# The Effects of Height on the Accumulation of *n*-Alkanes and Polycyclic Aromatic Hydrocarbons (PAHs) in Air-Conditioning Filter Dust from High-Rise Apartments

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## ABSTRACT

To assess the effect of floor levels of high-rise apartment buildings on the accumulation of contaminants in indoor environment, residential air-conditioner filter dust (ACFD) samples from the 1st, 10th, 20th and 30th floors of a high-rise apartment building were collected for the determination of *n*-alkanes and polycyclic aromatic hydrocarbons (PAHs). The results show that both *n*-alkanes and PAHs in the residential ACFD were ubiquitous but varied greatly in concentrations. The total concentrations of 27 *n*-alkanes ($\Sigma_{27}AK$) and 16 PAHs ($\Sigma_{16}PAH$) ranged from 1.35 to 9290 µg g$^{-1}$ and 278–34200 ng g$^{-1}$, respectively. Source apportionment revealed that *n*-alkanes were from mixed sources combining fossil fuel combustion, natural emission and solid biomass burning, but PAHs were mainly from indoor sources. Furthermore, the diagnostic ratio of paired low weight molecular PAH species may change during transportation and accumulation. Significantly higher concentrations of $\Sigma_{27}AK$ and $\Sigma_{16}PAH$ were observed in the samples from low floor levels (the 1st and 10th floors) compared to those from high floor levels (the 20th and 30th floors). The results of classification and regression tree analysis clearly suggested floor level is the most important factor influencing the accumulation of $\Sigma_{27}AK$ and $\Sigma_{16}PAH$ in the ACFD. Our findings imply that people living on lower floor levels have greater exposure risks to PAHs associated with indoor dust.

*Keywords:* High-rise apartment; Floor level; Air-conditioner filter dust; *n*-alkanes; PAHs.

## INTRODUCTION

Exposure to indoor air contaminants is a vital health risk for urban populations because they generally spend more than 80% of time indoors (Mirhoseini *et al.*, 2016). However, with rapid urbanization, people living in countryside tend to migrate to the cities for better work opportunities and livings. It is reported more than 50% of the world's population resided in urban areas in 2013, compared to only 40% in 1980 (The World Bank, 2016). The United Nations Department of Economic and Social Affairs (DESA)'s Population Division also noted that there were 28 mega-cities worldwide in 2014, home to approximate 12% of global urban populations (United Nations Department of Economic and Social Affairs, 2014). As a result, the increase of urban population has a direct influence on the types of dwellings that people live in. High-rise apartment buildings are common in metropolitan regions worldwide, especially in China. Previous studies have demonstrated that the height of the dwelling in urban areas is the key factor influencing the wind speed (Murakami *et al.*, 1979; Lu and Ip, 2009), microenvironment (Lee and Jo, 2006; Gao *et al.*, 2008) and inhabitant's health (Drennan *et al.*, 2016). More importantly, floor levels can influence indoor-outdoor exchange of contaminants, resulting in significant vertical variation of indoor and outdoor ambient pollution (Jo and Kim, 2002; Jo and Lee, 2006). The exposure to indoor airborne pollutants for the residents living in different floor levels may vary accordingly. Thus the information on how the floor level affects the indoor air quality lays groundwork for a comprehensive assessment of inhalation exposure risks for urban populations living in high-rise buildings. However, such information has not been well addressed to date.

The *n*-alkanes and polycyclic aromatic hydrocarbons (PAHs) are two groups of ubiquitous indoor environmental pollutants, both of which are commonly used to indicate the pollution significance because of their widespread sources including indoor and outdoor emissions (Huang *et al.*, 2015; Jung *et al.*, 2015; Mwangi *et al.*, 2015; Wang *et al.*, 2015). Furthermore, PAHs have obtained extensive attention because of their persistency and carcinogenicity nature (Zhang *et al.*, 2015). Thus numerous studies applied PAHs as model pollutants to assess the health risks (Bhargava *et al.*, 2004; Zhou and Zhao, 2012; Kavouras *et*

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*al.*, 2015; Lee *et al.*, 2016; Mohammed *et al.*, 2016). Due to the importance of indoor exposure, PAHs in indoor air (including gas phase and particle) and floor dust are commonly investigated to elucidate potential health risks (Essumang *et al.*, 2015; Tiwari *et al.*, 2015). However, most of these sampling strategies demand specialized equipment and are time-consuming and noise nuisance. Furthermore, there are many uncertainties with the results of health assessment based upon these monitoring data due to temporal and spatial variations of contaminant concentrations. For example, significantly higher concentrations of PAHs in both gaseous and particle phases were observed in winter season than in summer (Masih *et al.*, 2012). Muenhor and Harrad (2012) also demonstrated that floor dust from the same room and building varied greatly in the concentrations of polybrominated diphenyl ethers. Dust accumulated on the air condition filter (ACFD) may be a better candidate to elucidate indoor exposure risks because it can integrate contamination during a given time interval (Ni *et al.*, 2011; Chou *et al.*, 2016).

The main objectives of the present study were to investigate *n*-alkanes and PAHs in ACFD from a high-rise apartment building, and further to evaluate the potential exposure risk to indoor PAHs for the dwellers. Special attention was paid to the effect of floor levels on the accumulation of hydrocarbon compounds and associated human health risk.

## MATERIALS AND METHODS

### Sampling Location and Sampling

The City of Hefei is a capital city and the political, economic, and cultural center of Anhui Province in Eastern China. It is located in the middle altitude zone (30°57'–32°32'North latitude and 116°41'–117°58' East longitude) with an annual precipitation of ~1,000 mm. The city has a population of 7,790,000 inhabitants with over 70% of them living in urban areas. Undoubtedly, rapid population growth and urbanization has resulted in a concomitant increase on the high –rise apartment buildings in urban areas.

ACFD samples were collected from a new high-rise apartment building in Hefei. The apartment has 30 floors (about 90 m in height) and was built in 2006. Additional five buildings with the same height and floor levels are around the study building with the space between each other approximately 20 m. A total of 44 ACFD samples were collected in August 2014. Among these samples, seven of them (two in bedrooms and five in living rooms) were collected from seven homes on the 1st floor;thirteen samples (seven in bedrooms and six in living rooms) from nine homes on the 10th floor; eleven samples (eight in bedrooms and three in living rooms) from five homes on the 20th floor; thirteen samples (ten in bedrooms and three in living rooms) from five homes on the 30th floor. Less than 0.2 g to 1.3 g of dust particles on impervious surface of each air-conditioner filter were collected using plastic brush and dust pans by gentle sweeping motion. Before each sampling, brush and dust pans were cleaned with acetone and paper towels. All samples were wrapped with

solvent-rinsed aluminum foil and stored in polyethylene zip bags. After transported to the analytical laboratory, dust samples were stored at –20°C prior to chemical analysis.

A questionnaire survey was conducted to understand the potential indoor sources of hydrocarbons. All participating families returned with valid questionnaire. Most participants had ages ranging from 20 to 50 years old. The rooms had been interiorly decorated before the participating families moved in. There is noventilation and central air-conditioner system in the apartment, and only independent air conditionings were used during summer seasons. All but one family had significant cooking activities (generally 2–3 times d$^{-1}$) and only four families were impacted by indoor smoking. Six filters of air-conditioners in three families had been cleaned before the summer.

### Chemical and Reagents

*n*-hexane, dichloromethane, acetone and methanol supplied by Oceanpak Alexative Chemical Co., Ltd (Gothenburg, Sweden) were of pesticide grade. Neutral silica gel (80–100 mesh) was purchased from Guangzhou Xinshi Chemistry Experimental Equipment (Guangzhou, China) and was pre-cleaned by Soxhlet extraction for 48 hours using methanol and dichloromethane, respectively, prior to use. Sodium sulfate obtained from Sigma-Aldrich (St Louis, MO, USA) was baked at 450°C for 4 hours and stored in sealed containers. All glassware was cleaned with chromic acid and tap water, followed by rinsing with deionized water, and then baked at 450°C for 4 hours.

A mixture standard of *n*-alkanes including $nC_{10}$, $nC_{12}$, $nC_{14}$, $nC_{16}$, $nC_{18}$, $nC_{20}$, $nC_{22}$, $nC_{24}$, $nC_{26}$, $nC_{28}$, $nC_{30}$, $nC_{32}$, $nC_{32}$, $nC_{34}$, $nC_{36}$ and $nC_{38}$ was purchased from AccuStandard (New Haven, CT, USA). The standard solutions of $nC_{24}$–$d_{50}$ and $nC_{30}$–$d_{62}$ at 1000 µg mL$^{-1}$ each were purchased from Cambridge Isotope Laboratory (Anbe, MA, USA) as surrogate and internal standards of *n*-alkanes to monitor the performance of sample extraction and cleanup, respectively. A mixture solution containing sixteen PAHs identified as priority pollutants by the United States Environmental Protection Agency (USEPA), i.e., nathphalene (Nap), acenaphthylene (Ace), acenaphthene (Ac), fluorene (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthrancene (DahA), and benzo[g,h,i]perylene (BghiP), was purchased from AccuStandard. A deuterated standard mixture solution containing five PAHs (Nap-$d_8$, Ace-$d_{10}$, Phe-$d_{10}$, Chr-$d_{12}$, andperylene-$d_{12}$) at 100 µgm L$^{-1}$ each was purchased from Cambridge Isotope Laboratory (Massachusetts, USA) and applied as surrogate standards of PAHs. Reference standards of 2-fluoro-1,1-biphenyl, *p*-terbenyl-$d_{14}$, and DahA-$d_{14}$ at 100 µg mL$^{-1}$ each (Cambridge Isotope Laboratory, USA) were used as internal standards.

### Extraction and Analysis of PAHs

The procedures of extraction and purification of hydrocarbon compounds were described elsewhere (Chuang *et al.*, 1995). Briefly, the dust samples were spiked with a

known amount of $nC_{24}–d_{50}$ and five deuterated PAHs as surrogate standards and then extracted with 15 mL of a mixture of dichloromethane and hexane (50:50 in volume) in an ultrasonic bath for 30 min. The extraction was repeated three times. The combined extract was filtered through a 7 µm glass fiber filter (Whatman International, Maidstope, UK) and then concentrated to 1 mL. The condensed extract was further solvent exchanged to hexane by adding 10 mL of hexane and then reduced to 1 mL again. The sample was purified by passing through a glass column packed with 3 g of silica gel. After the column was conditioned with 5 mL of hexane and loaded with the sample, the first fraction containing *n*-alkanes was eluted using 12 mL of hexane, and the second fraction containing PAHs was eluted with 15 mL of a mixture of hexane and dichloromethane (7:3 in volume). Both fractions were condensed to 500 µL and spiked with known amounts of internal standards.

Both *n*-alkanes and PAHs were instrumentally analyzed using an Agilent 7890A gas chromatograph coupled with a 5975C mass spectrometer (Agilent Technologies, Palo Alto, California, US) in electron impact (EI) ionization mode. A 60 m × 0.25 mm × 0.25 µm DB-5 capillary column (Agilent Technologies) was employed for chromatographic separation. Ultra high purity helium (99.999%) was used as carrier gas with a constant flow rate of 1 mL min$^{-1}$. Sample (1 µL) was injected using an automatic sampling system AOC-20 Series in the splitless mode, and the temperature of injector was held at 280°C. The temperatures of ion source and quadrupole were both set at 150°C, and the transfer line temperature was at 280°C. For *n*-alkanes, the oven temperature was programmed from 60 (held 2 min) to 180°C at a rate of 8 °C min$^{-1}$, to 220 °C at a rate of 2 °C min$^{-1}$, and then to 320°C at a rate of 8 °C min$^{-1}$. For PAHs analysis, the GC oven was temperature-programmed from 60°C to 200°C at a rate of 10 °C min$^{-1}$, and to 250°C at a rate of 2 °C min$^{-1}$, then to 290°C at a rate of 20 °C min$^{-1}$, finally held isothermally for 5 min. The quantification and confirmation of each analyte was achieved via selected ion monitoring (SIM) mode. All *n*-alkanes with even numbers of carbon atoms were directly quantified by the calibration curves built using their corresponding reference standards at eight calibration concentrations (0.05, 0.1, 0.2, 0.5, 1, 2, 5, and 10 µg mL$^{-1}$). Each calibration solution contained 1.0 µg mL$^{-1}$ of internal standards. Odd number carbon *n*-alkanes were qualified with the average response factor of two adjacent even-numbered carbon components, given that their reference standards were not available (Wang *et al.*, 2012).

### *Quality Assurance and Quality Control (QA/QC)*

Quantification was performed using an internal calibration method based on eight-point calibration curves with regression coefficient ($R^2$) value higher than 0.99 for all target compounds. For every ten samples, a calibration solution containing 1.0 µg mL$^{-1}$ each of *n*-alkanes or 0.5 µg mL$^{-1}$ each of PAHs was analyzed to monitor the instrument performance. For each batch of 15 dust samples, one solvent blank was processed to monitor laboratory contamination. Solvent blanks contained *n*-alkanes and PAHs less than 10% of the mean concentrations from all

dust samples. The reporting limits (RLs) were calculated at 0.078 µg g$^{-1}$ for all *n*-alkanes and 7.76 ng g$^{-1}$ for all PAHs after including the lowest concentrations of the calibration curves and average sample weight into consideration (Wang *et al.*, 2007). The average recovery efficiency of $nC_{24}–d_{50}$ was 100.6 ± 25.9%, and five deuterated PAHs had the recoveries of 88.8 ± 23.3% for Nap-$d_8$, 87.2 ± 21.1 for Ace-$d_{10}$, 88.8 ± 21.8 for Phe-$d_{10}$, 104.4 ± 20.4 for Chr-$d_{12}$, and 107.0 ± 16.0 for perylene -$d_{12}$.

### *Data Analysis*

Because $nC_{37}$ and $nC_{38}$ were not detected in any sample, 27 *n*-alkane homologues (from $nC_{10}$ to $nC_{36}$) were discussed in the present study and their total concentration was referred to as $\Sigma_{27}AK$. The total concentration of 16 PAH compounds was defined as $\Sigma_{16}PAH$. For any measured concentration below RL, zero was used for concentration calculation. Data analyses were conducted using Excel 2007 (Microsoft Corporation, Silicon Valley, USA) and the statistical analyses including normality test and principal component analysis were performed using SPSS 16.0 (Chicago, IL, USA).

To assess the potential sources of *n*-alkanes in ACFD, carbon preference index (CPI) and plant wax alkanes (WaxCn(%)) were calculated according to the following equations:

$$CPI_1 = \frac{\sum odd(nC_{10} - nC_{36})}{\sum envn(nC_{10} - nC_{36})} \tag{1}$$

$$CPI_2 = \frac{nC_{27} + nC_{29} + nC_{31} + nC_{33}}{nC_{28} + nC_{30} + nC_{32} + nC_{34}} \tag{2}$$

$$WaxC_n(\%) = \frac{\sum \left[ n\text{-}C_n - \frac{1}{2}(n\text{-}C_{n-1} - n\text{-}C_{n+1}) \right]}{\sum (n\text{-}C_{15} - n\text{-}C_{34})} \tag{3}$$

It should be noted that $CPI_1$ was usually used to discriminate between petrogenic and biogenic sources of total *n*-alkanes; in general, $CPI_1$ close to 1 suggests incomplete combustion of fossil fuel, and it indicates contemporary biological sources when $CPI_1$ is greater than 3 (Simoneit, 1984). $CPI_2$ is utilized to distinguish the fraction of *n*-alkanes from high plant wax, a high value usually indicative of high plant wax input (Reddy *et al.*, 2000). WaxCn(%) is close to zero for petroleum or crude oil residue, whereas it approaches 100 for high terrestrial plants or marine plants (Gogou *et al.*, 1996).

In the present study, the classification and regression tree analysis was employed to uncover the relationships and interactions between PAHs or *n*-alkanes contents in ACFD and floor levels or room type (bedroom or living room) variables (JMP software, Version 10.0, SAS institute, Cary, NC, USA). As a nonparametric modeling method, the classification and regression tree analysis can justify the relationship between variables from a set of independent variables and recursively separate the data to obtain

increasingly homogeneous subsets based upon the independent variable splitting criteria using variance minimizing algorithms (Zheng *et al.*, 2009).

To better parameterize ACFD carcinogenicity related to the whole PAH fraction, the concentrations of PAHs in ACFD were further transferred to BaP equivalent concentration ($BaP_{eq}$) using individual toxic equivalency factor (TEF) (Pongpiachan, 2015). TEF values are 0.001, 0.001, 0.001, 0.001, 0.001, 0.01, 0.001, 0.001, 0.1, 0.01, 0.1, 0.1, 1, 0.1, 1, and 0.01 for Nap, Ace, Ac, Fl, Phe, Ant, Flu, Pyr, BaA, Chr, BbF, BkF, BaP, IcdP, DahA and BghiP, respectively (Yang *et al.*, 2015).

## RESULTS AND DISCUSSION

### *n-Alkanes in Residential ACFD*

The total concentrations of $\Sigma_{27}AK$ in residential ACFD ranged from 1.35 to 9290 µg g$^{-1}$dw. Because logarithm-transformed the concentration of $\Sigma_{27}AK$ followed normal distribution ($p = 0.244$, with One - Sample Kolmogorov - Smirnov Test in SPSS), the geometric mean was reported at 321 µg g$^{-1}$dw. Despite all *n*-alkane homologues between $nC_{10}$ to $nC_{36}$ were detectable in each residential ACFD, the predominated congeners belong to $nC_{24}$–$nC_{33}$ homologues, and the carbon maximum number ($C_{max}$, defined as the homologue with the highest concentration among all *n*-alkanes) was observed at $nC_{27}$, $nC_{29}$ or $nC_{31}$ in 6, 23 or 8 out of 44 samples, respectively. Interestingly, samples from the 10$^{th}$ floor generally contained $C_{max}$ at $nC_{27}$, while the $C_{max}$ was mainly at $nC_{29}$ for samples form the 20$^{th}$ and 30$^{th}$ floors. The $C_{max}$ in samples from the 1$^{st}$ floor was complex, i.e., one sample at $nC27$, one at $nC_{29}$, three at $nC_{31}$ and two at $nC_{33}$. These suggest that *n*-alkanes in most samples were

partly contributed by biological origins (Eglinton and Hamilton, 1967; Bi *et al.*, 2005b) or combustion of biomass (bin Abas *et al.*, 1995; Wang *et al.*, 2009). The left part of Fig. 1 illustrates the patterns of *n*-alkanes as a function of carbon number. The results show unimodal distribution without significant odd-carbon number preference. $CPI_1$ and $CPI_2$ values varied between 0.72–2.8 and 0.98–3.2 with arithmetic means of $1.4 \pm 0.38$ and $1.7 \pm 0.44$, respectively, suggesting *n*-alkanes in residential ACFD were predominately from anthropogenic activities combining biological materials. Human activities related sources of *n*-alkanes contain CPI values close to unit, whereas natural sources from vegetation generally exhibit high CPI values ($> 3.0$) (Bi *et al.*, 2005a; Wu *et al.*, 2007). Significantly higher $CPI_2$ values than $CPI_1$ suggest that long-chain alkanes in residential ACFD were more dominated by biogenic sources when compared with short-chain homologues. $WaxC_n(\%)$ value was determined in a range of 4.4%–24.9% with an arithmetic mean of 13.2 $\pm$ 4.3%, indicating a predominant contribution from anthropogenic sources. A similar contribution of *n*-alkanes in urban aerosol from biogenic origins was also investigated (Bi *et al.*, 2003; Lin and Lee, 2004; Feng *et al.*, 2006; Wu *et al.*, 2006). $WaxC_n(\%)$ significantly correlated against $CPI_1$ (Fig. 1(b)). Therefore, $CPI_1$ could be expected as a useful indicator for the prediction of biogenic sources derived *n*-alkanes.

PCA analysis shows three main factors which contributed the majority of the variance (87.0%). Factor 1 (accounting for 46.5% of the total variance) was mostly associated with short-chain alkanes (from $nC_{10}$ to $nC_{23}$), suggesting fossil fuel combustion (Cincinelli *et al.*, 2007; Alves *et al.*, 2016). Factor 2 contributing to 32.7% of total variance was mostly explained by high loadings of long-chain *n*-alkanes,
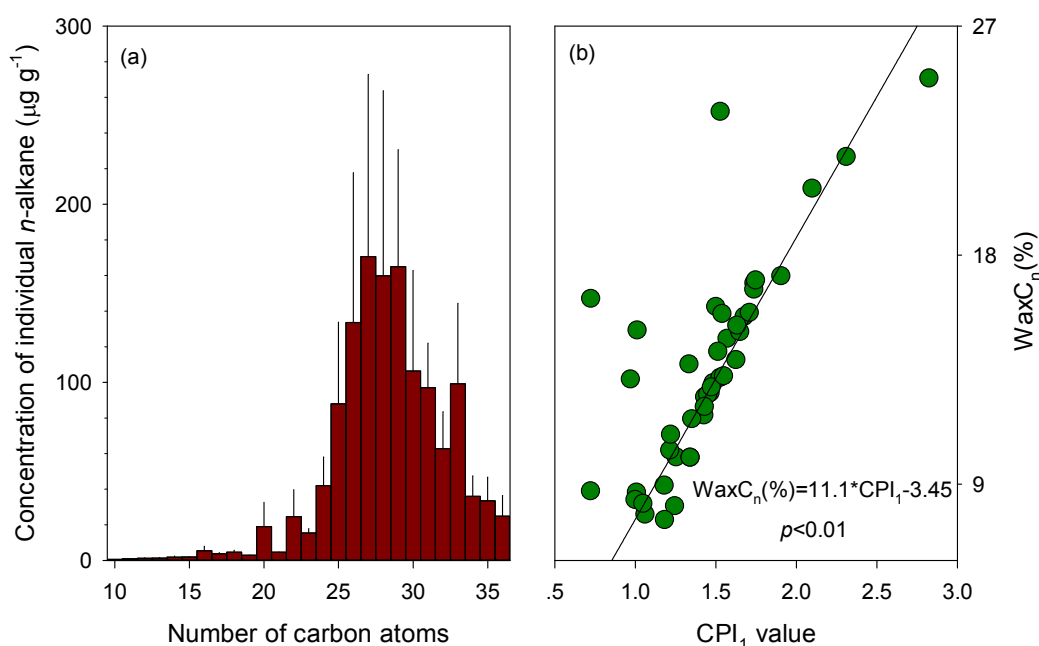


**Fig. 1.** The profile of *n*-alkanes in the air-conditioner filter dust (ACFD) from a high-rise apartment building (a), and the correlation between carbon preference index ($CPI_1$) and plant wax alkanes ($WaxC_n(\%)$) (b). It is noted that $CPI_1$ and $WaxC_n(\%)$ were calculated based upon Eqs. (1) and (3), respectively.

indicating natural sources because long-chain alkanes are largely emitted from plant wax (Eglinton and Hamilton, 1967; Bi *et al.*, 2005b). The third factor (factor 3) could explain 7.8% of the total variance, and was dominated by $nC_{21}$–$nC_{26}$. Previous studies demonstrated that solid fuel and biomass burning would produce moderate –chain alkanes (Oros and Simoneit, 2001; Bi *et al.*, 2008; Sun, 2012). Therefore, residual *n*-alkanes in residential ACFD were from mixed sources combining fossil fuel combustion, natural emission and solid biomass burning.

### Occurrence of PAHs in Residential ACFD

Nap, Fl, Phe, Flu, Pyr, and Chr were detected in all ACFD samples with the concentration ranges of 14.2–2240, 10.0–376, 56.0–3290, 51.7–5030, 36.8–4090, and 36.4–3610 ng $g^{-1}$, respectively (Fig. 2). Only 15 samples contained Ace greater than the report limit, and Ace was observed with the lowest mean concentration (5.1 ± 37.4 ng $g^{-1}$) among all target PAHs. The detection frequencies of Ac, Ant, BaA, B(b+k)F, BaP, IcdP, DahA and BghiP were 63.7%, 93.1%, 84.1%, 97.7%, 76.3%, 93.2%, 50.0% and 90.9%, respectively. Therefore, the residential ACFD was dominated by high molecular weight PAHs (from 4 to 6 rings), accounting for 68.6 ± 16.8% of the total concentration of $\Sigma_{16}$PAH in the present study. Low molecular weight PAHs with high volatility appeared to less accumulate in the ACFD (Zhou *et al.*, 2010). Because high molecular weight PAHs are more toxic and persistent in environment, TEF-adjusted concentrations of high molecular weight PAHs were 2–3 orders of magnitude higher than that of low molecular weight PAHs. The total concentration of $\Sigma_{16}$PAH varied from 278 to 34200 ng $g^{-1}$ and appeared to have a lognormal distribution (*p* = 0.84, with One - Sample Kolmogorov - Smirnov Test in SPSS). The geometric mean of $\Sigma_{16}$PAHs in the residential ACFD was determined at 2280 ng $g^{-1}$ and $BaP_{eq}$ concentrations ranged from 2.27 to 5290 ng $g^{-1}$.

Zhou *et al.* (2010) reported the total concentrations of PAHs in ACFD of different indoor environments, i.e., 4895 ng $g^{-1}$ for household, 4231 ng $g^{-1}$ for office, 475 ng $g^{-1}$ for meeting room, 740 ng $g^{-1}$ for chemical laboratory, and 13090 ng $g^{-1}$ for restaurant. ACFD samples from the commercial office, secondary school, shopping mall, hospital, electronic factory and manufacturing plant were also found with the total concentration of PAHs at 5850 ± 3190, 5610 ± 2330, 5530 ± 2070, 3720 ± 1820, 4090 ± 2380 and 22200 ± 6050 ng $g^{-1}$, respectively (Kang *et al.*, 2010). The concentrations of PAHs in ACFD were comparable to or slightly lower than those in door floor dust (Gevao *et al.*, 2007; Mannino and Orecchio, 2008; Peng *et al.*, 2012; Qi *et al.*, 2014). Therefore, our results, along with previous studies, indicate that dust retained on the AC filter is suitable for the evaluation of health risks from indoor exposure to indoor contaminants because they can reflect both the indoor dust and indoor air quality through air recirculation.

Paired isomers based diagnostic ratios of Ant/(Ant + Phe), Flu/(Flu + Pyr), BaA/(BaA + Chr) and IcdP/(IcdP + BghiP) were 0.07 ± 0.02, 0.58 ± 0.05, 0.16 ± 0.08 and 0.64 ± 0.11, respectively. High ratio values for Flu/(Flu + Pyr) and IcdP/(IcdP + BghiP) indicate incomplete combustion of fossil fuels and biomasses (Yan *et al.*, 2005). Cooking would also be expected as an important source of residual PAHs in ACFD because of the high value of BaP/BighP (2.26 ± 3.37) (Zhang *et al.*, 2009). However, low ratios of Ant/(Ant + Phe) and BaA/(BaA + Chr) suggest petroleum-related sources (Mwangi *et al.*, 2015). Therefore, the identification of the sources of residual PAHs in ACFD remained difficult, possibly due to the change in the diagnostic ratios during the transportation and accumulation. Previous studies indicated that subtle variation in the physicochemical properties of the paired PAH species would alter the ratios from sources to receptors (Schauer *et al.*, 1996; Zhang *et al.*, 2005). For example, Ant was more susceptible to degradation than Phe, resulting in a reduced
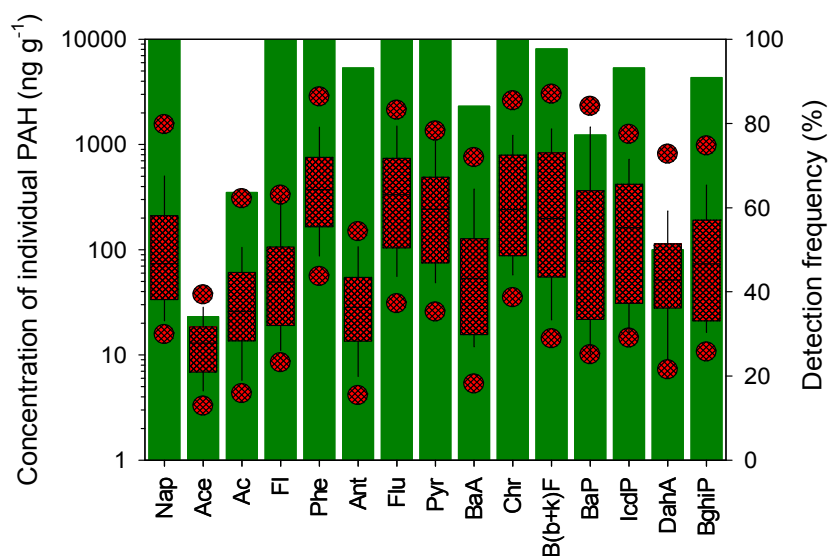


**Fig. 2.** The concentration distribution and detection frequency of individual PAHs in air-conditioner filter dust (ACFD) from a high-rise apartment building.

Ant/(Ant + Phe) ratio in pine needles (Hwang *et al.*, 2003). In the present study, very low ratios of Ant/(Ant + Phe) observed in ACFD would be explained as a result of the preferential decay or volatilization of Ant in the aged ACFD. However, ratios of high weight molecular PAH species, e.g., IcdP/(IcdP + BighP), were stable during the transportation (Wang *et al.*, 2011). High IcdP/(IcdP + BighP) ratios in the present study suggest sources from biomass produced by cigarettes burning (Du *et al.*, 2006) and cooking (Ye *et al.*, 2013). Therefore, PAHs accumulated in residential ACFD would be mainly from indoor sources.

### The Effect of Floor Level on the Accumulation of PAHs and n-Alkanes in ACFD

The results of classification and regression tree analysis indicate that greater concentrations of $\Sigma_{27}$AK and $\Sigma_{16}$PAH were determined in the samples collected from lower floor levels (1st and 10th floor) compared with higher floor levels (20th and 30th floor) (Figs. 4(a) and 4(b)). The mean concentrations of $\Sigma_{27}$AK and $\Sigma_{16}$PAH in ACFD from lower floor levels were 4.6 and 5.2 times greater than the concentrations from higher floor levels. Samples from the 1st and 10th floors contained comparable $\Sigma_{27}$AK concentrations ($p = 0.09$), and the concentrations in the dust from the 20th and 30th floors were also comparable ($p = 0.28$). Room type (bedroom and living room) did not significantly influence *n*-alkanes accumulation in the ACFD, but influenced PAHs accumulation. For example, greater concentrations of PAHs were found in ACFD from bedrooms versus living rooms located on the 10th floor ($p < 0.01$).

Obviously, the floor level is a critical factor influencing the accumulation of hydrocarbon contaminants in residential ACFD. Residents living on lower floor levels of high-rise buildings are exposed to elevated concentrations of particle-associated PAHs. Previous studies demonstrated that both outdoor and indoor airborne contaminants such as carbon monoxide, fine particles, volatile organic compounds (VOCs) and PAHs were significantly higher at low floor levels than at high floor levels (Jo *et al.*, 2003; Jo and Lee, 2006; Jung *et al.*, 2011).Therefore, our findings strongly supported the conclusion on a decreasing trend in indoor nonvolatile PAHs concentration with increasing floor levels. Additionally, concentrations of $\Sigma_{16}$PAH were significantly correlated with $\Sigma_{27}$AK ($p \leq 0.01$) in the samples from low floor levels (1st and 10th floors), but they were not related in the ACFD collected from high floor levels (Fig. 3). These results imply that PAHs and *n*-alkanes had similar sources and accumulative processes in ACFD. A number of studies attributed some indoor air pollutants mainly to the input from outdoor anthropogenic sources (Cheruiyot *et al.*, 2015). However, we were unable to determine the relative importance of outdoor and indoor sources in their contributions to *n*-alkanes and PAHs concentrations in residential ACFD, given that, PAHs in outdoor aerosol were not determined in the present study. It should be noted that the samples from the 20th and 30th floors generally contained $C_{max}$ at $nC_{29}$, but those with $C_{max}$ at $nC_{31}$ were mostly from the 1st and 10th floors. The $nC_{31}$ is usually considered as a chemical marker of grass plants, whereas $nC_{29}$ is mainly from wax plants (Oros *et al.*, 2006). Thus lower and higher floors of the apartment building are more affected by grass and wax plants, respectively. These results imply significant outdoor sources.

### Implication

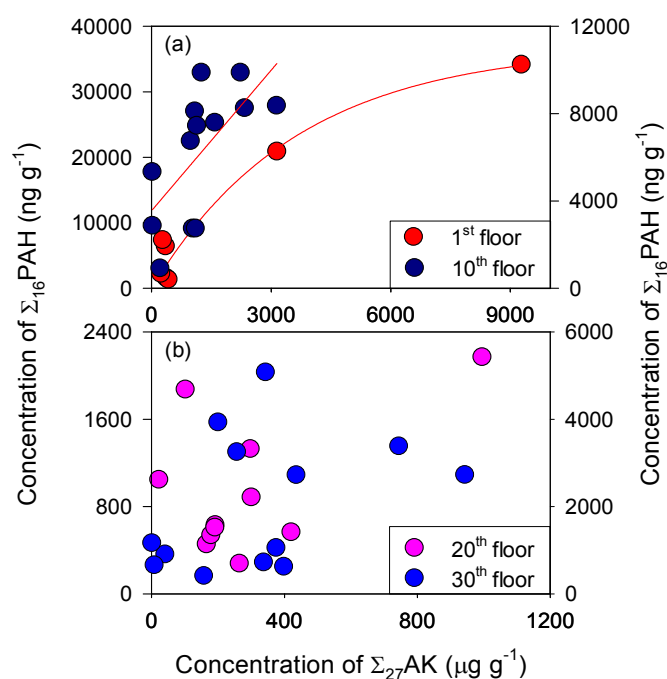Both concentrations of *n*-alkanes and $\Sigma_{16}$PAH varied over



**Fig. 3.** Plot of $\Sigma_{27}$AK and $\Sigma_{16}$PAH in air-conditioner filter dust (ACFD) from the 1st and 10th floors (a) and from the 20th and 30th floors (b).

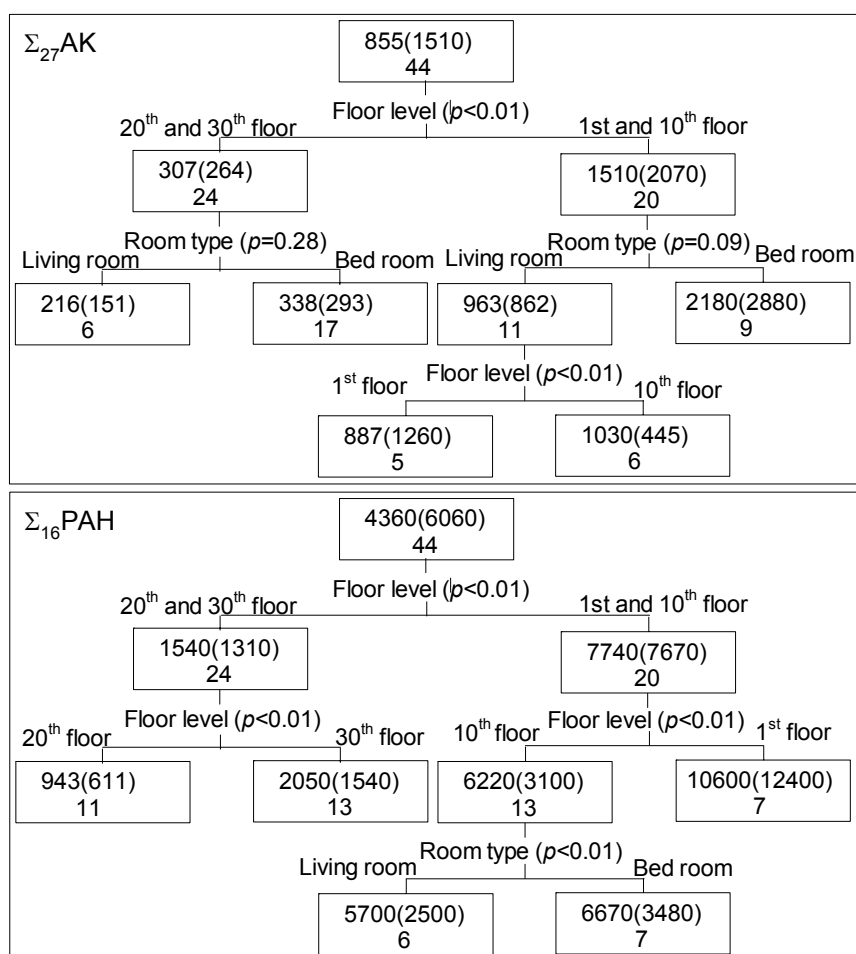**Fig. 4.** Regression tree predicting$\Sigma_{27}$AK (a) and $\Sigma_{16}$PAH (b) from floor levels and room types. Each node (rectangle) is labeled with mean (standard deviation, with a unit of µg g$^{-1}$ for $\Sigma_{27}$AK and ng g$^{-1}$ for $\Sigma_{16}$PAH) and the number of samples. The model is read using a top-down approach until terminal nodes appear.

2–3 orders of magnitude in the present study and exhibited significant spatial variations within apartments or rooms. This was mainly due to large variations in contamination on different floor levels of the building. Additional factors, such as the dwellers' living habits (smoking and nonsmoking, cooking and seldom cooking) or additional emission sources, may also contribute to temporal and spatial variations in *n*-alkanes and PAHs concentrations in ACFD (Mannino and Orecchio, 2008; Harrad *et al.*, 2009; Jung *et al.*, 2011; Muenhor and Harrad, 2012; Qi *et al.*, 2014)). Particularly, indoor smoking and cooking are generally considered as the primary contributors of indoor dust bound PAHs (Ren *et al.*, 2006; Qi *et al.*, 2014). Nevertheless, there were no significant differences in concentrations between the samples from smoking and nonsmoking or between cooking and noncooking apartments (all $p > 0.05$), likely due to insufficient number of samples for statistical analysis.

Overall, *n*-alkanes and PAHs were investigated in the residential ACFD from different floor levels of a high-rise apartment. Results clearly suggest that elevated concentrations of *n*-alkanes and PAHs occurred in the samples from low floor levels (1$^{st}$ and 10$^{th}$ floor). Consequently, the cancer risk from exposure to dust bound PAHs would be predicted to be greater for people living on lower floor levels than on higher floor levels. Previous studies have demonstrated that the vertical variations of ambient air pollution are influenced by emission sources and meteorological conditions. Several investigators also found significant declining trends on aerosol associated PAHs concentrations with height in urban areas (Li *et al.*, 2005; Tao *et al.*, 2007). Thus our results clearly demonstrated that elevated pollution by particle-associated contaminants occurred on lower floor levels compared with high floor levels of high-rise buildings. As mentioned above, the height of a building can greatly influence wind speed and microenvironment; thus it may further affect the vertical diffusion of different sizes of aerosols and the exchange of particle bounded pollutants between indoor and outdoor air. However, knowledge on these aspects remains limited. Additional efforts should also be taken to explore the change of contaminants in particles with different sizes between indoor and outdoor environment at different vertical heights.

**CONFLICTS OF INTEREST**

The authors declare no competing financial interest.

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