( $\mu g g^{-1}$ ) with maximum incremental reactivity (MIR) of MIR<sub>i</sub>. The 138 FOQAT 139 OFP was calculated inside the packages developed by Tianshu Chen 140 (https://github.com/tianshu129/foqat). The MIR used in this work can be found in Table S3.

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$$OFP = \sum [EF_i] \times MIR_i$$
(2)

142 Secondary organic aerosol (SOA) was estimated by equation (3).

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$$SOA = \sum [EF_i] \times (1 - e^{-k_{OH,i} \times [OH] \times \Delta t}) \times Y_i \quad (3)$$

Where  $k_{OH,i}$  and  $Y_i$  represent the OH reaction rate and SOA yield of precursor *i*, respectively (Table S3). The SOA yields of precursors were from literature (Loza et al., 2014; Harvey and Petrucci, 2015; Tkacik et al., 2012; Shah et al., 2020; McDonald et al., 2018; Chan et al., 2010, 2009; Wu et al., 2017; Li et al., 2016; Matsunaga et al., 2009; Algrim and Ziemann, 2019, 2016; Liu et al., 2018; Charan et al., 2020) or surrogates from *n*-alkanes in the same volatility bins (Zhao et al., 2017).  $k_{OH}$  and *Y* could be found in Table S3.  $[OH] \times \Delta t$  is the OH exposure and was set to be 13 ×10<sup>10</sup> molecules cm<sup>-3</sup>s (24 hours in OH concentration of 1.5 ×10<sup>6</sup> molecules cm<sup>-3</sup>).

## 151 2.4 Pixel-based risk assessments of incense-burning pollutants

Octanol-air partition coefficient ( $K_{o-a}$ ), air-water partition coefficient ( $K_{a-w}$ ), and octanol-water partition coefficient ( $K_{o-w}$ ) were estimated by a linear free-energy relationship (LFER) model (Nabi et al., 2014; Zushi et al., 2019). Partition coefficients of chemicals are associated with their two-dimensional retention times (Song et al., 2022b). Chemicals with high bioaccumulation potential (BAP) are defined as contaminants with partition coefficients of ( $2 < \log K_{o-w} < 11$ ) and ( $6 < \log K_{o-a}$ < 12). See Zushi et al. (Zushi et al., 2019) for more information. The R source code was obtained from GitHub (https://github.com/Yasuyuki-Zushi).

#### 159 **3 Results and discussions**

#### 160 **3.1 Emission profiles of different incense-burning organics**

161 Figure S2 is a typical chromatogram of incense burning emissions, which is also set as the 162 reference chromatogram during the pixel-based analysis. As much as 90.2% of the total percent 163 response could be explained. The ratio is similar to a recent study resolving biomass burning emissions (98%) (Huo et al., 2021). The emission factor (EF) of total organics is 791.8  $\pm$  300.6 µg g<sup>-1</sup>, 164 consistent with previous work (100 ~ 19100  $\mu$ g g<sup>-1</sup>) (Lee and Wang, 2004), and comparable to rice 165  $(475.9 \pm 61.2 \ \mu g \ g^{-1})$ , pine  $(558.6 \pm 103.6 \ \mu g \ g^{-1})$  and poplar  $(564.6 \pm 124.1 \ \mu g \ g^{-1})$  combustions 166 (Zhu et al., 2022), but much lower than coal combustion (6.3 mg  $g^{-1}$ ) (Huo et al., 2021). The 167 168 contributions of different chemical categories are displayed in Figure S3. Oxygenated compounds 169 dominate the total EFs, accounting for 48.4%, followed by aromatics (29.8%), b-alkanes (5.3%), 170 nitrogen-containing compounds (4.0%), alkenes (4.0%), and *n*-alkanes (2.3%). Unresolved complex 171 mixtures (UCMs) are further separated into aliphatic, cyclic, and oxygenated UCM due to retention 172 times and mass spectrums. The UCM ratio in this work (2.3% in EFs) is comparable to biomass 173 burning (Huo et al., 2021) and diesel exhaust (He et al., 2022) analyzed by GC×GC-MS, and is much 174 smaller than the UCM ratio (>50%) in biomass burning smoke analyzed by 1D GC-MS (Zhu et al., 175 2022). Ketones are the most abundant oxygenated compounds, accounting for 13.6% of the total EFs, 176 followed by aldehydes (9.7%), esters (8.1%), alcohols (6.9%), phenols (3.6%), and acids (3.1%). The 177 emission profiles are comparable to corncob and wood combustion, which are also dominated by 178 ketones and esters (Huo et al., 2021). However, the abundance of phenol is much lower than in 179 biomass-burning smoke (>15%) (Zhu et al., 2022; Huo et al., 2021), while comparable to coal 180 combustion (5.4%) (Huo et al., 2021).

EFs of selected compounds are listed in Table S4, which were comparable with other incense burning studies (Lee and Wang, 2004; Yang et al., 2007; Manoukian et al., 2016), while the EF of benzene ( $59.6 \pm 43.1 \ \mu g \ g^{-1}$ ) is slightly lower than other studies ( $188 \sim 1826 \ \mu g \ g^{-1}$ ) (Lee and Wang, 2004; Yang et al., 2007; Manoukian et al., 2016). The Tenax TA liner in the CIS system does not capture benzene at an initial temperature of 20 °C, while it is efficient for the trapping of most I/SVOC compounds. Lower CIS temperature may trap benzene while causing water condensation. As a result, the tailing of benzene on the second column (Figure S2) causes an underestimation ofblob integration and results in an underestimation of EF.

189 The top 10 compounds are all VOC compounds (Figure S4), accounting for 35.3% of the total EFs. Toluene  $(70.8 \pm 35.7 \ \mu g \ g^{-1})$  is the most abundant compound in incensing-burning smoke, 190 benzene, furfural, phenol, styrene, 2-oxo-propanoic 191 followed by acid methyl ester. 3-methyl-2-butanone, ethylbenzene, 1-hydroxy-2-propanone, and benzyl alcohol. Note that VOC 192 193 compounds discussed here are part of volatile organics captured by Tenax-TA, not the common 194 VOCs detected by SUMMA-GC-MS. The top 5 IVOCs are B17 b-alkanes, B16 b-alkanes, B18 195 *b*-alkanes, diethyl phthalate, and 1,6-dioxacyclododecane-7,12-dione. The naphthalene (a typical PAH, 2 rings) EF is 3.0  $\pm$  1.5  $\mu$ g g<sup>-1</sup>, comparable to rice straw combustion (Zhu et al., 2022). SVOCs 196 197 are all *n*-alkane species and only account for less than 1% of the total EFs.

198 The average VBS distribution of incense burning is displayed in Figure 1, and the 199 volatility-polarity distribution is exhibited in Figure S5. In general, the EF decreases as the volatility 200 decreases, following the trend of VOC-EF (80.8%) > IVOC-EF(19.2%) >> SVOC-EF (<0.1%). The 201 chemical compositions in the VOC-IVOC range are shown in Figure S6. Oxygenated compounds 202 (53.5% of the total VOC EFs) and aromatics (37.6%) are largely detected in the VOC range, while 203 *b*-alkanes, *n*-alkanes, and oxygenated compounds are the main components of IVOC compounds. 204 The average VBS distribution is similar to cooking emissions (Song et al., 2022a) and wood 205 combustion (Stewart et al., 2021), but less volatile than gasoline exhausts (Lu et al., 2018) and more 206 volatile than diesel emissions (Lu et al., 2018). For example, the proportion of chemicals with saturated vapor concentration (C\*) more than  $10^6 \ \mu g \ m^{-3}$  (Figure 1 a) is 80.8% (incense burning), 207 208 80.7% (cooking emissions) (Song et al., 2022a), 77.6% (wood combustion) (Stewart et al., 2021), 209 94.2% (gasoline exhaust) (Lu et al., 2018), and 41.0% (diesel exhaust) (Lu et al., 2018). The polarity 210 of incense burning is dominated by non-polar and intermediate-polarity organics (P1 ~ P5, Figure 211 S5). The volatility-polarity distribution of incense burning is quite similar to cooking emissions 212 (Song et al., 2022a), dominant by VOCs in the volatility range of before B13 and the polarity range 213 of P1 ~ P5.

A similar emission pattern but different EFs of different incense-burning emissions are observed.

Similarities among incense burning are more dominant than diversities. First, pixel-based partial least squares-discriminant analysis (PLS-DA) elucidates that there is no systemic difference between different chromatograms of incense burning emission, no matter different incense shapes (Figure S7) or materials (Figure S8). Second, the compositions of different types of incense emissions are indeed quite similar (Figure S9 and Figure S10). Third, the multiway principal component analysis (MPCA) positive loadings are much larger than negative loadings, indicating that the similarities between samples are much more important than the differences (Figure 2).

222 However, the absolute EFs significantly diverge according to different incense forms (p = 0.03, 223 Figure S11) and different materials (p < 0.01, Figure S12). Incense made in stick form (incense stick: 893.2  $\pm$  335.6 µg g<sup>-1</sup>, Thailand incense stick: 877.5  $\pm$  123.8 µg g<sup>-1</sup>) emits more organics than made in 224 coil form (incense coil: 835.5  $\pm$  306.0 µg g<sup>-1</sup>). The EF of mosquito coil is the smallest (382.5  $\pm$  175.0 225 ug g<sup>-1</sup>). A similar pattern was observed in previous work (Jetter et al., 2002). Concerning the incense 226 materials, we spot that the so-called smokeless sandalwood stick emits more abundant organics 227  $(1195.8 \pm 83.3 \ \mu g \ g^{-1})$  than common sandalwood sticks  $(633.7 \pm 6.6 \ \mu g \ g^{-1})$ . The emission of 228 smokeless sandalwood sticks is even greater than aromatic sticks (893.2  $\pm$  335.6  $\mu$ g g<sup>-1</sup>) and coils 229  $(824.8 \pm 228.5 \ \mu g \ g^{-1})$ . Our results demonstrate that although smokeless sandalwood stick is 230 231 preferred as fewer particulates are generated during the combustion process, the gaseous emissions 232 are enhanced compared to other incenses.

# 3.2 Contributions of home-use incense burning to ozone and secondary organic aerosols (SOA)

The total OFP is 1513.4  $\pm$  551.0  $\mu g~g^{-1}$  which is 1.91 g O\_3/g VOC-IVOCs. The OFP 235 enhancement ratio (OFP per mass of precursor) is much smaller than gasoline exhaust  $(3.53 \text{ g O}_3/\text{g})$ 236 237 VOCs) (Wang et al., 2013) and evaporation  $(2.3 \sim 4.9 \text{ g O}_3/\text{g VOC})$  (Yue et al., 2017), showing that 238 incense burning is less efficient on ozone formation than gasoline-related sources. The lack of IVOC 239 measurements in previous work could also cause an overestimation of the OFP enhancement ratio as 240 IVOCs are less efficient in ozone formation. Toluene, furfural, p-xylene, benzyl alcohol, phenol, 241 2-furanmethanol, o-xylene, ethylbenzene, 1-hydroxy-2-propanone, and benzene are top 10 species 242 that contribute most to OFP (Figure S4). Oxygenated compounds take up 48.2% of the total OFP,

followed by aromatics (41.0%), and alkenes (6.7%) (Figure S3). VOCs dominate the total OFP, 243 244 accounting for 92.4% while IVOCs take up 7.6% (Figure 1). Aromandendrene, naphthalene, and 245 α-cedrene are the top 3 IVOC-OFP contributors. The volatility distribution of OFP contribution is 246 comparable to cooking emissions, as VOCs account for 88.8 ~ 99.9% of the total cooking OFP 247 estimation (Song et al., 2022a). Toluene contributes the most OFP in both cooking emissions and 248 incense burning. Short-chain linear aldehydes (pentanal, hexanal, nonanal) originating from the 249 degradation of oils play a more important role in OFP contribution in cooking emissions (Song et al., 250 2022a), while benzenes, furfural, alcohols, and phenols are non-negligible OFP contributors in 251 incense burning.

252 Figure 1 shows the volatility distribution of estimated SOA estimation, with the top 10 253 contributors displayed in Figure S4. IVOCs contribute 19.2% of the EFs while accounting for 40.0% 254 of the total SOA estimation, highlighting the importance of IVOCs in SOA formation. The 255 contribution of IVOC species to SOA is higher than EFs due to the relatively higher yields and  $k_{OH}$ . 256 which has already been reported in cooking emissions (Song et al., 2022a; Yu et al., 2022), gasoline 257 exhaust (Zhao et al., 2014), diesel exhaust (Zhao et al., 2015), and biomass burning (Stewart et al., 258 2021). Oxygenated compounds account for 32.9% of the SOA estimation, followed by aromatics 259 (23.7%), and *b*-alkanes (11.5%) (Figure S3). Phenol, benzyl alcohol, styrene, toluene, B18 cyclic 260 UCM, aromandendrene, 2-furanmethanol, B17 b-alkanes, benzene, and phenyethyne are the top 10 261 SOA contributors. The incense-burning SOA formation profiles are distinct from cooking emissions (Song et al., 2022a) and biomass burning (Huo et al., 2021). Cooking SOA is largely derived from 262 263 the oxidation of short-chain acids and aromatics (Song et al., 2022a), while phenols account for more 264 than 65% of the SOA estimation from biomass burning (Huo et al., 2021). Phenols only account for 265 11.0% of SOA estimation in this work. Alcohols (7.3%) and furans (7.6%) are much more important 266 SOA precursors in incense burning compared to biomass burning and cooking emissions. Compared 267 with other sources, we stress the importance of incense-burning benzenes, furfural, alcohols, and 268 phenols in OFP formation and alcohols and furans in SOA formation. The secondary formation 269 potential of mosquito coils is the lowest, while OFP and SOA of burning smokeless sandalwood 270 sticks are the highest. Compared to other incense, the higher aromatic contents of smokeless

sandalwood sticks burning fumes result in much more ozone and SOA formation.

## 272 **3.3** Identification of molecular markers from incense burning

273 Pixel-based MPCA is utilized to identify tracers of incense burning emissions. In brief, MPCA 274 decomposes a matrix X into a scoring matrix (S) and a loading matrix (L). Similarities and 275 differences in chromatograms are revealed by positive and negative loadings, respectively (Figure 2) 276 (Song et al., 2022b). The similarities of chromatograms could be explained by benzenes (toluene, 277 ethylbenzene), *p*-xylene, o-xylene, and ketones (3-methyl-2-cyclopenten-1-one, 2-hydroxy-2-cyclopenten-1-one, 3-ethyl-2-pentanone), aldehydes (furfural, succindialdehyde, 278 279 2-methyl-2-butenal), 2-methyl-propanoic acid, 1-methyl-1H-pyrazole, 2(5H)-furanone, and 280 2-furanmethanol. The differences between samples could be largely explained by 2-methyl-2-butenal, 281 2(5H)-furanone, 3,4-dimethylfuran, 2,3-dihydro-1H-inden-1-one, 2-methoxy-naphthalene, and 282 1,2-dihydro-2,2,4-trimethyl-quinoline. The negative loadings (0.006) are significantly smaller than 283 the positive loadings (0.07), confirming the dominance of similarities among chromatograms. The 284 relationship between the EFs of these compounds among different incense types is displayed in 285 Figure S13. Although the total EFs are significantly different (p = 0.03), the EFs of selected 286 compounds (2-hydroxy-2-cyclopenten-1-one, 2-furanmethanol, 3-ethyl-2-pentanone, and furfural are 287 significantly no different (p > 0.08). As a result, we recommend these compounds as incense-burning 288 tracers. It is reported that furfural is formed during the thermal degradation of hemicelluloses (Uhde 289 and Salthammer, 2007), while the oxidation of furfural under harsher conditions forms 290 2(5H)-furanone (Depoorter et al., 2021). The formation mechanism of furfural from xylose and D-xylopyranose is displayed in Figure S14 (Ahmad et al., 1995; Bonner and Roth1, 1959; Nimlos et 291 292 al., 2006). The initiation of the degradation of five-carbon sugars is from the acyclic form of pentoses 293 or directly via a 2,3- $(\alpha,\beta)$ -)unsaturated aldehyde. The dehydrating of the intermediate compounds 294 finally forms furfural (Figure S14). The addressed tracers, furfural, 2-furanmethanol, and 295 2(5H)-furanone, have already been identified in incense burning smoke in previous work (Depoorter 296 et al., 2021; Tran and Marriott, 2007).

Furthermore, we compare the chemical profiles with an odor database (Aroma Office 2D, Gerstel). Among the top 20 chemicals contributing to EFs, furfural (bread-like, alcoholic, 299 incense-like), phenol (mushroom, acid, burnt plastics), 1-hydroxy-2-propanone (buttery, caramellic, 300 fruity), benzyl alcohol (burning taste, flower, roasted), limonene (citrus-like, fruity, lemon-like), and 301 2-methyl-propanoic acid (apple-like, cheese-like, sweat) could be the aroma compounds. As for 302 tracers identified above, 2-furanmethanol (burnt sugar, honey, sweet) could also be another aroma 303 compound. Among them, furfural is widely and largely detected which could be the most important 304 molecular marker of incense burning (Silva et al., 2021; Ho and Yu, 2002). Note that 305 aromandendrene, a cucumber-like, woody, and floral compound, is only detected in one incense coil 306 sample (incense coil 2, Figure S1). Aromandendrene is also detected in plants, such as in dry flowers 307 of Lonicera japonica (Shang et al., 2011). The emission factor of aromandendrene is rather large (4.3  $\mu g g^{-1}$ , 0.7% of the total EFs), and is a significant SOA precursor (2.3  $\mu g g^{-1}$ , 3.9% of the total SOA 308 estimation). The importance of aromandendrene on incense flavor and SOA formation could not be 309 310 neglected. Aromandendrene could also be responsible for the distinct flavor of a certain incense coil. 311 As above said, we recommend furfural be used as a molecular indicator of incense burning 312 regardless of the incense type or additives, especially responsible for the flavor of incense burning.

#### 313

#### 3.4 Risk assessment of incense burning organics

314 The hazardous compounds from incense burning could cause adverse health effects on human 315 health (Wong et al., 2020; Yang et al., 2007; Chen et al., 2021; Yang et al., 2017). To evaluate the 316 potential risks of these compounds, we conducted a pixel-based risk assessment (bioaccumulation 317 potential, BAP) for partition coefficient estimation. Chemicals with high BAP concerns are listed in 318 Figure 3. 2-methoxy-naphthalene, acenaphthylene, dibenzofuran, diethyl phthalate, dibutyl phthalate, 319 benzoic acid 2-ethylhexyl ester, C15 - C19 n, and b-alkanes are regarded as high BAP concern 320 (Figure 3). Among them, acenaphthylene is a toxic polycyclic aromatic hydrocarbon (PAH) that is 321 widely detected in incense smoke (Yadav et al., 2022). Dibenzofuran, an oxygenated compound with 322 detrimental effects on human health (Suzuki et al., 2021), is also detected in the smoke of incense 323 burning (Tran and Marriott, 2007). Diethyl phthalate and dibutyl phthalate are phthalate esters (PAEs) 324 widely used as plasticizers which are endocrine disruptors (Wang and Qian, 2021). PAEs are 325 abundant in incense smoke (Tran and Marriott, 2007). We propose that acenaphthylene, dibenzofuran, 326 and PAEs could be chemicals of high-risk concern in incense smoke. We also assess the arctic

327 contamination potential (ACP) as shown in Text S1. Further epidemiologic studies should be carried328 on to demonstrate the health effect of these hazardous compounds.

#### 329 **4 Implication**

330 The non-target approach of GC×GC-MS gives us a full glimpse of incense smoke, spotting a 331 large pool of organics (317 compounds) covering the VOC-IVOC-SVOC range. We have provided a 332 detailed description of both primary emission and secondary estimation of incense burning organics 333 which is ready-to-use in SOA simulation models. IVOCs (130 compounds) are crucial organics accounting for 19.2% of the total EFs and 40.0% of the SOA estimation, highlighting the importance 334 335 of incorporating IVOCs into SOA models. Further investigation should be carried on to elucidate 336 emission characteristics of short-chain compounds that are lacking in our research, such as alkanes 337 (<C7), (<C7), and aldehydes (<C5). By combining alkenes data obtained from 338 gas-chromatography-flame ionization detector (GC-FID) and proton transfer mass spectrometer 339 (PTR-MS), the emission pattern of incense burning could be well demonstrated. Comparisons of 340 IVOC capture efficiency on different sampling materials should also be taken into account to obtain 341 a reliable quantification result of IVOC species. High-time resolution measurement should also be 342 carried on to understand the time-resolved pattern of incense burning.

We also suggest furfural as the molecular marker of incense burning as the EFs of furfural among samples are relatively stable. Pixel-based MPCA also indicates that furfural is responsible for the similarities between chromatograms. Furfural may be the key aroma compound of incense smoke. This key component identified in this work could be implemented in source apportionment. Furfural is also a key component contributing to OFP (rank 2). Phenol, toluene, 2-furanmethanol, benzene, and benzyl alcohol are the main contributors to both OFP and SOA.

Surprisingly, we find that the EF of burning smokeless sandalwood sticks is the highest, with a remarkable contribution to OFP and SOA, due to the high aromatic contents. We recommend that both gaseous and particulate organics should also be taken into consideration when burning incense. The single reduction of particles does not mean fewer emissions of gas-phase organics. A comprehensive assessment of incense-burning organics in both gas- and particle-phase should be implemented.

355 Combing pixel-based property estimation and blob identification, the risk assessment analysis of 356 compounds could benefit analysts with less experience with GC×GC. The risk assessment in this 357 work demonstrates that acenaphthylene, dibenzofuran, and PAEs are chemicals of high-risk concern 358 and warrant further control. It was reported that more than half of Chinese residents burn incense 359 every day at home for more than 20 years (Salvi and Apte, 2016). The toxic PAHs detected in indoor 360 air could be 19 times higher than in outdoor air (Salvi and Apte, 2016). Exposure to these hazardous 361 compounds could result in significant health threats. As a result, it is of vital importance to reveal and assess the epidemiological influences of incense burning in future work. 362

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# 364 Acknowledgment

This research is supported by the National Natural Science Foundation of China (No. 22221004), the National Key Research and Development Program of China (2022YFC3701000, Task 2), the National Natural Science Foundation of China (No. 42107115, 41977179, 42275104), the Natural Science Foundation of Shandong Province, China (No. ZR2021QD111), the Hong Kong Research Grants Council (No. 11304121).

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# 584 Figures

- **Figure 1.** Volatility distributions of EF, OFP, and SOA with chemical class in each volatility bin. The
- *x*-axis is the unsaturated vapor concentration in logarithmic form (log C\*,  $\mu$ g m<sup>-3</sup>). The *y*-axis is the
- normalized mass emission factor (100%).
- 588 Figure 2. Positive (a) and negative loadings (b) of incense burning samples describing similarities
- and differences between chromatograms. The color bar is the loading.
- **Figure 3.** Chemicals with high bioaccumulation potential (BAP) assesses by pixel-based approaches.



**Figure 1.** Volatility distributions of EF, OFP, and SOA with chemical class in each volatility bin. The *x*-axis is the unsaturated vapor concentration in logarithmic form (log C\*,  $\mu$ g m<sup>-3</sup>). The *y*-axis is the normalized mass emission factor (100%).



Figure 2. Positive (a) and negative loadings (b) of incense burning samples describing similarities
and differences between chromatograms. The color bar is the loading.



**Figure 3.** Chemicals with high bioaccumulation potential (BAP) assesses by pixel-based approaches.