

CHARACTERISTICS OF POLYCYCLIC AROMATIC HYDROCARBON FORMATION DURING SMOKING

*Haruki Shimazu¹, Tsuyoshi Yata¹ and Naoto Ozaki¹

¹School of Science & Engineering, Kindai University, Japan

*Corresponding Author, Received: 11 Nov. 2017, Revised: 8 Dec. 2017, Accepted: 25 Dec. 2017

ABSTRACT: This study examines the concentrations of polycyclic aromatic hydrocarbons (PAHs) in cigarettes and sidestream cigarette smoke. All 16 gas-phase PAHs and fourteen particulate-phase PAHs were determined in sidestream cigarette smoke for four brands of cigarettes. The median total of gas-phase and particulate-phase PAH concentrations after two cigarettes were smoked were 79.6 ng/m³ and 10.0 ng/m³, and the concentrations were several times higher than before smoking. Median PAH increases in the air samples were 1050 ng per cigarette for gas-phase PAHs and 228 ng per cigarette for particulate-phase PAHs. Gas-particle partition coefficients (K_p) of PAHs in air samples before and after smoking were correlated with the subcooled liquid vapor pressures (PL^o) of PAHs. The slopes of log-log plots of K_p versus PL^o before and after smoking were -0.270 and -0.575, respectively. PAH partitioning between the gas and particle phases after smoking was closer to equilibrium state because the slope was closer to -1. Furthermore, all 16 PAHs were determined in the cigarettes. The medians of total PAH concentrations in the four brands of cigarettes were 353 ng/g before smoking and 1130 ng/g after smoking. Median PAH increase in the cigarette samples during smoking was 1410 ng per cigarette. The relationships in the concentration of the between total suspended particulate (TSP) and total particulate-phase PAHs, and between total gas-phase PAHs and total particulate-phase PAHs were significantly positive. These findings imply that lowering the amount of TSP generated will lower the amounts of PAHs generated.

Keywords: Cigarette, Sidestream cigarette smoke, Polycyclic aromatic hydrocarbons, Gas-particulate partition coefficient, Total suspended particulate

1. INTRODUCTION

Certain organic compounds in cigarette smoke can cause respiratory diseases and lung cancers [1]–[3]. Among these constituents are polycyclic aromatic hydrocarbons (PAHs), which are produced by incomplete combustion of fossil fuels and other organic materials in vehicular emissions, incinerators, and factories [4]–[8]. PAHs are a group of chemicals composed of two or more fused benzenoid rings and known for their carcinogenic and mutagenic properties. The International Agency for Research on Cancer (IARC) has classified the PAH benzo[a]pyrene (BaP) as a Group 1 carcinogen (carcinogenic to humans) [9]. Dibenz[a,h]anthracene (DahA), dibenz[a,j]acridine, and dibenzo[a,l]pyrene have been classified as Group 2A carcinogens (probably carcinogenic to humans). Many PAHs, including BaP and benzo[b]fluoranthene (BbF), are also considered mutagenic [10].

Some studies on the toxicity of PAHs in mainstream cigarette smoke have been conducted to evaluate their health effects on active smokers [11]–[14]. However, little information is available on PAHs in sidestream cigarette smoke, which mainly affects passive smokers. The health effects of PAHs in cigarette smoke depend on the distribution of

PAHs between the gas and particulate phases. The first objective of this study was to examine the occurrence of PAHs in gas and particulate phases and their gas/particulate partitioning in sidestream cigarette smoke.

In some experimental studies, PAHs were found in the cigarette body [15], [16]. PAHs have been detected in some smokeless cigarette containing fire-cured cigarette varieties [17], [18]. This is because when cigarette is fire-cured, PAHs formed by smoldering wood are absorbed in the cigarette leaves [15]. The second objective of this study was to examine the occurrence of PAHs in cigarette and cigarette butt samples. Last, the formation characteristics of PAHs during smoking were investigated from the PAH mass balance in cigarettes and cigarette smoke.

2. MATERIALS AND METHODS

2.1 Samples

Four brands of cigarettes were used in this study. The tar and nicotine content was 6–19 mg/cigarette and 0.5–1.4 mg/cigarette, respectively. PAH concentrations in air samples collected before and after cigarettes had been smoked were measured.

Four air samples were collected before and after

cigarettes of brands A-D had been smoked. The volume of the air sampling room was approximately 66 m³, with 2.5 m high, 5.3 m long, and 5.0 m wide. Each pre-smoking air sample was collected using a high-volume air sampler (HV-500R; Sibata Scientific Technology Ltd., Souka, Japan) over a period of 1 h at a flow rate of 400 L/min (giving a sampling volume of 24 m³). Particulate matter was collected on a quartz fiber filter (QR-100; Advantec, Tokyo, Japan). The filter had a minimum particle collection efficiency of 99.99% for particles with diameters of 0.3 µm when the air was passed through the sampler at a velocity of 5 cm/s. Gas-phase PAHs were collected on two polyurethane foam plugs (080130-0941A; Sibata Scientific Technology Ltd.). After the initial air samples had been collected, the door and windows of the sampling room were left open for about 1 h. The door and windows were then closed, and two cigarettes were smoked in the room. Sidestream smoke samples were collected using the high-volume air sampler over a period of 2.75 h at a flow rate of 400 L/min (giving a sampling volume of 66 m³). Particulate-phase PAHs were collected on a quartz fiber filter, and gas-phase PAHs were collected on two polyurethane foam plugs. The used filters and polyurethane foam plugs were analyzed as described in the analytical methods and instruments section below.

PAH levels in cigarettes before and after smoking were also determined. Cigarette samples (before smoking) and butt samples (after smoking) were analyzed. Table 1 lists the filter and leaf weights per cigarette. The arithmetic means of the weights of the four brands of cigarettes before smoking (n = 4) were 0.107–0.181 g for the filters and 0.525–0.586 g for the leaves. The weights of cigarettes after smoking (n = 4) were 0.158–0.285 g for the filters and 0.075–0.078 g for the leaves. Whole filters and leaves of cigarette samples were analyzed as reported in the analytical methods and instruments section below. After each of four brands of cigarettes were smoked, whole filters and leaves of the butt samples were also analyzed.

Table 1 Cigarette weights before and after smoking

	Cigarette		Cigarette butt	
	Filter	Leaf	Filter	Leaf
Brand A	0.170	0.525	0.272	0.077
Brand B	0.178	0.546	0.285	0.077
Brand C	0.181	0.586	0.267	0.075
Brand D	0.107	0.578	0.158	0.078

Note: The weights are the arithmetic means (n = 4). All units are g.

2.2 PAHs

The target compounds were the 16 PAHs listed

in Table 2. These PAHs were frequently detected in various environmental media in multiple studies. The US Environmental Protection Agency regulates the concentrations of these 16 PAHs in the air, water, and soil. Naphthalene (Nap) is a two-ring PAH. Acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), and anthracene (Ant) are three-ring PAHs. Fluoranthene (Flt), pyrene (Pyr), benz[a]anthracene (BaA), and chrysene (Chr) are four-ring PAHs. BbF, benzo[k]fluoranthene (BkF), BaP, and DahA are five-ring PAHs. Indeno[1,2,3-cd]pyrene (IP) and benzo[ghi]perylene (BP) are six-ring PAHs.

Table 2 PAHs measured in this study

PAHs	Abbr.	CAS No.
Naphthalene	Nap	91-20-3
Acenaphthylene	Acy	208-96-8
Acenaphthene	Ace	83-32-9
Fluorene	Flu	86-73-7
Phenanthrene	Phe	85-01-8
Anthracene	Ant	120-12-7
Fluoranthene	Flt	206-44-0
Pyrene	Pyr	129-00-0
Benzo[a]anthracene	BaA	56-55-3
Chrysene	Chr	218-01-9
Benzo[b]fluoranthene	BbF	205-99-2
Benzo[k]fluoranthene	BkF	207-08-9
Benzo[a]pyrene	BaP	50-32-8
Indeno[1,2,3-cd]pyrene	IP	193-39-5
Dibenz[a,h]anthracene	DahA	53-70-3
Benzo[ghi]perylene	BP	191-24-2

Note: The abbreviations are used in this study.

2.3 Analytical methods and instruments

Each quartz fiber filter was weighed using an electronic balance before and after smoking. Each filter that had been used to sample air after smoking was cut into 16 sections. All portions were placed in 260-mL bottles and extracted with 100 mL of hexane for 15 min in an ultrasonic bath. The extract was concentrated to 3 mL using a rotary evaporator, then passed through a disposable filter (Whatman Puradisc 25 TF; GE Healthcare Bio-Sciences, Piscataway, NJ, USA). The extract was then concentrated to 1.5 mL under N₂ flow. Finally, 200 µL of internal standard mixture and hexane were added to bring the final volume to 2 mL.

Two polyurethane foam plugs were used in each experiment. Each polyurethane foam plug that had been used to sample air after smoking was cut into 16 sections. All portions were placed in 260-mL bottles and extracted with 220 mL of hexane for 15 min in an ultrasonic bath. The separate extracts of the two foam plugs were each concentrated to 3 mL using a rotary evaporator and then mixed it. Next,

the extract was passed through a disposable filter and concentrated to 1.5 mL under N₂ flow. Finally, 200 µL of internal standard mixture and hexane were added to bring the final volume to 2 mL.

Cigarette filters and leaves before and after smoking were analyzed. Each sample was weighed, placed into a cellulose extraction thimble (Whatman high-performance cellulose extraction thimbles; GE Healthcare Life Sciences, Little Chalfont, UK), and extracted with 100 mL of hexane for 15 min in an ultrasonic bath. The extract was concentrated to 3 mL using a rotary evaporator, then passed through a disposable filter device. Next, the extract was concentrated to 1.5 mL under N₂ flow. Finally, 200 µL of internal standard mixture and hexane were added to bring the final volume to 2 mL.

PAH concentrations in the extracts were detected by a gas chromatography-mass spectrometer (5975B inert XL E/CI MSD; Agilent Technologies, Santa Clara, CA, USA) equipped with an HP-5MS capillary column (30-m long, 0.25-mm i.d., 0.25-µm film thickness; Agilent Technologies). The gas chromatograph was using in splitless injection mode, with a sample volume of 2 µL and an injection port temperature of 250°C. The oven temperature program commenced at 70°C (held for 1.5 min), and was then increased at 20°C/min to 180°C, followed by an increase of 5°C/min to 280°C (held for 1 min). The carrier gas was helium. The mass spectrometer was operated in electron impact ionization mode and an electron energy of 70 eV. Each analyte was identified using two representative fragment ions, then quantified using the most abundant fragment ion. Quantification was performed with an internal standard calibration method using standards containing the 16 PAHs shown in Table 1 (TCL PAH Mix, CRM48905 Supelco, Bellefonte, PA, USA) and three deuterated PAHs (525 Fortification Solution, 48230-U, Supelco).

Total suspended particulate (TSP) concentration (µg/m³) was defined as the difference between the weight of a quartz fiber filter before and after air sampling divided by 24 m³ for a sample collected before cigarettes had been smoked or 66 m³ for a sample collected after cigarettes had been smoked.

3. RESULTS AND DISCUSSION

3.1 PAHs in air samples before and after smoking

Fifteen PAHs were detected in the gas-phase air samples collected before cigarettes had been smoked. Concentrations ranged from 29.4 to 166 ng/m³ (median: 48.0 ng/m³). Median values were 90.9 ng/m³ for Brand A, 74.8 ng/m³ for Brand B, 45.9 ng/m³ for Brand C, and 52.3 ng/m³ for Brand

D (Figure 1). Nap, Acy, Flu, Phe, and Flt were detected in all 16 samples. Ace, Pyr, and BaA were detected in almost all of the samples. The mean mole ratios for the individual PAHs were 35.2% for Phe, 32.6% for Nap, 11.1% for Flu, 5.5% for Ace, 3.7% for Flt, 3.6% for Pyr, 3.5% for Acy, 3.1% for BaA, 1.4% for BkF, and 0.4% for Ant.

Fifteen PAHs were detected in the particulate-phase air samples collected before cigarettes had been smoked. Concentrations ranged from 1.35 to 13.2 ng/m³ (median: 3.13 ng/m³). Median values were 2.15 ng/m³ for Brand A, 3.01 ng/m³ for Brand B, 2.74 ng/m³ for Brand C, and 7.17 ng/m³ for Brand D (Figure 1). BkF was detected in all 16 samples, and Nap, Phe, Flt, Pyr, BaA, Chr, and BbF were detected in almost all of the samples. The mean mole ratios for the individual PAHs in the particulate-phase samples were 53.5% for Nap, 14.2% for BkF, 7.1% for BbF, 6.6% for BaP, 4.3% for Phe, 2.9% for Pyr, 2.5% for BaA and Chr, 2.4% for Flt, 2.2% for Flu, and 1.8% for Ant.

All 16 PAHs were detected in the gas-phase air samples collected after cigarettes had been smoked. Concentrations ranged from 35.5 to 128 ng/m³ (median: 79.6 ng/m³). Median values were 58.0 ng/m³ for Brand A, 103 ng/m³ for Brand B, 74.0 ng/m³ for Brand C, and 87.9 ng/m³ for Brand D (Figure 2). Nap, Acy, Flu, Phe, Ant, Flt, and Pyr were detected in all 16 samples. Ace, BaA, and Chr were detected in almost all of the samples.

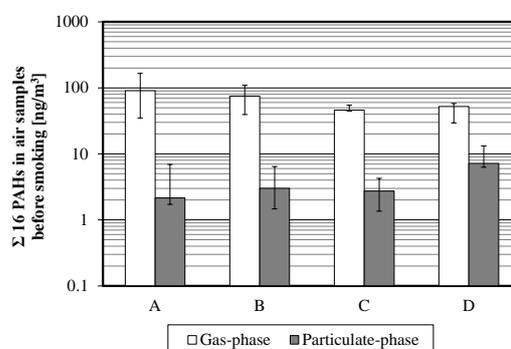


Fig.1 PAHs in air samples before smoking

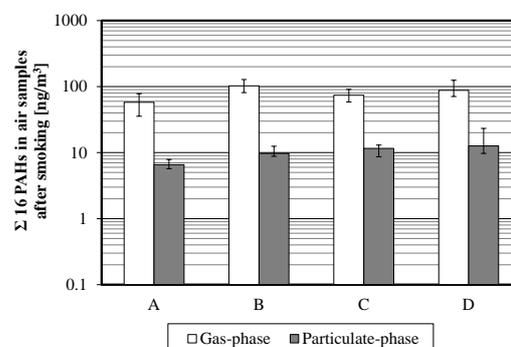


Fig.2 PAHs in air samples after smoking

The mean mole ratios for the individual PAHs were 35.7% for Phe, 15.0% for Flu, 14.4% for Nap, 9.4% for Acy, 7.0% for Flt, 5.9% for Ant, 5.3% for Pyr, 4.8% for Ace, 1.1% for Chr, 1.0% for BaA, and 0.3% for BkF.

Fourteen PAHs were detected in the particulate-phase air samples collected after cigarettes had been smoked. Concentrations ranged from 5.71 to 23.4 ng/m³ (median: 10.0 ng/m³). Median values were 6.58 ng/m³ for Brand A, 9.70 ng/m³ for Brand B, 11.6 ng/m³ for Brand C, and 12.6 ng/m³ for Brand D (Figure 2). Nap, Chr, BbF, BkF, Bap, IP, and BP were detected in all 16 samples. Flt, Pyr, and BaA were detected in almost all of the samples. The mean mole ratios for the individual PAHs were 20.1% for BaP, 16.7% for BbF, 16.2% for Chr, 12.9% for BP, 11.7% for IP, 8.7% for BaA, 5.4% for Nap, 4.5% for BkF, 1.8% for DahA, 0.8% for Ace, 0.7% for Flt, and 0.5% for Pyr. The gas-phase PAH concentrations were much higher than the particulate-phase PAH concentrations both before and after smoking. The medians of gas-phase and particulate-phase PAH concentrations were higher after smoking.

3.2 PAHs in cigarette samples before and after smoking

Fifteen PAHs were detected in the cigarette leaves before cigarettes had been smoked. Concentrations ranged from 136 to 504 ng/g (median: 291 ng/g). Median values were 339 ng/g for Brand A, 431 ng/g for Brand B, 164 ng/g for Brand C, and 284 ng/g for Brand D (Figure 3). Nap, Phe, Flt, Pyr, BaA, and Chr were detected in all 16 samples. Ace, Ant, and BbF were detected in almost all of the samples. The mean mole ratios for the individual PAHs were 25.9% for Nap, 21.5% for Phe, 17.4% for Pyr, 14.2% for Flt, 3.9% for BbF, 3.5% for BkF, 2.7% for Ace, 2.5% for BaP, 2.2% for Ant, 1.9% for Chr, 1.7% for BaA, 1.5% for BP, and 1.1% for Flu.

Fifteen PAHs were detected in the cigarette filters before cigarettes had been smoked. Concentrations ranged from 68.3 to 1010 ng/g (median: 330 ng/g). Median values were 517 ng/g for Brand A, 563 ng/g for Brand B, 110 ng/g for Brand C, and 287 ng/g for Brand D (Figure 3). Nap and Phe were detected in all 16 samples. Flt, Pyr, and BbF were detected in almost all of the samples. The mean mole ratios for the individual PAHs were 60.5% for Nap, 18.8% for BkF, 8.3% for Phe, 3.4% for Ace, 2.7% for Flu, 2.6% for Flt, 2.5% for Pyr, and 1.2% for BaA.

Sixteen PAHs were detected in the cigarette leaves after cigarettes had been smoked. Concentrations ranged from 890 to 2850 ng/g (median: 1390 ng/g). Median values were 1550 ng/g for Brand A, 1350 ng/g for Brand B, 1300 ng/g

for Brand C, and 2250 ng/g for Brand D (Figure 4). Phe, Flt, and Pyr were detected in all 16 samples. Ant, BaA, and Chr were detected in almost all of the samples. The mean mole ratios for the individual PAHs were 33.0% for Phe, 13.4% for Chr, 13.3% for Flt, 11.6% for Pyr, 8.8% for Ant, 7.0% for BaA, 4.1% for BaP, 3.6% for Nap, 3.2% for BbF, and 2.1% for Acy.

Thirteen PAHs were detected in the cigarette filters after cigarettes had been smoked. Concentrations ranged from 631 to 5020 ng/g (median: 957 ng/g). Median values were 1040 ng/g for Brand A, 895 ng/g for Brand B, 898 ng/g for Brand C, and 3710 ng/g for Brand D (Figure 4). Nap, Acy, Flu, Phe, Flt, and Pyr were detected in all 16 samples. BkF was detected in almost all of the samples. The mean mole ratios for the individual PAHs were 56.9% for Nap, 11.3% for Flu, 9.1% for Acy, 7.9% for Phe, 5.0% for Ace, 3.4% for Flt, 3.3% for Pyr, 1.9% for Ant, and 1.3% for BkF. PAH concentrations in cigarette leaves and filters were higher after smoking.

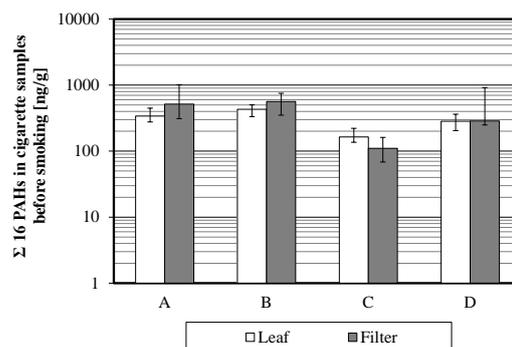


Fig.3 PAHs in cigarette samples before smoking

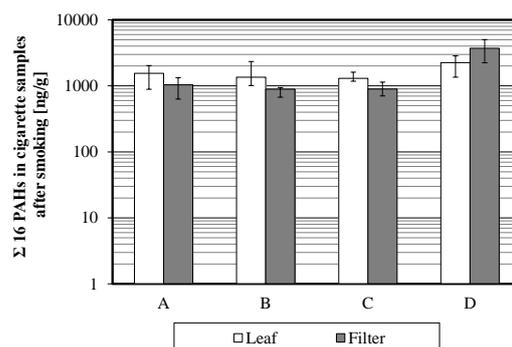


Fig.4 PAHs in cigarette samples after smoking

3.3 Gas-particle partitioning of PAHs

The fate of PAHs in the air is strongly affected by gas-particle partitioning, which is controlled by the vapor-liquid pressures of PAHs [19]-[21]. The gas-particle partitioning of PAHs in the air is presented using the gas-particle partition coefficient (K_p, m³/μg) calculated for PAHs determined simultaneously in the gas and particle phases.

$$K_p = (F / TSP) / A$$

Where F is the particle-phase concentration of PAHs (ng/m³), A is the gas-phase concentration (ng/m³), and TSP is the total suspended particulate concentration (µg/m³). Multiplying K_p by TSP gives a ratio of the particle- to gas-phase concentrations:

$$K_p \times TSP = F / A$$

PAHs partition primarily to the particle phase for $K_p \times TSP > 1$, and to the gas phase for $K_p \times TSP < 1$. For the PAHs detected in both the gas and particulate phases before cigarettes had been smoked ($n \geq 5$), the mean values of $K_p \times TSP$ for Nap, Ace, Flu, Phe, Ant, Flt, Pyr, and BaA were < 1 , and those for BbF and BkF were > 1 . For the PAHs detected in both the gas and particulate phases after cigarettes had been smoked ($n \geq 5$), the mean values of $K_p \times TSP$ for Nap, Ace, Phe, Flt, and Pyr were < 1 , and those for BaA, Chr, BbF, BkF, and IP were > 1 . High-molecular-weight PAHs that originate from combustion sources could be strongly absorbed to particles in the air. These findings indicate that the low-molecular-weight PAHs for $K_p \times TSP < 1$ tend to exist in the gas phase, and the high-molecular-weight PAHs for $K_p \times TSP > 1$ tend to exist in the particulate phase.

The mean values of $\log K_p$ for the PAHs detected in both the gas and particulate phases ($n \geq 5$) ranged from -4.15 (Phe) to -1.87 (BbF) before smoking and -4.67 (Phe) to -0.76 (BbF) after smoking. The values were in good agreement with those shown in previous studies of atmospheric PAHs [22]-[24].

The calculated partition coefficient K_p for PAHs was correlated with the subcooled liquid vapor pressures (PL°) of PAHs using the following equation:

$$\log K_p = mr \times \log PL^\circ + br$$

Where mr and br are the slope and the intercept of the linear regression, respectively. The subcooled liquid vapor pressures (PL° , Torr) were calculated using air temperature [25], [26]. The relationships between $\log K_p$ and $\log PL^\circ$ before and after smoking are shown in Figure 5 and Figure 6, respectively. To compare the findings with those of other studies [22]-[24], [27], the plots for Nap and high- $\log PL^\circ$ PAHs were excluded from the figures. Both correlation coefficients (r^2) were statistically significant ($p < 0.01$). The slope and intercept of \log - \log plots of K_p versus PL° before cigarettes had been smoked were -0.270 and -4.33 , respectively; the slope and intercept of the plots after cigarettes had been smoked were -0.575 and -5.87 , respectively. The slope mr should be equal to -1 under equilibrium conditions between the gas and particle phases [28]. PAH partitioning between the gas and particle phases was closer to equilibrium state after smoking.

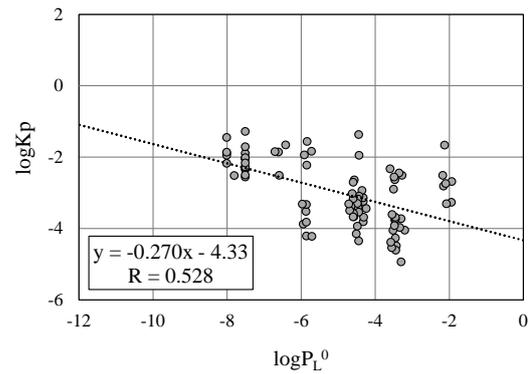


Fig.5 Relationships between $\log K_p$ and $\log PL^\circ$ before smoking

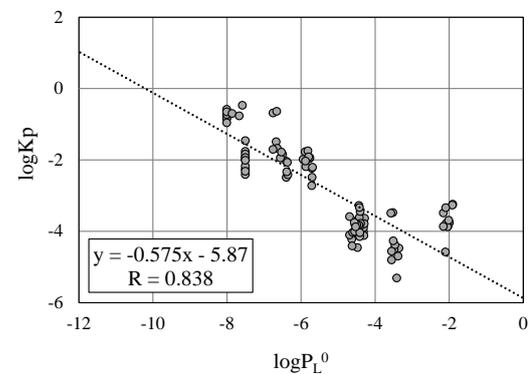


Fig.6 Relationships between $\log K_p$ and $\log PL^\circ$ after smoking

3.4 PAH formation while smoking

Figure 7 shows the median contents of the 16 PAHs in air and cigarettes in the air sampling room before and after smoking. Levels in air were calculated as the median concentrations for the 16 PAHs multiplied by 24 m³ for a sample collected before smoking or 66 m³ for a sample collected after smoking. PAH content in cigarette filters and leaves was calculated as the median concentration multiplied by weight times two, because two cigarettes of each brand were smoked during the experiments. PAH content in cigarettes was calculated as the sum of PAH content in cigarette filters and leaves. PAH levels in air before cigarettes had been smoked were 1930–10 900 ng for the gas-phase PAHs and 88.8–867 ng for the particulate-phase PAHs; the medians were 3150 and 205 ng, respectively. PAH levels in air after cigarettes had been smoked were 2340–8440 ng for the gas-phase PAHs and 376–1540 ng for the particulate-phase PAHs; the medians were 5240 and 660 ng, respectively. PAH content in two cigarettes before smoking was 188–803 ng; the median was 505 ng. PAH content in two cigarettes after smoking was 511–1730 ng; the median was 774 ng. PAH content in the cigarettes was approximately equal before and after smoking.

However, levels of gas-phase PAHs and particulate-phase PAHs were higher after smoking. Figure 8 displays the relationship between the gas-phase and particulate-phase PAH levels in the air after smoking; the correlation coefficient was 0.640 ($p < 0.001$). Gas-phase levels of all 16 PAHs increased as the total particulate-phase levels increased.

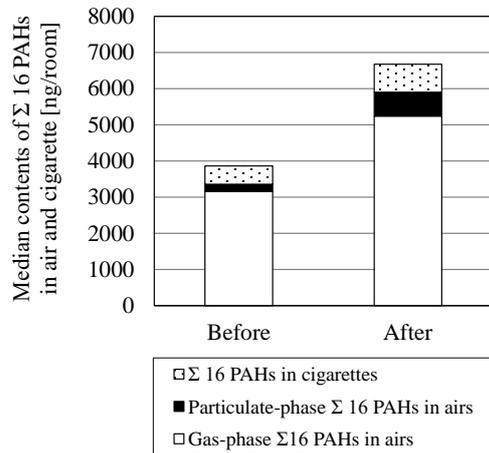


Fig.7 Relationships between logKp and log PL° after smoking

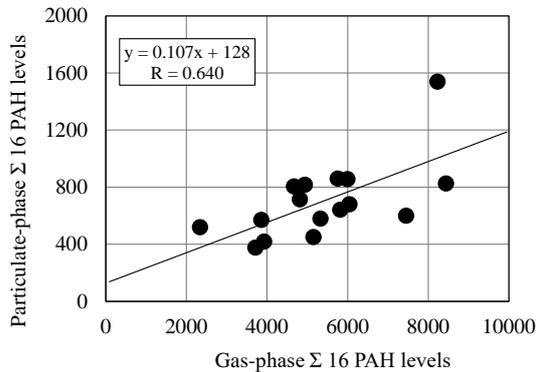


Fig.8 Relationship between gas-phase Σ 16 PAH levels and particulate-phase Σ 16 PAH levels after smoking

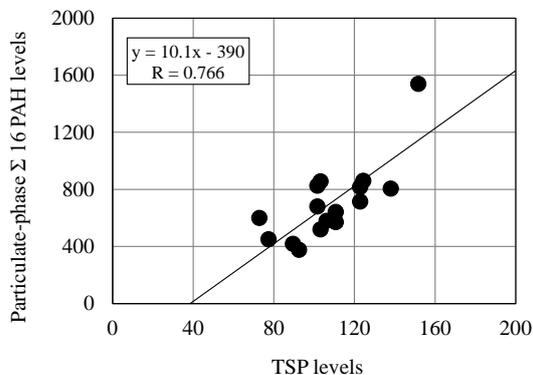


Fig.9 Relationships between TSP levels and particulate-phase Σ 16 PAH levels after smoking

Figure 9 displays the relationship between TSP levels and particulate-phase PAH levels in the air sampling room after smoking; the correlation coefficient was 0.766 ($p < 0.001$). These findings imply that lowering the amount of TSP generated will lower the amounts of PAHs generated.

4. CONCLUSION

In this study, the concentrations of PAHs in cigarettes and sidestream cigarette smoke for four brands of cigarettes were examined and the formation characteristics of PAHs during smoking were investigated from the PAH mass balance in cigarettes and the smoke. The results are summarized as follows.

(1) PAH levels in sidestream cigarette smoke before and after smoking were determined in the air sampling room with a volume of approximately 66 m³. The median total of gas-phase and particulate-phase PAH concentrations after two cigarettes were smoked were 79.6 ng/m³ and 10.0 ng/m³, and the concentrations were several times higher than before smoking. Median PAH increases in the air samples were 1050 ng per cigarette for gas-phase PAHs and 228 ng per cigarette for particulate-phase PAHs.

(2) Gas-particle partition coefficients (Kp) of PAHs in air samples before and after smoking were correlated with the subcooled liquid vapor pressures (PL°) of PAHs. The slopes of log-log plots of Kp versus PL° before and after smoking were -0.270 and -0.575, respectively. PAH partitioning between the gas and particle phases after smoking was closer to equilibrium state because the slope was closer to -1.

(3) PAH levels in cigarettes before and after smoking were determined. The medians of total PAH concentrations before smoking were 291 ng/g for cigarette leaves and 330 ng/g for cigarette filters. Those after smoking were 1390 ng/g for cigarette leaves and 957 ng/g for cigarette filters. The concentrations in cigarette leaves and filters were higher after smoking. Median PAH increase in the cigarette samples during smoking was 1410 ng per cigarette.

(4) The relationships in the levels for the between TSP and total particulate-phase PAHs, and between total gas-phase PAHs and total particulate-phase PAHs after cigarettes had been smoked were significantly positive. These findings imply that lowering the amount of TSP generated will lower the amounts of PAHs generated.

5. ACKNOWLEDGEMENTS

The research described in this paper was financially supported by Kindai University. The authors thank Dean Meyer, PhD, ELS, from Edanz Group (www.edanzediting.com/ac) for editing a draft of this manuscript.

6. REFERENCES

- [1] OEHHA, Proposed identification of environmental tobacco smoke as a toxic air contaminant, part B: Health effects assessment for environmental tobacco smoke, 2005.
- [2] International Agency for Research on Cancer, Tobacco smoke and involuntary smoking, Vol. 83 IARC monographs on the evaluation of the carcinogenic risk of chemicals to human, 2004.
- [3] Lee, C.H., Ko, Y.C., Cheng, L.S., Lin, Y.C., Lin, H.J., Huang, M.S., Huang, J.J., Kao, E.L., and Wang, H.Z., The heterogeneity in risk factors of lung cancer and the difference of histologic distribution between genders in Taiwan, *Cancer Causes and Control*, Vol.12, 2001, pp.289–300.
- [4] Yang, H.H., Hsieh, L.T., Liu, H.C., and Mi, H.H., Polycyclic aromatic hydrocarbon emissions from motorcycles, *Atmospheric Environment*, Vol.39, 2005, pp.17-25.
- [5] Shen, H., Tao, S., Wang, R., Wang, B., Shen, G., Li, W., Su, S., Huang, Y., Wang, X., Liu, W., Li, B., and Sun, K., Global time trends in PAH emissions from motor vehicles, *Atmospheric Environment*, Vol.45, 2011, pp.2067-2073.
- [6] Besombes, J.L., Maitre, A., Patissier, O., Marchand, N., Chevron, N., Stoklov, M., and Masclat, P., Particulate PAHs observed in the surrounding of a municipal incinerator, *Atmospheric Environment*, Vol.35, 2001, pp.6093-6104.
- [7] Sadhra, S. and Wheatley, A.D., Polycyclic aromatic hydrocarbons emissions from clinical waste incineration, *Chemosphere*, Vol.66, 2007, pp.2177-2184.
- [8] Wan, X., Chen, J., Tian, F., Sun, W., Yang, F., and Kaoru Saiki, Source apportionment of PAHs in atmospheric particulates of Dalian: Factor analysis with nonnegative constraints and emission inventory analysis, *Atmospheric Environment*, Vol.40, 2006, pp.6666-6675.
- [9] IARC, Some Industrial Chemicals, Vol. 115. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, 2016.
- [10] Luch, A., *The Carcinogenic Effects of Polycyclic Aromatic Hydrocarbons*. London: Imperial College Press, 2005.
- [11] Risner, C.H., The determination of benzo[a]pyrene in the total particulate matter of cigarette smoke, *Journal of Chromatographic Science*, Vol.26, 1988, pp.113–120.
- [12] Ding, Y.S., Ashley, D.L., and Watson, C.H., Determination of 10 carcinogenic polycyclic aromatic hydrocarbons in mainstream cigarette smoke, *Journal of Agricultural and Food Chemistry*, Vol.55, 2007, pp.5966-5973.
- [13] Ding, Y. S., Zhang, L., Jain, R. B., Jain, N., Wang, R. Y., Ashley, D. L., and Watson, C.H., Levels of tobacco-specific nitrosamines and polycyclic aromatic hydrocarbons in mainstream smoke from different tobacco varieties, *Cancer Epidemiology, Biomarkers and Prevention*, Vol.17, 2008, pp.3366–3371.
- [14] Vu, A.T., Taylor, K.M., Holman, M.R., Ding, Y.S., Hearn, B., and Watson, C.H., Polycyclic aromatic hydrocarbons in the mainstream smoke of popular U.S. cigarettes. *Chemical Research in Toxicology*, Vol.28, 2015, pp.1616–1626.
- [15] Bentley, H.R., and Burgan, J.G., Polynuclear hydrocarbons in tobacco and tobacco smoke Part 1 3:4-Benzopyrene, *Analyst*, Vol.85, 1960, pp.723–727.
- [16] Shimazu H., Determination of polycyclic aromatic hydrocarbons in cigarettes and cigarette smoke, *Environment and Pollution*, Vol.5, 2016, pp.15-25.
- [17] Stepanov, I., Villalta, P.W., Knezevich, A., Jensen, J., Hatsukami, D., and Hecht, S.S., Analysis of 23 polycyclic aromatic hydrocarbons in smokeless tobacco by gas chromatography-mass spectrometry, *Chemical Research in Toxicology*, Vol.23, 2010, pp.66–73.
- [18] McAdam, K.G., Faizi, A., Kimpton, H., Porter, A., and Rodu, B., Polycyclic aromatic hydrocarbons in US and Swedish smokeless tobacco products, *Chemistry Central Journal*, Vol.7, 2013, p.151.
- [19] Yamasaki, H., Kuwata, K. and Miyamoto, H., Effects of ambient temperature on aspects of airborne polycyclic aromatic hydrocarbons, *Environmental Science & Technology*, Vol.16, 1982, pp.189–194.
- [20] Pankow, J.F., Review and comparative analysis of the theories on partitioning between the gas and aerosol particulate phases in the atmosphere, *Atmospheric Environment*, Vol.21, 1987, pp.2275–2283.
- [21] Lohmann, R., Harner, T., Thomas, G.O. and Jones, K.C., A comparative study of the gas-particle partitioning of PCDD/Fs, PCBs, and PAHs, *Environmental Science & Technology*, Vol.34, 2000, pp.4943–4951.
- [22] Liang, C. and Pankow, J.F., Gas/particle

- partitioning of organic compounds to environmental tobacco smoke: partition coefficient measurements by desorption and comparison to urban particulate material, *Environmental Science & Technology*, Vol.30, 1996, pp.2800–2805.
- [23] Tsapakis, M. and Stephanou, E.G., Polycyclic aromatic hydrocarbons in the atmosphere of the Eastern Mediterranean, *Environmental Science & Technology*, Vol.39, 2005, pp.6584-6590.
- [24] Akyüz, M. and Çabuk, H., Gas–particle partitioning and seasonal variation of polycyclic aromatic hydrocarbons in the atmosphere of Zonguldak, Turkey, *Science of the Total Environment*, Vol.408, 2010, pp.5550-5558.
- [25] Paasivirta, J., Sinkkonen, S., Mikkelsen, P., Rantio, T. and Wania, F., Estimation of vapor pressures, solubilities and Henry's law constants of selected persistent organic pollutants as functions of temperature, *Chemosphere*, Vol.39, 1999, pp.811-832.
- [26] Allen, J.O., Sarofim, A.F. and Smith, K.A., Thermodynamic properties of polycyclic aromatic hydrocarbons in the subcooled liquid state, *Polycyclic Aromatic Compounds*, Vol.13, 1999, pp.261-283.
- [27] Cincinelli, A., Bubba, M.D., Martellini, T., Gambaro, A. and Lepri, L., Gas-particle concentration and distribution of n-alkanes and polycyclic aromatic hydrocarbons in the atmosphere of Prato (Italy), *Chemosphere*, Vol.68, 2007, pp.472-478.
- [28] Pankow, J.F., Bidleman, T.F., Interdependence of the slopes and intercepts from log-log correlations of measured gas-particle partitioning and vapor pressure. I. Theory and analysis of available data, *Atmospheric Environment*, Vol.26, 1992, pp.1071–1080.

Copyright © Int. J. of GEOMATE. All rights reserved, including the making of copies unless permission is obtained from the copyright proprietors.
