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Contributions of Indoor Household Activities to Inhalation Health Risks Induced by Gaseous Air Pollutants in Hong Kong Home

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ABSTRACT

To understand inhalation health risks at home, a comprehensive sampling campaign was conducted in a Hong Kong residential apartment from October to December 2019. Emissions of nine typical household activities, household background release and outdoor pollution were continuously monitored using a suite of the state-of-the-art instruments. Health risks were evaluated in each one-year exposure period. Acrolein and nitrogen dioxide (NO_2) likely caused chronic non-carcinogenic risks to residents in all exposure stages. Furthermore, the health risk of the respiratory system was proved in all age groups. For the first time, several household activities were found to cause acute health risks due to exposure to formaldehyde, benzene, NO_2 and acrolein in normal urban daily life. The probability distributions of cancer risks from household activities revealed that formaldehyde was the main carcinogen with an average risk of 2.00×10^{-4}, followed by benzene, acetaldehyde, ethylbenzene and dichlorobenzene. Among all indoor activities, incense burning was the largest contributor to total cancer risk (46%), as this source was respectively responsible for 47%, 54%, and 39% of cancer risks from formaldehyde, benzene, and acetaldehyde, followed by cooking and smoking. Notably, source-related health risk analysis showed that household background release dominated regardless of cancer risk (35.7–58.5%) or non-carcinogenic risk (57.1%). Further, the cancer risk from birth to 18 years was approximately 1.8–2.7 times that of 18–60 and 60–85 years of age. This study shed light on health risks posed by various household activities and highlighted the importance of indoor activities and household background release on acute and chronic health risks.

Keywords: Indoor air pollution, Household activities, Non-carcinogenic health risk, Incremental lifetime cancer risk, Hazardous air pollutants

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1 INTRODUCTION

Indoor health risks of inhalation have become a public issue of widespread concern (Ahmed *et al.*, 2019; Kumar *et al.*, 2016). In indoor environments, there are various hazardous air pollutants (HAPs) which can cause health risks through inhalation, such as hazardous volatile organic compounds, polycyclic aromatic hydrocarbons (PAHs), nitrogen dioxide (NO_2), and carbon monoxide (CO). Exposure to these HAPs can potentially induce a series of diseases, such as sensory irritation to eye and skin (Araviiskaia *et al.*, 2019; Tsai, 2019), airway inflammation (Pope *et al.*, 2015), rhinitis (Kim *et al.*, 2013), asthma (Billionnet *et al.*, 2011), chronic obstructive pulmonary diseases (Ghozikali *et al.*, 2016), and different types of cancer (IARC, 2021).

In the past two decades, increasing studies have investigated the inhalation health risks indoors. These studies clearly proved that household air pollution made a dominant contribution to indoor inhalation health risks and even the total inhalation health risks of indoor and outdoor exposures

(Du *et al.*, 2014a). For examples, de Bruin *et al.* (2008) reported that household exposure contributed as high as 99% to the inhalation health risk of formaldehyde in European Union. In China, household exposure accounted for averagely 48% of the total inhalation cancer risk encountered by working adults (Du *et al.*, 2014a).

So far, approximately 60 HAPs were identified to be able to cause health risks in household environments, including carbonyls (Liang, 2021; Xiang *et al.*, 2017), aromatics (Du *et al.*, 2014b), PAHs (Rostami *et al.*, 2019; Wu *et al.*, 2022) and so on. These HAPs come from various sources which can be classified into three groups, i.e., (i) household background release, including building and furniture materials emissions (Du *et al.*, 2014b; Gao *et al.*, 2014), and microbial activity emissions (Mensah-Attipoe *et al.*, 2016); (ii) outdoor air infiltration (Ielpo *et al.*, 2019); and (iii) household activity emissions, such as cooking (Arı *et al.*, 2020), tobacco smoking (Naddafi *et al.*, 2019), and incense burning (Lee and Wang, 2004). Plenty of studies emphasized on the total inhalation health risks in different indoor environments, such as restaurants (Arı *et al.*, 2020), waterpipe cafés (Naddafi *et al.*, 2019), or homes (Fang *et al.*, 2019), to represent the health risks induced by typical indoor activities. However, a dearth of studies quantifies the contributions of the aforementioned source groups to household inhalation health risks though explicit understanding of the contributions of these three source groups is closely related to appropriate measures that prevent residents from possible inhalation health risks at homes. HAPs emitted from different indoor activities and their risks to residents were seldom studied simultaneously before. Therefore, it was difficult to make comparison of different indoor activities due to diversity in analytical techniques and sampling environments.

In previous studies, long-term inhalation health risks have been extensively investigated in various household environments, whereas acute inhalation health risks were only observed for certain situations, i.e., rural indoor biomass or solid fuel combustions (Sun *et al.*, 2019), and no studies have provided direct evidence to show whether urban residents may experience acute inhalation health risks in normal daily life. Moreover, previous studies have typically assessed inhalation health risks over longer exposure periods, such as 35 or 40 years (Du *et al.*, 2014a), which may obscure or underestimate the health risks of certain HAPs and certain age groups. Assessment strategy taking into account different exposure periods (Fang *et al.*, 2019) would specifically guide the mitigation of indoor air pollution for each age group.

Hence, based on a carefully designed sampling campaign and the application of a suite of state-of-the-art instruments, we comprehensively measured a variety of air pollutants in a residential apartment of Hong Kong. Various indoor activities were regularly conducted during the sampling period. Using a recognized health risk assessment strategy, we evaluated the chronic and acute non-carcinogenic health risks and cancer risks of exposure to typical HAPs and household activities for residents in different age groups. Furthermore, contributions of household background release, outdoor air infiltration and household activity emissions to total inhalation health risks were estimated. The findings are helpful to reveal the household inhalation health risks and promote targeted control strategies by estimating health risks of each household source.

2 METHODS

2.1 Sampling

The sampling campaign was conducted from October to December 2019 in a residential apartment on the 7th floor of an apartment building located in Kowloon, Hong Kong. This is a residential apartment in Hong Kong, with a floor area of 25.4 m^2 and an inner volume of 66.1 m^3, which is divided into a living room, two bedrooms, a kitchen, and a bathroom, as displayed in Fig. S1 in Supplementary Information (SI). No interior painting or decorating activity has been undertaken in the past six months. Surrounding sources of outdoor air pollution included vehicles, restaurants, and gas stations. During the sampling campaign, the average outdoor temperature and relative humidity were 22.2°C and 66%, respectively, and the average wind speed was 7.6 m s^{-1} with predominant wind directions of 50°–90°.

For the measurements of air pollutants, minutely VOC measurements were performed online using a Proton Transfer Reaction-Mass Spectrometer (PTR-Q-MS 500, Ionicon Analytik Ges.m.b.H., Austria). The mass detection range was 1–512 amu with a mass resolution of $<$ 1 amu, while the

response time was 100 msec and the detection limit was 1 pptv. The PTR-Q-MS was calibrated with standard gas including 12 VOC species (Table S1) before sampling to determine the concentrations of main HAPs more accurately. The influence of relative humidity on the concentration of formaldehyde was removed through the correlation between relative humidity and formaldehyde concentrations, and the interferences of ethanol to formaldehyde and acetaldehyde were also excluded by determining the correlation between the concentrations of ethanol and formaldehyde/acetaldehyde. Furthermore, the interferences of isomers to the concentrations of some HAPs such as acrolein, propanal, and 1,4-dichlorobenzne were discussed in the assessment of health risks. In addition, a Thermal-desorption Aerosol Gas-chromatography-Time of Flight-Mass Spectrometry (TAG-TOF-MS) was deployed to monitor the hourly concentrations of selected PAHs occurring as organic aerosols. Detailed information about this instrument can be found in our previous publications (Huo *et al.*, 2022; Lyu *et al.*, 2021). Furthermore, a set of trace gas analyzers (Advanced Pollution Instrumentation (API), USA) were utilized to measure O_3 (API model 400 with a detection limit of $<$ 0.4 ppb, and precision of $<$ 0.5% for readings $>$ 100 ppb), SO_2 (API model 100E with a detection limit of 0.4 ppb and precision of $\leq$ 0.5% for readings $>$ 50 ppb), CO (API model 300EU with a detection limit of $<$ 20 ppb and precision of 0.5%), and NO_2 (API model T200U with a detection limit of $<$ 50 ppt and precision of 0.5% for readings $>$ 5 ppb). All the trace gas analyzers were calibrated before and after the measurements, and details were described in our previous studies (Guo *et al.*, 2009, 2013). The HAPs identified during the sampling campaign are listed in Table S2 in SI.

During the sampling, all the instruments were placed in the living room of the residential apartment, and the sampling inlets were fixed at the center of the apartment above the floor of 1–1.2 meters, as shown in Fig. S1. A switch timer controlled the instruments to alternatively collect indoor and outdoor air samples every other hour. As clarified in our recent publications (Huo *et al.*, 2022; Lyu *et al.*, 2021), this method was acceptable as it would not affect temporal variations of air pollutants. Moreover, only the average concentrations during the sampling period would be used to calculate chronic health risks. Thus, the uncertainty introduced by this sampling method was negligible. For indoor air sampling, besides collecting the samples of indoor background air, air samples during typical household activities, including candle burning, incense burning, tobacco smoking, cooking (breakfast, lunch and dinner), coffee/tea brewing, cleaning, laundry, bathing, and air freshener spraying, were also monitored. All the household activities were repeated at least three times up to over thirty times. Table S3 in SI summarizes the type and number of daily activities in the sampling period. The concentration of a HAP during each activity was obtained by averaging the values from the beginning of the activity to the end when its concentration reached the background level. This duration was also deemed as the exposure time of residents to this activity. Further details for this sampling are summarized in Text S1 or can be referred to our recent publications (Huo *et al.*, 2022; Lyu *et al.*, 2021).

2.2 Health Risk Assessment

In this study, chronic non-carcinogenic and carcinogenic health risks were adopted to assess the health risks encountered by residents, according to equations developed and updated by U.S. Environmental Protection Agency (U.S. EPA) (U.S. EPA, 2009), which have been widely applied in previous studies (Dai *et al.*, 2017; Lyu *et al.*, 2020; Mo *et al.*, 2021). However, different from other studies, we adopted different living patterns and age sensitivity to HAPs of residents in each age group. The time-activity pattern model of residents constructed by us is summarized in Table S4 and Table S5 in SI. Age sensitivity factor of residents to HAPs is given in Table S6 in SI.

Hazard quotient (HQ) and hazard index (HI) were used to characterize chronic non-carcinogenic health risks as follows (U.S. EPA, 2009):

$$HQ_i = \frac{EC_i}{RfC_i} \tag{1}$$

$$HI_j = \sum_{1}^{i} HQ_i \tag{2}$$

where *i* and *j* refer to a HAP and a health endpoint, respectively. HQ_i is the hazard quotient of HAP_i (unitless). EC_i represents the exposure concentration of HAP_i that acts on human body through inhalation. RfC_i is the reference concentration of HAP_i (μg m^{-3}), as listed in Table S2. HI_j is the health risk encountered by a specific health endpoint *j*. It equals the sum of the HQs of HAPs posing health risk on this health endpoint. The health endpoint of each HAP is also given in Table S2. For both HQ_i and HI_j, corresponding health risks likely occur when their values are higher than 1, and vice versa. Besides, an HQ_i or HI_j value larger than 0.1 might indicate the potential health risks (McCarthy *et al.*, 2009).

To estimate time-cumulative health risks, the following Eq. (3) was used (U.S. EPA, 2009):

$$HQ_c = \frac{1}{t}\sum_{1}^{i} HQ_m \tag{3}$$

where, HQ_c is time-cumulative HQ for any one consecutive exposure period, *t* is year month^{-1} counts covered by this consecutive exposure period, and HQ_m is the HQ in each age exposure stage across this consecutive exposure period.

Exposure concentration is quantified by Eq. (4) (U.S. EPA, 2009):

$$EC_i = \frac{C_i \times ET \times EF \times ED}{AT} \tag{4}$$

where C_i is the concentration of HAP *i* in air, μg m^{-3}; ET is exposure time, hours day^{-1}; EF is exposure frequency, days $year^{-1}$; ED is exposure duration, years; and AT is averaging time, referring to lifetime in years × 365 days $year^{-1}$ × 24 hours day^{-1}.

Cancer risk is quantified by Eq. (5) (U.S. EPA, 2009):

$$ILCR\chi = EC\chi \times IUR\chi \times ASF \tag{5}$$

where, χ refers to a carcinogen, and $ILCR_\chi$ is the incremental lifetime cancer risk of carcinogen χ (unitless). EC_χ is the exposure concentration of carcinogen χ, which is quantified by Eq. (3), where the AT is calculated by human life expectancy × 365 (days). The default value of lifetime is 70 years, but 85 years was used as the life expectancy in this study. IUR_χ is the inhalation unit risk of carcinogen χ (μg^{-1} m^{-3}), as listed in Table S2. ASF is age sensitivity factor (unitless) (U.S. EPA, 2009), as shown in Table S6.

Apart from chronic non-carcinogenic and carcinogenic health risks, this study also investigated possible acute health risks. By determining the maximum average concentration of a HAP in a specific time range, i.e., maximum 30-min or hourly average concentration, this study compared the value with corresponding acute health risk guidelines to judge whether an acute health risk occurred.

Furthermore, based on the measurement data and the above time-activity pattern models, the health risks of exposure to household background release, outdoor air infiltration and household activity emissions were quantified, respectively. Here, the average concentrations of indoor air pollutants when there were no indoor intensive emission activities were used for the estimation of the health risks of household background release, and the average concentrations of outdoor air pollutants were applied to evaluate the health risks of outdoor air infiltration, while the concentration of indoor air pollutants with each intensive emission activity was used to calculate the health risks of each household activity emission, which were summed up to represent the health risks of total household activity emissions.

2.3 Uncertainty and Sensitivity Analysis

To more accurately estimate the health risks of HAPs from indoor activities, this study employed Monte Carlo simulations and sensitivity analysis to assess the uncertainties in risk assessment (Dai *et al.*, 2017; Mo *et al.*, 2021; Zhou *et al.*, 2011). This method utilizes the probability distribution

of input data (i.e., EC_χ, IUR_χ) to generate a probability distribution of health risks, which will provide more information for risk assessment than a single point average value.

In this study, different distributions were chosen for each input parameter, including exposure concentration and IUR value for each HAP. According to U.S. EPA (U.S. EPA, 2021), the IUR is defined as the upper-bound excess lifetime cancer risk caused by sustained exposure to a HAP at an ambient concentration of 1 μg m^{-3}. In the simulations, IUR values recommended by U.S. EPA were specified as the maximum values with the highest probability and 0 was set as the minimum value, thus adopting a triangular distribution to characterize the IUR values. Exposure concentrations of HAPs were frequently described as lognormally distributed in previous studies (Dai *et al.*, 2017; Mo *et al.*, 2021). In this study, the exposure concentrations of HAPs were mostly lognormally distributed or skewed to some extent. Fitted distributions were used to represent indoor personal exposures.

3 RESULTS AND DISCUSSION

3.1 Characteristics of Household Air Pollution

Fig. 1 shows the average concentrations with standard errors of major HAPs identified in outdoor air, indoor background air and during each typical household activity in the sampling campaign. The average ratios of indoor background to outdoor concentrations, i.e., I/O, were also calculated. I/O ratio along with two-tail t-test ($p < 0.01$, Table S7 in SI) proved that the outdoor average concentrations of trace gases, PAHs, heptanal and trimethylbenzenes/acetophenone were significantly higher than indoors, suggesting dominant impact of outdoor air infiltration on indoor levels of these pollutants. In contrast, all the other HAPs possessed significantly higher indoor average concentrations than outdoors, implying long-standing important indoor sources of these HAPs (Alves *et al.*, 2020), such as construction and furniture materials, indoor deodorants, rubber, paint (Du *et al.*, 2014a).

The outdoor levels of the HAPs are basically consistent with other studies. For example, the outdoor concentrations of benzene and toluene were respectively 1.30 ± 0.21 μg m^{-3} and 5.22 ± 1.55 μg m^{-3} in this study, comparable to the levels of 0.95 ± 0.02 μg m^{-3} and 4.58 ± 0.15μg m^{-3} in suburban Hong Kong and 1.11 ± 0.03 μg m^{-3} and 4.07 ± 0.14 μg m^{-3} in roadside Hong Kong in 2018 (Zeng *et al.*, 2022). The outdoor levels of O_3, NO_2, CO and SO_2 were 46.75 ± 8.69 μg m^{-3}, 45.59 ± 14.36 μg m^{-3}, 658.32 ± 91.76 μg m^{-3}, and 10.15 ± 0.71 μg m^{-3}, respectively, which were close to the annual average concentrations of 60.00 μg m^{-3}, 38.00 μg m^{-3}, 595.00 μg m^{-3}, and 5.00 μg m^{-3} of 2019 in Hong Kong (EPD, 2019). However, the respective indoor background levels of benzene, toluene, and xylenes/ethylbenzene (1.34 ± 0.11 μg m^{-3}, 5.87 ± 0.63 μg m^{-3}, and 3.57 ± 0.44 μg m^{-3} in this study) were lower than the average levels of 5.31 ± 3.48 μg m^{-3}, 51.13 ± 23.07 μg m^{-3}, and 12.27 μg m^{-3} in six Hong Kong homes (Lee *et al.*, 2002). The reason was that the indoor background levels in this study excluded interferences from household activities, which made the observed concentrations well below the general indoor levels reported in previous studies in Hong Kong.

Fig. 1 and two-tail t-test ($p < 0.01$, Table S7) revealed that household activities led to significant rises in indoor concentrations of many HAPs. Candle burning caused increase of C_1–C_3 aldehydes, C_6–C_8 aromatics, chrysene, and benzo[a]anthracene, while incense burning enhanced levels of C_1–C_6 aldehydes, C_6–C_8 aromatics, NO_2, and CO, which were similar to previous studies. Furthermore, tobacco smoking caused rise of C_1–C_4 aldehydes and C_6–C_9 aromatics while cooking increased C_1–C_6 aldehydes, acetonitrile, C_6–C_9 aromatics, cyclohexane, NO_2 and CO. It was also found that coffee/tea brewing lifted the concentrations of C_2–C_9 aldehydes, C_6–C_8 aromatics and cyclohexane, and household hygiene activities, including cleaning, bathing, laundry, and air freshener spraying, led to the increase in C_1–C_3 aldehydes and C_6–C_8 aromatics. To sum up, species profiles of HAPs emitted from each household activities were consistent with previous studies (Ahn *et al.*, 2015; Dinh *et al.*, 2015; Lopez *et al.*, 2016; Wang *et al.*, 2018; Yener *et al.*, 2016; Zhao and Zhao, 2018).

The concentration profiles of HAPs during each household activity varied greatly among different studies. For example, the peak concentration of formaldehyde was 169.77 μg m^{-3} during candle burning in this study, while it was 13.39–397.33 μg m^{-3} and 1136–2577 μg m^{-3} in other

studies (Ahn *et al.*, 2015; Petry *et al.*, 2014). This is mainly due to limited studies and different experimental settings. So far, there has not been a study that comprehensively measures HAP emissions during various household activities in the actual household environment at least in Hong Kong. This study fills the gap and can be used to more accurately assess the health risks of household inhalation.

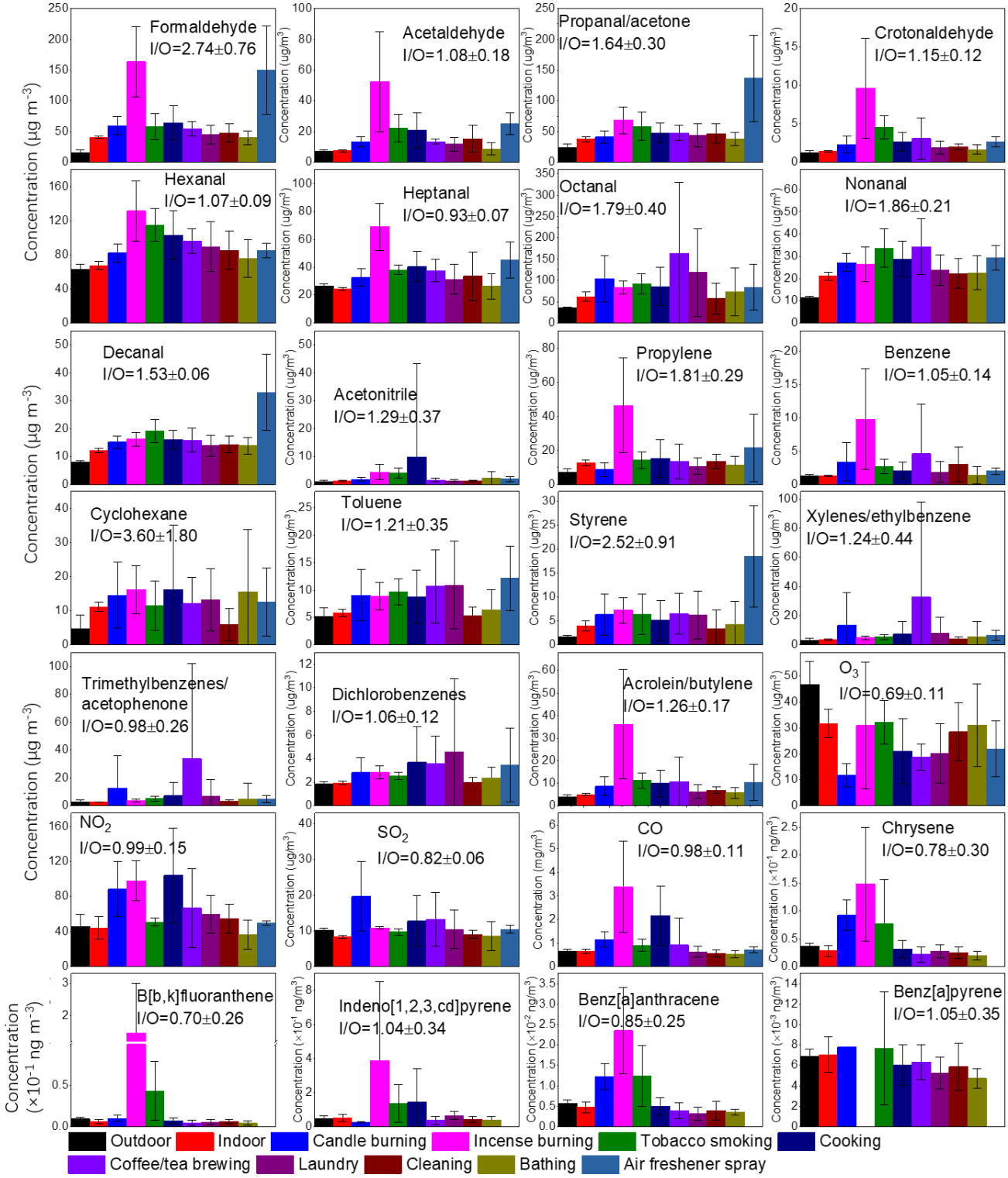

Fig. 1. Average concentrations of HAPs in outdoor air, indoor background air, and during various household activities.

3.2 Health Risk Assessments

3.2.1 Chronic non-carcinogenic health risks

Fig. 2 shows the average HQs of 24 HAPs measured in this study. The HQs in different age groups are summarized in Table S8 in SI. As shown in Fig. 2, based on lifetime exposure period, the HQs of propanal, acrolein, and NO_2 were higher than 1 in all exposure stages, implying that corresponding health risks were likely to occur in any one of these exposure stages. As for hexanal, octanal, O_3, formaldehyde, heptanal, SO_2, nonanal and decanal, they had the potential to cause non-carcinogenic health risk in all exposure stages, with HQs larger than 0.1. Besides, this study found that HAPs possessed severer health risks to young people, especially children (Table S8). Moreover, other HAPs had no non-carcinogenic detrimental effects in household environments in this study.

Previous study reported that NO_2 likely has chronic non-carcinogenic health risks to residents in Japanese dwellings in winter seasons (Azuma *et al.*, 2016). Furthermore, the chronic non-carcinogenic health risks caused by inhalation of household acrolein and propanal (HQs > 1) identified in this study need further clarification. Acrolein was previously reported to be the primary HAP likely causing chronic non-carcinogenic health risk to residents in Japan (Azuma *et al.*, 2016). And propanal was likely to cause health risk in some cases, since it had a maxima margin of exposure (MOE) value at 0.2, which was equal to a HQ value at 2.0. In this study, however, the HQs of acrolein and propanal were overestimated with an uncertain level, because the PTR-Q-MS could not accurately determine the concentrations of acrolein and propanal, owing to the interferences of substances having the same mass-to-charge ratios, such as butene and acetone, respectively. According to measurement results of whole air sample, the concentration of butene in this study was 0.16 ± 0.16 μg m^{-3}, when measured acrolein ranged from 4.90 to 36.2 μg m^{-3}. The interference of butene, therefore, might cause an uncertainty less than 3.3% on results regarding acrolein. However, the uncertainty of propanal induced by acetone would be larger, since the concentration of household acetone could reach 20 μg m^{-3} (Azuma *et al.*, 2016), while measured propanal was within the range of 39.3–149.5 μg m^{-3}. Nevertheless, with the aim of protecting residents from health risks, overestimation might be beneficial to people. Furthermore, to our knowledge, no studies have reported potential chronic non-carcinogenic health risks of octanal, hexanal, O_3, etc., but their HQs reached 0.49–0.70 in this study. Hence, residents should pay full attention to the possible health risks of NO_2, octanal, hexanal, and especially acrolein.

Fig. 3 shows the HIs for different endpoints and Table S9 summarizes HIs in different exposure stages. The HI for respiratory system was higher than 1 in all exposure stages even when the adverse effects of propanal to respiratory system were not considered. Therefore, year-cumulative HI for

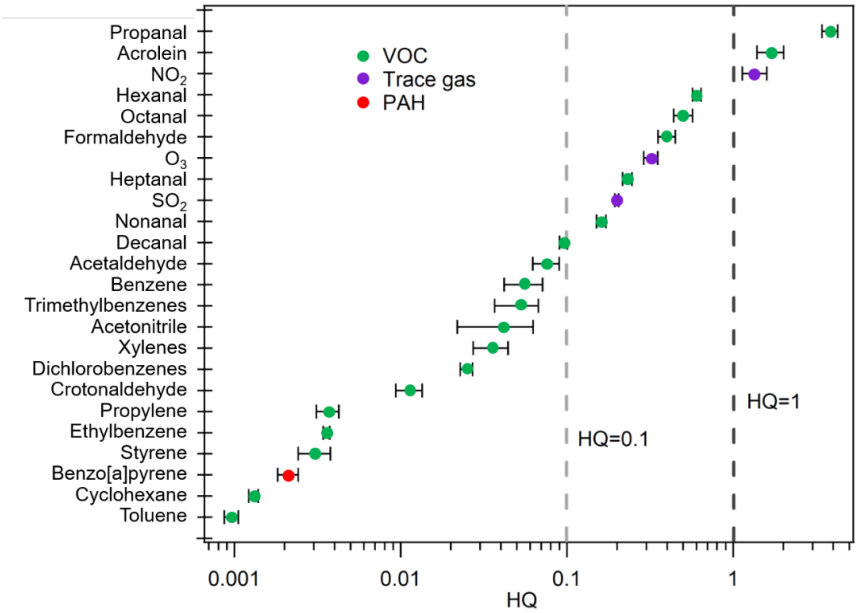

Fig. 2. HQs of 24 indoor HAPs measured in this study.

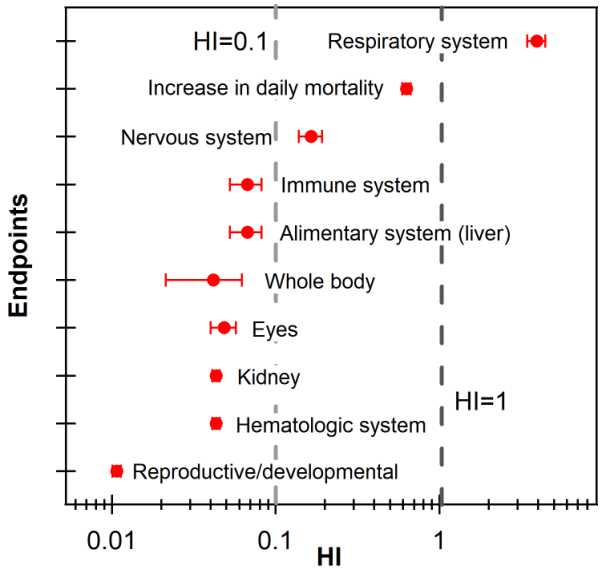

Fig. 3. HIs for different health endpoints.

respiratory system was also higher than 1 in any a consecutive exposure period. This meant that corresponding respiratory illnesses were likely to happen to residents of various age stages. This was because of many HAPs posing health risks to humans' respiratory system, such as formaldehyde, toluene, and NO_2. Besides, HIs for nervous system and daily mortality were larger than 0.1 in all exposure periods (Table S9), meaning continuous potential risks. Moreover, HIs were lower than 0.1 for other health endpoints in all exposure stages (Fig. 3, Table S9), indicating that corresponding health risks were not likely to happen to these health endpoints.

3.2.2 Acute non-carcinogenic health risks

Fig. 4(a) shows the maximum 30-min. and hourly average concentrations of formaldehyde in different activities. The maximum 30-min. and hourly average concentrations of formaldehyde due to candle burning, incense burning, cooking, and air freshener spraying all exceeded the 30-min. (100 μg m^{-3}) (WHO, 2010) and hourly (55 μg m^{-3}) (OEHHA, 2021) acute health risk guidelines, while tobacco smoking, coffee/tea brewing and cleaning only caused the exceedance of the maximum hourly average concentration of formaldehyde over the hourly acute health risk guideline (55 μg m^{-3}) (OEHHA, 2021). The results indicated that each of these household activities likely caused acute health risk to our eyes (sensory irritation) because of sharp rise of formaldehyde.

The acute health risks could occur when several household activities were carried out simultaneously. For example, candle burning, incense burning and tobacco smoking together enhanced the maximum hourly average concentration of benzene (28.32 μg m^{-3}) above the hourly acute health risk guideline (27 μg m^{-3}) (OEHHA, 2021), likely leading to acute health risks to the developmental, immune and hematologic systems of the residents. Moreover, the combination of candle and incense burning caused the exceedance of the maximum hourly average concentration of NO_2 (207.83 μg m^{-3}) over the acute health risk guideline (200 μg m^{-3}) (WHO, 2010), resulting in acute health risk to the respiratory system. Furthermore, acrolein could be an important HAP causing acute health risk to residents (Fig. 2), since its concentration increased sharply during incense burning, implying that incense burning might lead to acute health risks to the eyes and respiratory system (sensory irritation). This section for the first time provided evidence on the acute health risks, which is helpful to take effective measures to protect residents from possible acute health risks.

3.2.3 Carcinogenic health risk assessments

Fig. 5 shows the ILCRs of 10 carcinogens in Hong Kong home caused by household activities (background release excluded), based on Monte Carlo simulations reflecting the distributions of

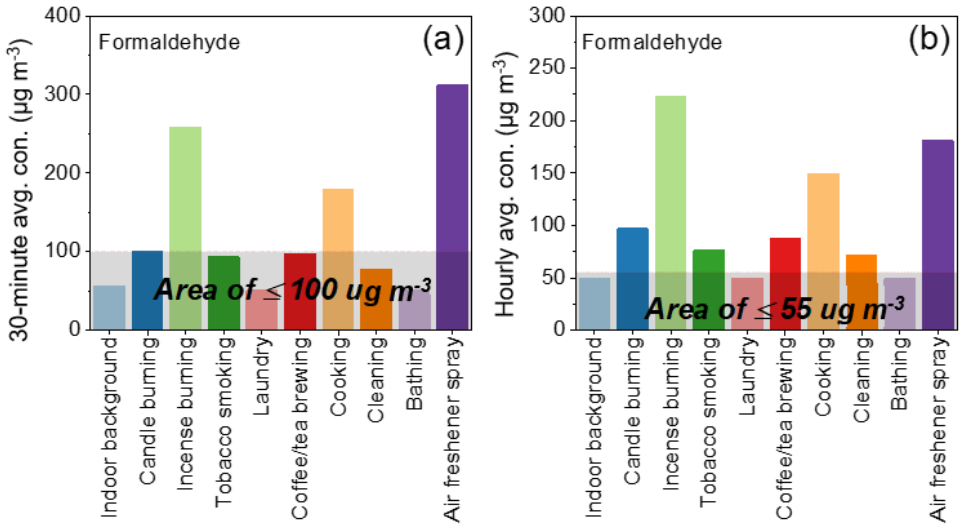

Fig. 4. Maximum 30-min. and hourly average concentrations of formaldehyde in different activities.

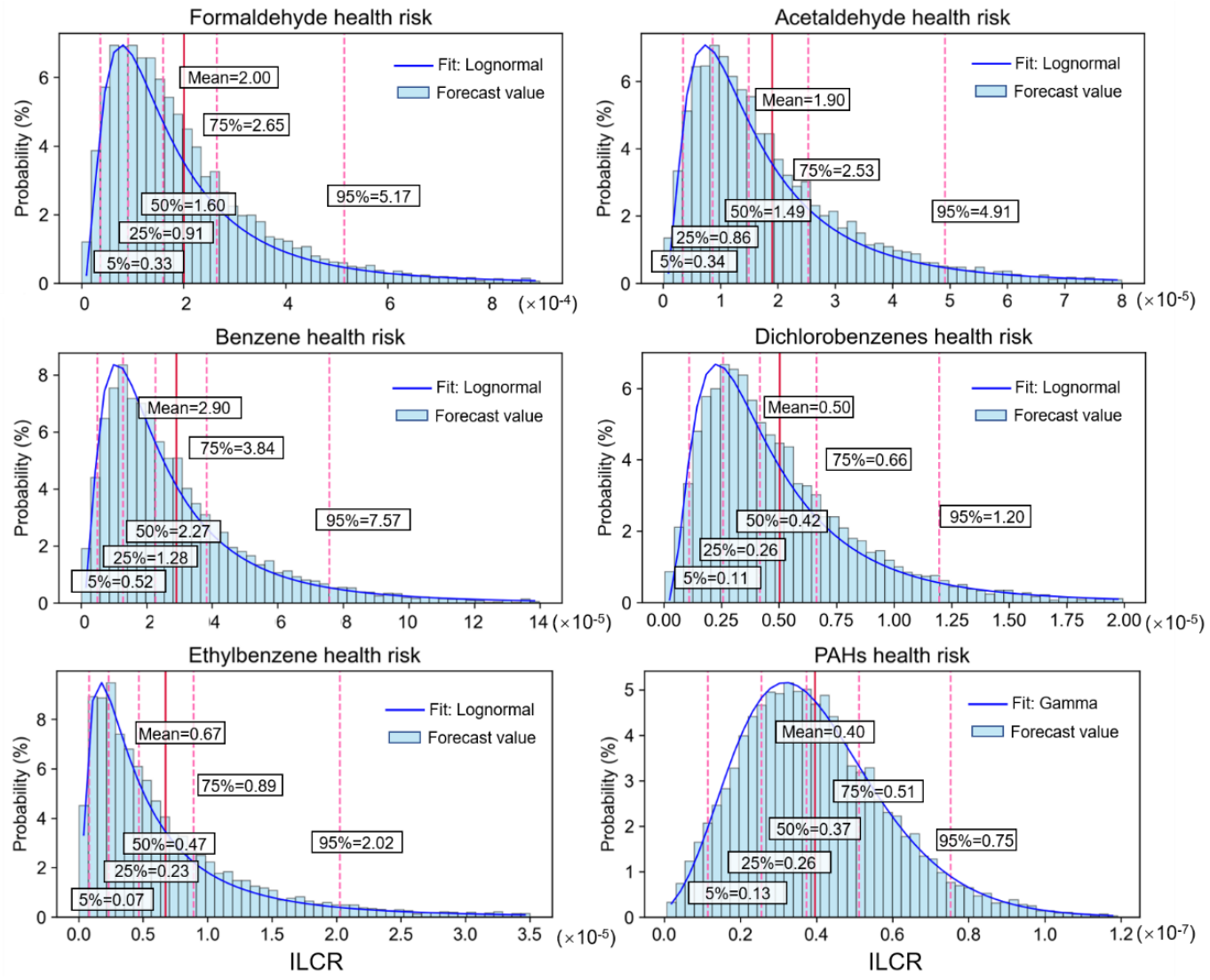

Fig. 5. Incremental lifetime cancer risk of carcinogens from household activities exposure.

personal risks. Probability distributions are presented together with typical parameters such as mean, 5th, 25th, 50th, 75th and 95th percentiles. Since the mean ILCRs of chrysene, benzo[b,k]fluoranthene, indeno[1,2,3-cd]pyrene, and benzo[a]anthracene all were over 10 times lower than that of benzo[a]pyrene, their ILCRs were summed up as ILCR of PAHs. ILCRs of individual PAH species are displayed in Fig. S3.

The mean and median carcinogenic risk of formaldehyde was the highest at 2.00×10^{-4} and 1.60×10^{-4} respectively, followed by benzene (2.11×10^{-5} and 1.66×10^{-5}), acetaldehyde (1.90×10^{-5} and 1.49×10^{-5}), ethylbenzene (6.72×10^{-6} and 4.67×10^{-6}), 1,4-dichlorobenzene (5.03×10^{-6} and 4.16×10^{-6}), all above the benchmark released by the U.S. EPA (1×10^{-6}). Besides, the carcinogenic risks of PAHs induced by indoor activities were 3.95×10^{-8} (mean) and 3.73×10^{-8} (median), respectively, indicating no carcinogenic risks. The difference between the mean and median values indicated a skewed distribution of estimated risk. As for the 95th percentile value, formaldehyde ranked first, and all estimated HAPs in Fig. 5 presented probable carcinogenic risks. Strikingly, at the 5th percentile, formaldehyde, benzene, acetaldehyde, and dichlorobenzenes emitted from indoor activities still caused carcinogenic risks, indicating the severity of indoor air pollution. Besides, the estimated carcinogenic risks of most VOCs were best fitted with lognormal distributions, but PAHs were better fitted with Gamma distributions. This might be because PAHs mainly came from incense burning or smoking, while other VOCs originated from different indoor activities, which would be further clarified in the next section.

By summing the carcinogenic risks of all measured species, the cumulative carcinogenic risks were obtained, with a mean value of 2.60×10^{-4} and a median value of 2.06×10^{-4}. For mean values, formaldehyde was the top contributor to total carcinogenic risk with a proportion of 77.0%, followed successively by benzene (11.1%), acetaldehyde (7.3%), ethylbenzene (2.6%), 1,4-dichlorobenzene (1.9%), and PAHs (0.02%). Previous study also reported that formaldehyde was the leading air pollutant causing cancer risk to Chinese working adults, and benzene, acetaldehyde, 1,4-dichlorobenzene and ethylbenzene all were non-negligible carcinogens (Du *et al.*, 2014a). By reviewing dozens of publications, Lyu *et al.* (2020) reported that formaldehyde was the primary carcinogen in ambient air in China, where benzene and acetaldehyde posed probable risk as well. In this study, due to the interferences of 1,2-dichlorobenzene, 1,3-dichlorobenzene, and xylenes, the ILCRs of 1,4-dichlorobenzene and ethylbenzene might be overestimated. However, the ILCR values of 1,4-dichlorobenzene and ethylbenzene only accounted for about 4.8% of the total ILCRs.

3.3 Health Risk Apportionment

Obviously, even if residents were only exposed to HAPs emitted during the household activities, the ILCRs in the lifetime exposure period were averaged at 2.60×10^{-4}, which meant that Hong Kong residents were at risk of cancer. Fig. 6 displays proportions of different indoor activities in household cancer risks. Among various household activities, incense burning caused an ILCR of 1.21×10^{-4}, accounting for 45.6% of total ILCR, mainly due to emissions of elevated formaldehyde, benzene and acetaldehyde. Furthermore, cooking (4.59×10^{-5}, 17.3%), smoking (3.23×10^{-5}, 12.1%), air freshener spraying (2.76×10^{-5}, 10.4%) and candle burning (1.78×10^{-5}, 6.7%) resulted in probable cancer risks while cleaning and coffee/tea brewing led to possible cancer risks, contributing to ILCRs of 4.18×10^{-6} (1.6%) and 1.43×10^{-5} (5.4%), respectively. The same exposure frequency was applied to each household activity to compare their unit health risks. It was found that incense burning had the highest risk with equivalent exposure frequency, followed by air freshener spraying, coffee/tea brewing, cooking, candle burning and smoking.

As the main carcinogen, ILCR of formaldehyde mostly came from incense burning (46.8%), which also contributed the most to those of benzene (54.2%) and acetaldehyde (38.8%). Differently, cooking activities (44.7%) and candle burning (25.6%) contributed the most to indoor 1,4-dichlorobenzene and ethylbenzene, respectively. Briefly, household exposure concentrations of the above-mentioned HAPs were contributed by multiple (8 to 9) indoor sources. However, PAHs mostly came from three indoor activities (i.e., smoking, candle burning and incense burning), accounting for 90.8%. It is noteworthy that the risks associated with the particulate matters generated by these activities were not considered for the risk assessment in this study, so the overall risks from these activities might be even higher.

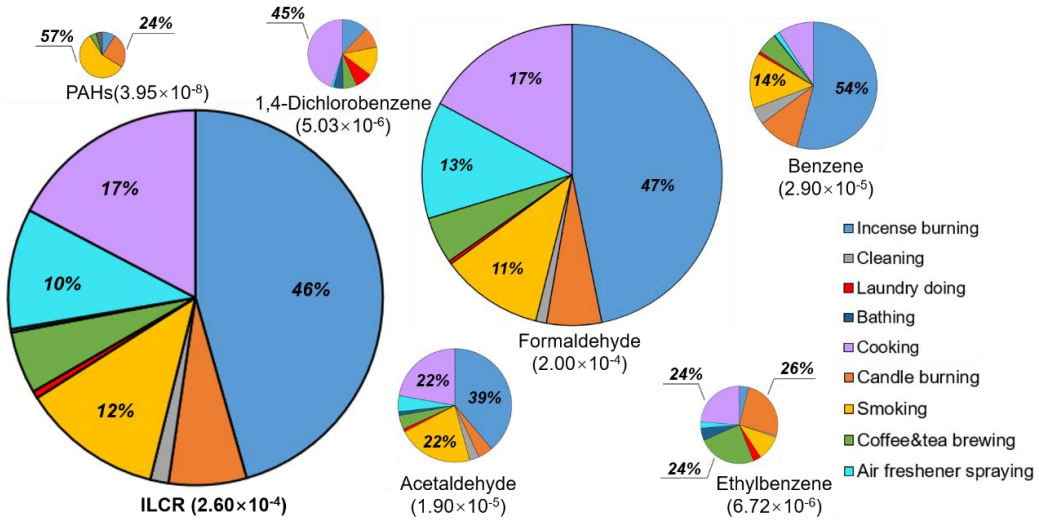

Fig. 6. Portions of different indoor activities in ILCR and cancer risks of HAPs. Pie areas are proportional to total cancer risk.

In order to estimate the household inhalation health risks comprehensively, the contributions of household background and outdoor air infiltration were estimated and summed up to total cancer risks, as shown in Fig. S4. Briefly, formaldehyde posed the highest carcinogenic risks (8.35×10^{-4}), followed by benzene (9.34×10^{-5}), acetaldehyde (4.58×10^{-5}), 1,4-dichlorobenzene (2.29×10^{-5}) and ethylbenzene (1.94×10^{-5}). Moreover, the total carcinogenic risks were divided into three exposure stages of life. It was found that the ILCRs of measured HAPs during birth–18 years of age was around 1.8 and 2.7 times those in the exposure stages of 18–60 and 60–85 years of age. This is mainly because infants and children spend more time at homes; and are much more sensible to carcinogens (Table S6). Hence, more stringent control measures should be taken to protect infants and children from potential cancer risks at homes. Specifically, the total ILCRs for the group of 18–60 years of age (2.97×10^{-4}) were at a similar magnitude of cancer risk to previous studies. We ever reported that the average ILCRs suffered by working adults were 1.13×10^{-4} in a 40-year exposure period in living rooms and bedrooms of Hong Kong (Guo *et al.*, 2004). Du *et al.* (2014a) found that the total ILCR for Chinese working adults was about 1.82×10^{-4} in a 42-year exposure period at homes. In this study, continuous measurements were conducted on indoor air pollutant levels, especially the emissions of typical household activities, which were more precise to determine the actual exposure levels of residents. In contrast, previous studies intermittently measured household air pollutants for certain duration, and emissions of intensive household activities were not well considered, thereby resulting in underestimation of overall exposure levels of residents.

Fig. S5 compares the contributions of household background release, outdoor air infiltration and household activity emissions to the health risks in the lifetime exposure period. Though their contributions to the ILCRs of different HAPs varied, the household background release were the primary contributor to cancer risk, as it accounted for over 50% of the ILCR of formaldehyde and 1,4-dichlorobenzene. In terms of non-carcinogenic health risks, the HI for respiratory system, which is an integrated indicator of multiple HAPs, was dominated (80.2%) by the household background release, while outdoor infiltration contributed the most to household NO_2, possibly attributed to surrounding anthropogenic activities, such as vehicle emissions on the street.

The above results demonstrate the prominent contributions of background release to the household inhalation health risks. The leading reason is that residents are constantly exposed to the indoor background pollution (i.e., emissions from building materials or wall/floor coverings) but intermittently to the household activities at homes. Another reason is that some key HAPs remained high levels in indoor background air, which were higher than the average concentration increments caused by most of the household activities. For example, the average concentrations of formaldehyde and propanal were 40.15 ± 1.76 and 36.24 ± 3.36 μg m^{-3} in indoor background

air, while on average it only increased by 17.48–20.78 and 4.54–21.99 μg m^{-3} during candle burning, smoking and cooking, respectively. Clearly, the health risks of the prolonged exposure to indoor emission sources such as furniture and building materials, and the continuous outdoor air pollutant infiltration were much more severe than the transient and intense household activity emissions at homes.

4 UNCERTAINTY EVALUATION

In order to better compare concentrations of HAPs in different indoor activities and background release, the sample inlet was fixed in the living room where most indoor activities (except cooking and bathing) were performed. Uncertainties arose from the spatial distribution of air pollutants in different rooms. As demonstrated in previous studies (Cheng *et al.*, 2011), the time to reach a well-mixed indoor state is typically less than one hour, usually in the range of 2–42 min., so that concentrations of HAPs in the background release were unlikely to be affected as the exposure concentrations were stable and the duration was more than 1157 min. per day. As for indoor activities, Ott *et al.* (2003) measured respirable suspended particles (RSP) concentrations in a house when a cigar was smoked in the kitchen and reported that the RSP concentrations in the living room were –25% lower than those in the kitchen, and after 45 min. they became similar. Thus, there might be an underestimation of about 25%, especially in cooking activities, as it accounted for 17% of the total ILCR, while bathing only contributed 0.3%. This meant that the probability of developing cancer was still underestimated in this study. The findings deepened our understanding of the health risks posed by household air pollution.

It should be noted that the sampling campaign was only conducted in one Hong Kong home for three months herein. This was attributed that in this study we focused on the species profiles of HAPs emitted during various household activities and quantified the contributions of each potential household source regardless of the influences of background release, outdoor infiltration and ventilation system in different household environments. According to Fig. 1, concentrations of most HAPs in the same activity showed good reproducibility, reflecting the typical species profiles and concentrations of HAPs for each household activity. Some previous studies also evaluated lifetime health risks in specific indoor environments based on limited sample size (i.e., 27 samples) (Mo *et al.*, 2021) and limited sampling period (i.e., 30 days in a restaurant) (Arı *et al.*, 2020). Seasonal effects on meteorological conditions (i.e., temperature and relative humidity) and activity patterns (i.e., the use of air conditioners) would cause uncertainty under the premise of limited sampling days. Generally, high temperature and humidity might enhance the release of some air pollutants, i.e., formaldehyde. The average outdoor temperature and humidity during the sampling period were 9% and 16% lower than the yearly average (HKO, 2019), which indicated a possible underestimation of concentrations of air pollutants. Less indoor pollutants were reported in summer due to stronger air ventilation caused by more frequent use of air conditioners and larger indoor/outdoor temperature differences, compared to those in winter (Chamseddine *et al.*, 2019; Abdel-Salam, 2021). In some studies (Dėdelė and Miškinytė, 2016; Stamp *et al.*, 2021), however, the differences in indoor concentrations between warm and cold seasons were without significance ($p \geq 0.05$). The sampling campaign in autumn and winter generated a possible overestimation of indoor concentrations of pollutants in this study.

Uncertainty might also be induced by the vertical distribution of outdoor air pollutants. As indicated previously (Hong *et al.*, 2022), concentrations of air pollutants (i.e., NO_2 and formaldehyde) decreased obviously as the altitude increased within 0–3 km with a height resolution of 200 m. Nevertheless, concentrations of air pollutants were more stable within 200 m which was higher than most of residential buildings (Samad *et al.*, 2020). Villa *et al.* (2017) reported that the impacts of ground-level vehicle emissions on the concentrations of air pollutants (i.e., NO_2) were constrained within the height of 0–40 m, and the declining trend of concentrations became gentler above 30 m. Additionally, the vertical mixing of pollutants would be better during the daytime when no temperature inversion was observed. To sum up, residences living in lower floors were tended to be influenced more by the outdoor infiltration and obtained higher health risks.

Overall, the limitations in the number of residences and sampling days might bring uncertainties

to the evaluation of health risks, thus further sampling campaigns should be performed in the future to obtain more accurate and comprehensive results.

5 CONCLUSIONS

An intensive sampling campaign was conducted from October to December of 2019 in an apartment of Hong Kong. In total, over 28 main HAPs including VOCs, PAHs and trace gases were identified in the household environment, while outdoor air pollution significantly affected the indoor background levels of trace gases, PAHs, heptanal and trimethylbenzenes/acetophenone. However, other HAPs mainly originated from the prolonged emissions of indoor sources, such as building and furniture materials, indoor deodorants, rubber and paint. Although household activities caused obvious increase in concentrations of many HAPs indoors, the indoor background pollution dominantly contributed to some key HAPs. For example, the average concentration of formaldehyde was 40.15 $\pm$ 1.76 $\mu g\ m^{-3}$ in indoor background air, which was about two times higher than the average concentration increments of formaldehyde during candle burning (19.39 $\mu g\ m^{-3}$), smoking (17.48 $\mu g\ m^{-3}$), and cooking (20.78 $\mu g\ m^{-3}$), respectively.

Non-carcinogenic health risk assessments found that acrolein and NO_2 likely had chronic health risks to residents in all exposure stages. In addition, health risks of many HAPs to the respiratory system of residents were found in all age groups. Furthermore, the acute health risks of formaldehyde, benzene, NO_2 and acrolein were observed in normal urban daily life for the first time, caused by the transient and intensive emissions of household activities such as incense burning, smoking, and cooking. In terms of carcinogenic risks induced by household activities, formaldehyde presented highest risks, followed by benzene, acetaldehyde, ethylbenzene and 1,4-dichlorobenzene, all of which exceeded the acceptable benchmark. Probability distributions of health risks revealed that carcinogenic risks of most HAPs were well fitted to lognormal distribution, except benzo[a]pyrene, which was fitted to gamma distribution better. Further looking into contributions of indoor activities, incense burning contributed the most to carcinogenic risks, followed by cooking, smoking and air freshener spraying. Compared to PAHs, other HAPs were derived from more diverse indoor sources. Besides, the minor (birth–18 years of age) suffered from the most severe cancer risks, approximately 1.8–2.7 times higher than that faced by working adults (18–60 and 60–85 years of age). Health risk apportionment indicated that household background release was the major source of household inhalation health risks, contributing over 50% to the total and 80.2% to HI for respiratory system.

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SUPPLEMENTARY MATERIAL

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