Size Distributions of Particulate Matter and Particle-bound Polycyclic Aromatic Hydrocarbons and Their Risk Assessments during Cable Sheath Burning

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Abstract

Burning of electric cable sheath leads to the emission of small particles and toxic pollutants that cause severe air pollution and human health effects. In this study, size distributions of particulate matter and p-PAHs during open burning of cable insulation were examined. Lifetime cancer risk of PAHs was also assessed. The particulate samples were collected on quartz fiber filters using an eight-stage cascade impactor with flow rate of 28.3 L min^{-1}. The exposed filter was extracted with acetonitrile and then measured by GC-MS in the SIM mode for 16 PAHs identification. It was found that average concentrations of ultrafine, fine and coarse particles were 1,045.82 µg m^{-3} (11.45 % of the total mass), 3,557.50 µg m^{-3} (38.96 % of the total mass) and 4,529.03 µg m^{-3} (49.59 % of the total mass), respectively. The particle size distributions were bimodal with one major peak in the size range of 5.8-4.7 µm and another minor peak in the size range of 1.1-0.65 µm. The concentrations of 16 PAHs adsorbed on ultrafine, fine and coarse particle were 717.86 ng m^{-3} (11.47% of the total PAHs), 3,645.43 ng m^{-3} (58.23% of the total PAHs) and 1,897.19 ng m^{-3} (30.30% of the total PAHs), respectively. Distributions of BaA, BaP, DbA and BgP were unimodal with a peak in accumulation mode while those of Acy, Ace and Fla were bimodal with two peaks in accumulation mode. Distributions of Flu, Phe, Ant, Pyr, Chr, BbF and BkF were multimodal with peaks in accumulation and coarse modes whereas InP was not detected. The inhalable particles (PM$_{10}$) contained mainly 5-ring PAHs (55.67% of total PAHs) followed by 4-ring PAHs (27.58% of total PAHs), and 3-ring PAHs (14.98% of total PAHs). Only small amount of 2-ring PAHs (1.36% of total PAHs) and 6-ring PAHs (0.41% of total PAHs) was observed. The fraction of PAHs adsorbed on PM$_{10}$ was ranked in the order Group 2B (53.45%) > Group 3 (33.86%) > Group 1 (10.71%) > Group 2A (1.98%). The average concentrations of 16 PAHs and B[a]P$_{eq}$ were 6,260.47 ng m^{-3} and 1,014.35 ng m^{-3}, respectively. The estimated lifetime lung cancer risk during wire burning was 8.83E-02.

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1. Introduction

Electric cable wire is an essential part of all electrical and electronic devices. Nowadays, large quantities of electronic waste (e-waste) such as discarded computers, televisions, refrigerators are produced worldwide due to high consumption and shorter life span of electronic products [1]. This leads subsequently to massive generation of used cable wire. As it is composed of copper wire, which is high economic value, recycling of used cable is increasingly interested. There are several techniques used to recover copper from cable wire such as stripping, crushing, burning, high pressure water jet, cryogenic process and chemical treatment [2]. However, open burning is a common technique for cable wire recycling in informal sector, where most of e-waste is managed unsafely. As cable sheath consists of polyvinyl chloride (PVC), polyethylene (PE), silicone rubber and flame retardant [3], burning of the plastic insulation releases a large amount of small particles and particle-bound toxic elements such as heavy metals, polychlorinated biphenyls and polycyclic aromatic hydrocarbons (PAHs) into the surroundings [4, 5].

Atmospheric particles comprise a wide range of particle sizes. They can be classified based on their origin into primary and secondary particles. The former is directly released from an emission source while the latter is formed from chemical or photochemical reactions of pollutants both in gas and particle phases [6, 7]. The compositions and sizes of individual particles are changed during distribution in the atmosphere, relying on formation and destruction processes. Small particles can remain suspended for long time and be dispersed for long distance in the air. Thus, size distribution of particles and their compositions are able to indicate their emission sources and fate of transport in the environment [6]. Particulate matters, especially less than 2.5 micrometers in diameter, are correlated with the increased respiratory and cardiovascular diseases because they can penetrate into the respiratory tract, and some may transfer to bloodstream [8]. Several studies also indicated that smaller particle sizes potentially caused more health risk [9].

Burning of cable sheath is one of the main sources of PAHs derived from informal recycling of e-waste [5, 10]. Generally, PAHs in the atmosphere are found in vapor phase (g-PAHs) and particle phase (p-PAHs) [11]. The partitioning of a given PAH compound between particle and vapor phases depends on its vapor pressure, particle size and composition as well as ambient temperature [12, 13]. Allen $et\ al$. [14] reported that size distribution of atmospheric particle-bound PAHs related to vaporization and condensation mechanisms. In the atmosphere, low-molecular weight PAHs (2-3 rings; MW $\leq$ 178) are mainly found in a vapor phase (g-PAHs) while high molecular weight species (MW $\geq$ 228) are largely adsorbed on particles (p-PAHs) [15, 16]. Harrison $et\ al$. [17] also proposed that most PAHs, especially those with more than four aromatic rings, easily adsorb onto combustion aerosols. Specific compounds of PAHs are potentially toxic or mutagenic to humans that are associated with the increased risk of developing lung cancer [18, 19]. Previous studies revealed that high molecular weight PAHs were potentially more carcinogenic than low molecular weight ones [20]. It was found that carcinogenic PAHs were likely attached on fine particles [21-23]. In addition, p-PAHs remain in the surroundings for long period because they cannot be easily removed from the atmosphere by photolysis and other degradation pathways [24]. Since inhalation deposition of airborne pollutants depends on particle sizes, fine particle-bound PAHs can travel deep into the human respiratory system and cause adverse health impact [25, 26]. However, the effects on human health of PAHs depend on their toxicity, concentration, exposure time and exposure routes. The available information on size distribution of particle bound-PAHs (p-PAHs) released from open burning of cable sheath is still limited. This study aimed to investigate size distributions of particles and p-PAHs in different particle size ranges emitted from open burning of electric cable sheath. Risk assessment of PAHs via inhalation exposure was also determined using lifetime cancer risk of PAHs.

2.   Materials and Methods

2.1 Air sampling

Size distributions of particulate matter and p-PAHs adsorbed on inhalable particles emitted from burning of electric cable sheath were examined in a laboratory scale at Faculty of Science, King Mongkut's Institute of Technology Ladkrabang. One kg of cable sheath was burnt in open air. The air sample was collected on quartz fiber filters (Toyo Roshi Kaisha, Japan) using an eight-stage cascade impactor (Thermo Andersen, USA). The air sample was taken at a flow rate of 28.3 L min^{-1} for 30 min. The sampler was placed at 1.5 m above ground level in the downwind direction (1.5 meters). The particle sizes were separated into nine interval fractions: 10.0-9.0, 9.0-5.8, 5.8-4.7, 4.7-3.3, 3.3-2.1, 2.1-1.1, 1.1-0.65, 0.65-0.43, and <0.43 μm. The sampling procedure was conducted following US EPA method [27]. The gravimetric measurement was run in triplicate. Details of the analysis are explained in Phoothiwut and Junyapoon [28].

2.2 PAHs analysis

In this study, 16 PAHs consisting of naphthalene (Nap, 2-ring) acenaphthylene (Acy, 3-ring), acenaphthene (Ace, 3-ring), fluorene (Flu, 3-ring) phenanthrene (Phe, 3-ring), anthracene (Ant, 3-ring), fluoranthene (Fla, 4-ring), pyrene (Pyr, 4-ring), benzo[*a*]anthracene (BaA, 4-ring), chrysene (Chr, 4-ring), benzo[*b*]fluoranthene (BbF, 5-ring), benzo[*k*]fluoranthene (BkF, 5-ring), benzo[*a*]pyrene (BaP, 5-ring), dibenzo[*a,h*]anthracene (DbA, 5-ring), indeno[1,2,3-*cd*]pyrene (InP, 6-ring) and benzo[*ghi*]perylene (BgP, 6-ring) adsorbed on particles were extracted and analyzed using the Compendium Method TO-13A, modification from US EPA [29]. Each sample filter was cut into small pieces and then sonicated with 15 ml acetonitrile (99.9% HPLC grade, Sigma-Aldrich, USA) for 30 min. The extracts were analyzed by GC-MS in SIM mode (Agilent Technologies, USA) with an HP-5MS GC fused-silica capillary column, 30 m × 0.25 mm i.d. × 0.25 μm film thickness (J&W Scientific, USA). The carrier gas used was helium (99.999% purity, Praxair, Thailand) at a flow rate of 1.0 ml min^{-1}. Field blank sample was also measured using the same method as the sample. Recovery efficiencies were examined in triplicate using spike method. Details of the PAHs analysis are described in Phoothiwut and Junyapoon [28].

2.3 Calculation of lifetime lung cancer risk of PAHs

Lifetime lung cancer risk of PAHs was presented as excess cancer risk (ECR) from PAHs associated with particles through inhalation pathway. A mixture of PAHs concentrations was converted to benzo(a)pyrene toxic equivalent (B[a]P$_{eq}$). Total B[a]P$_{eq}$ can be calculated from equation 1.

$$\sum B[a]P_{eq} = \sum_{i=1}^{n}(C_i \times TEF_i) \tag{1}$$

Where C_i was the concentration of individual PAH congener attached on particles, TEF_i was the toxic equivalence factor of individual PAHs. TEF_i value of Nap, Acy, Ace used the factors provided by Nisbet and LaGoy [30] and the other PAHs used the values proposed by Larsen and Larsen [31]. ECR was calculated following the Office of Environmental Health Hazard Assessment (OEHHA) of the California Environmental Protection Agency (Cal EPA) [32, 33] as shown in equation 2.

$$ECR = \sum B[a]P_{eq} \times UR_{B[a]P} \tag{2}$$

Where $UR_{B[a]P}$ (unit risk) of 8.7×10^{-5} was the estimate number of people for lifetime cancer risks from inhalation at a B[a]P equivalent concentration of 1 ng m^{-3} within a lifetime of 70 years [34, 35].

3. Results and Discussion

3.1 Concentrations and size distributions of particulate matter

In this study, nine inhalable fractions of particles were divided into three size ranges according to their formation mechanisms: ultrafine (nuclei mode, $d_{ae}<0.4$ μm), fine (accumulation mode, $0.4<d_{ae}<2.1$ μm) and coarse particles ($d_{ae}>2.1$ μm). The concentrations of ultrafine, fine and coarse particles in ambient air during burning of cable sheath were in the range of 364.73-1,694.72 μg m^{-3} with an average of 1,045.82±665.58 μg m^{-3}, 1,953.33-4,431.41 μg m^{-3} with an average of 3,557.50±1,391.11 μg m^{-3} and 1,385.97-6,126.13 μg m^{-3} with an average of 4,529.03±2,722.09 μg m^{-3}, respectively as shown in Figure 1(a). The wide concentration ranges of particles were observed. This may be due to the variation
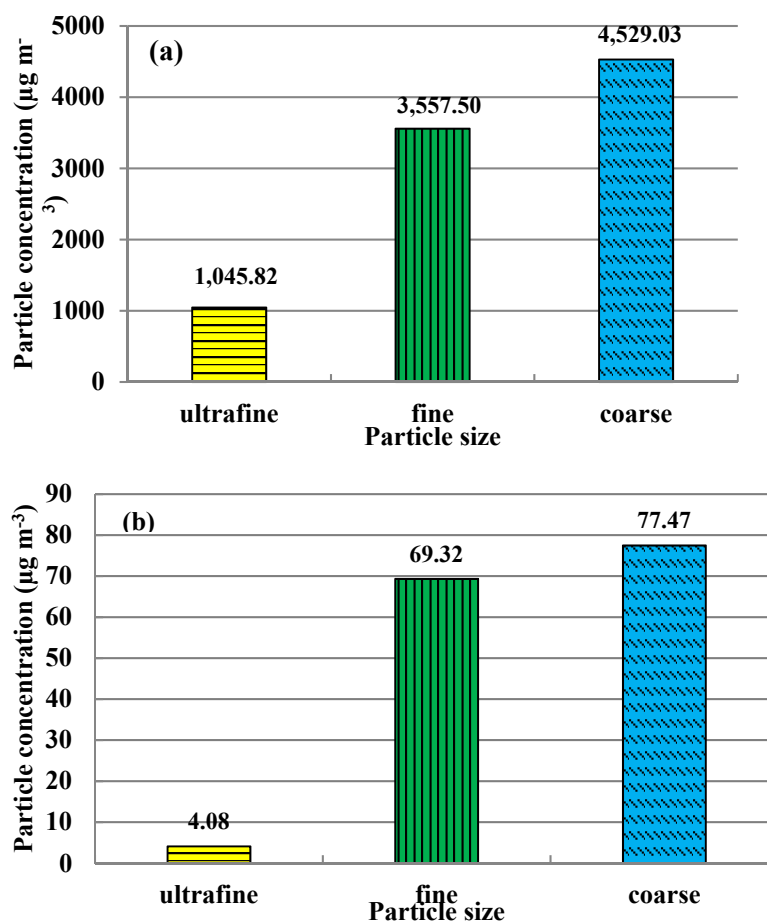

Figure 1. Concentrations of particle size range during open burning of cable sheath (a) and during no burning activity (b)

of burning and atmospheric conditions. The concentration range of each particle size during burning of cable sheath were significantly higher than those during no burning activity (Figure 1(b)). These results reveal that two main atmospheric components were coarse (49.59 % of the total mass) and fine particles (38.96 % of the total mass), and the minor component was ultrafine particles (11.45 % of the total mass). There was no mean difference between concentrations of coarse and fine particles (p-value>0.05). The size distribution plot of atmospheric particles during burning of cable sheath was bimodal with one major peak in the coarse particle size range (5.8-4.7 μm) and another minor peak in the fine particle size range (1.1-0.65 μm) as shown in Figure 2(a) while trimodal distribution was found during no burning activity (Figure 2(b)). Coarse particles are likely originated from incomplete combustion of plastic sheath whereas fine particles may be formed through different atmospheric formation mechanisms such as condensation of vapor species and accumulation of ultrafine particles [6].
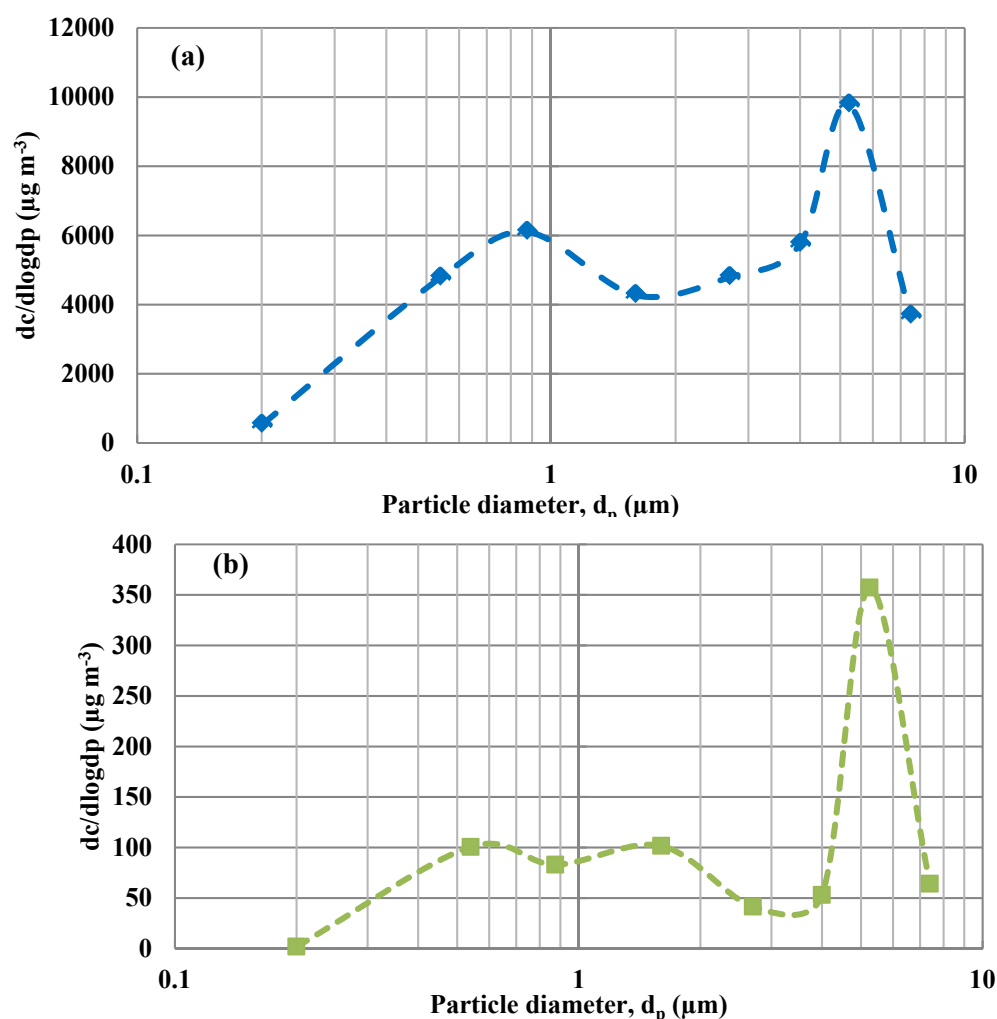

Figure 2. Size distributions of atmospheric aerosols during open burning of cable sheath (a) and during no burning activity (b)

3.2 Concentrations and size distributions of p-PAHs

Concentrations of 16 PAHs adsorbed on each particle size range during open burning of cable sheath are presented in Table 1. In this work, recovery efficiencies of 16 PAHs (n = 3) were ranged between 94.73 and 98.06 %. PAHs were likely generated from thermal degradation of PVC sheath through cracking and cross-linking processes of polymers [10]. The concentrations of total 16 PAH adsorbed on ultrafine, fine and coarse particles during burning of cable sheath were 717.86, 3,645.43 and 1,897.19 ng m^{-3}, respectively as shown in Figure 3. Total 16 PAHs were dominantly bound on fine particles (58.23% of the total PAHs) followed by coarse particles (30.30% of the total PAHs) and ultrafine particles (11.47% of the total PAHs). This is likely because p-PAHs in the accumulation mode were formed from condensation of vapor-phase PAHs as well as coagulation of their primary components. These results are similar to the findings of previous studies reporting that carcinogenic PAHs were likely attached on fine particles [21-23].

In this work, 16 PAHs attached on particles were divided into four groups, depending on the numbers of aromatic rings in their structures: 3-ring PAHs (Acy, Ace, Flu, Phe, Ant), 4-ring PAHs (Fla, Pyr, BaA, Chr), 5-ring PAHs (BbF, BkF, BaP, DbA) and 6-ring PAHs (InP, BgP). For 3-ring PAHs, distributions of Acy and Ace were bimodal with two peaks in accumulation mode whereas those of Flu, Phe and Ant were multimodal with peaks in accumulation and coarse modes as shown in Figure 4(a). For 4-ring PAHs, BaA had a unimodal distribution with a main peak in accumulation mode while Fla had a bimodal distribution with two peaks in accumulation mode. Multimodal distributions with peaks in accumulation and coarse modes were found in Pyr and Chr as shown in Figure 4(b). For 5-ring PAHs, distributions of BaP and DbA were unimodal with a main peak in accumulation mode while those of BbF and BkF were multimodal with peaks in accumulation and coarse modes as shown in Figure 4(c). For 6-ring PAHs, BgP had a unimodal distribution with a dominant peak in accumulation mode while InP was not detected (Figure 4(d)). These results indicate that most of less volatile PAHs preferentially condensed on fine particles, which is likely correlated with the previous work [14, 20]. However, burning parameters such as heat release rate, temperature, char length, afterburn time are required to be identified in future work.
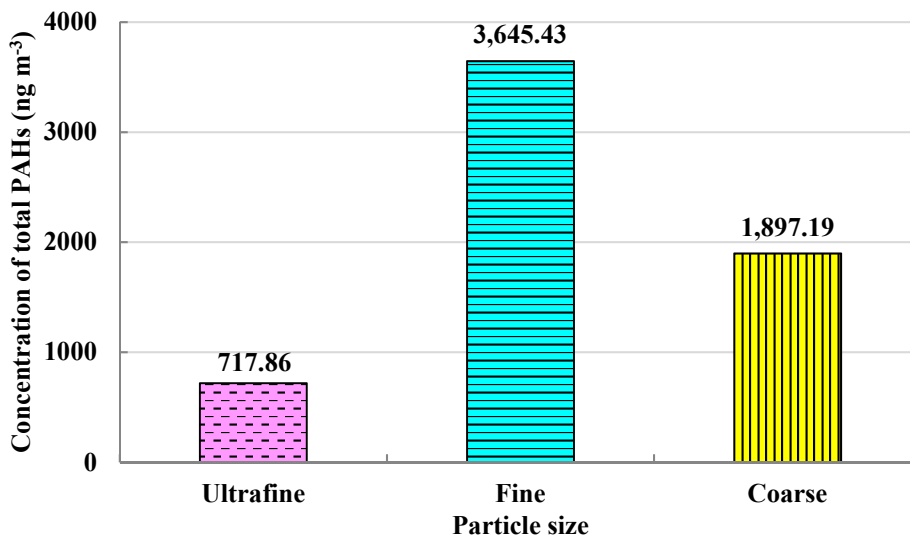

Figure 3. Concentrations of total 16 PAHs adsorbed on ultrafine, fine and coarse particles

Table 1. Concentrations of 16 PAHs adsorbed on each particle size range during open burning of cable sheath

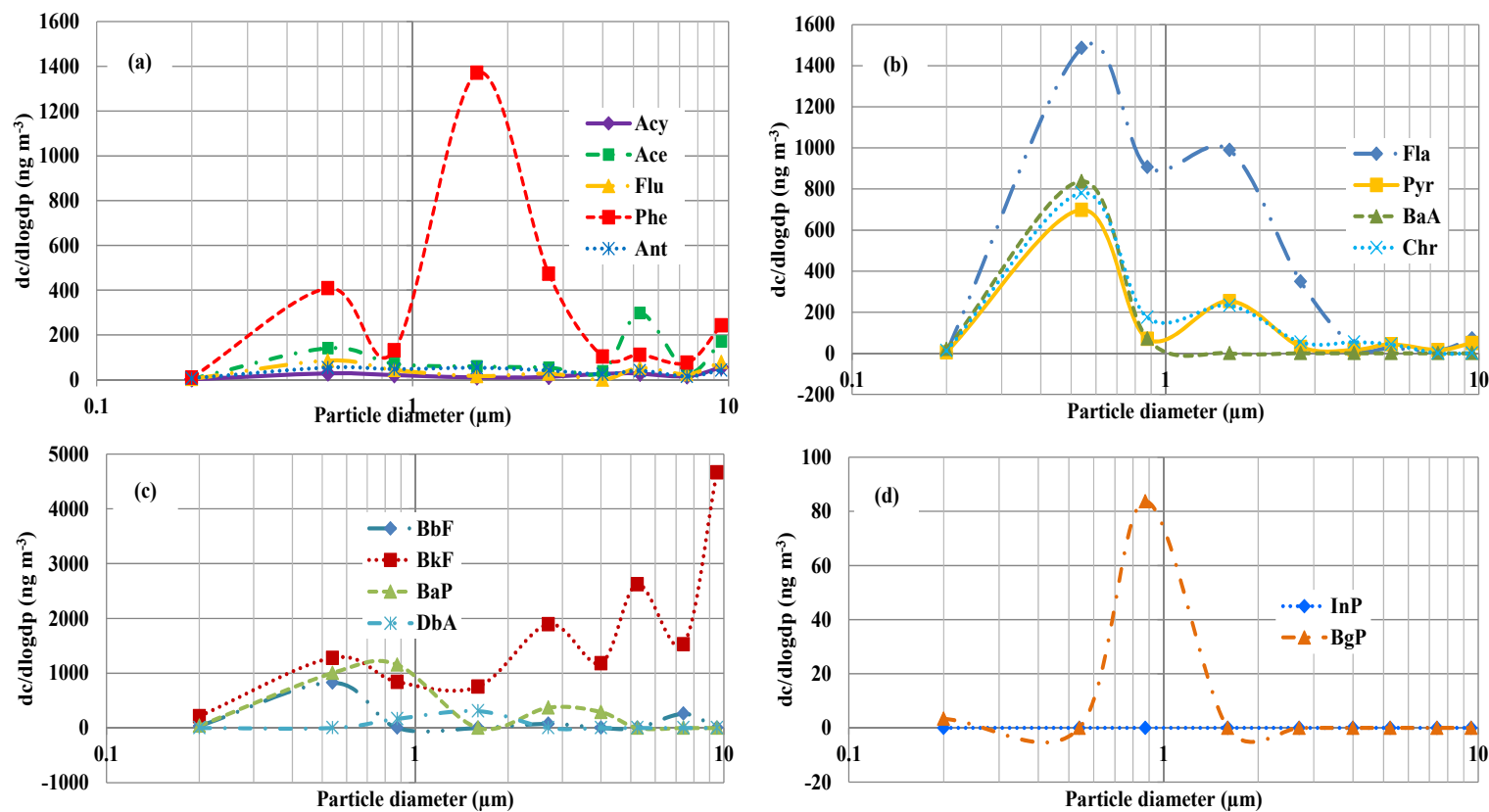| Particle size (µm)	PAHs concentration (ng m^{-3})															
	Nap	Acy	Ace	Flu	Phe	Ant	Fla	Pyr	BaA	Chr	BbF	BkF	BaP	InP	DbA	BgP
<0.4	6.98	2.68	6.71	5.47	17.51	11.82	8.97	8.71	38.98	23.65	71.06	430.69	77.90	n.d.	n.d.	6.73
0.43-0.65	19.57	5.92	29.59	17.93	86.09	11.36	313.24	147.22	176.58	164.54	174.17	270.02	211.42	n.d.	n.d.	n.d.
0.65-1.1	7.33	4.54	16.13	9.76	29.91	10.70	207.03	16.06	16.30	39.89	n.d.	192.53	263.70	n.d.	37.94	19.13
1.1-2.1	11.22	2.68	16.41	4.47	385.14	14.82	277.64	71.40	n.d.	65.30	n.d.	211.58	n.d.	n.d.	86.17	n.d.
2.1-3.3	8.60	2.43	10.44	5.21	92.74	7.92	68.57	6.13	n.d.	10.59	14.91	371.76	72.63	n.d.	n.d.	n.d.
3.3-4.7	9.94	3.97	5.61	n.d.	15.73	3.62	5.18	2.55	n.d.	8.33	n.d.	181.50	44.59	n.d.	n.d.	n.d.
4.7-5.8	7.45	2.43	27.14	4.81	10.13	3.64	2.36	4.00	n.d.	3.89	n.d.	239.71	n.d.	n.d.	n.d.	n.d.
5.8-9.0	7.47	2.84	6.72	4.61	14.43	2.81	2.67	3.08	n.d.	n.d.	49.29	291.59	n.d.	n.d.	n.d.	n.d.
9.0-10.0	7.16	2.51	7.77	3.61	11.11	1.97	3.24	2.28	n.d.	n.d.	n.d.	213.52	n.d.	n.d.	n.d.	n.d.
Total	**85.72**	**30.00**	**126.52**	**55.87**	**662.79**	**68.67**	**888.88**	**261.43**	**231.86**	**316.19**	**309.43**	**2,402.90**	**670.24**	**n.d.**	**124.11**	**25.86**

Figure 4. Size distributions of 3-6 ring PAHs adsorbed on particles during open burning of cable sheath (a) 3-ring PAHs, (b) 4-ring PAHs, (c) 5-ring PAHs and (d) 6-ring PAHs

3.3 Risk assessment

Figure 5 shows the ratios of 3-6 ring PAHs and carcinogenic PAHs adsorbed on particles during open burning of cable sheath. Ultrafine particles were composed of 2-ring PAHs (0.92% of total PAHs), 3-ring PAHs (5.84% of total PAHs), 4-ring PAHs (15.76% of total PAHs), 5-ring PAHs (76.59% of total PAHs) and 6-ring PAHs (0.89% of total PAHs). Fine particles contained 2-ring PAHs (1.05% of total PAHs), 3-ring PAHs (17.70% of total PAHs), 4-ring PAHs (41.01% of total PAHs), 5-ring PAHs (39.71% of total PAHs) and 6-ring PAHs (0.53% of total PAHs). Coarse particles comprised 2-ring PAHs (2.14% of total PAHs), 3-ring PAHs (13.40% of total PAHs), 4-ring PAHs (6.48% of total aPAHs) and 5-ring PAHs (77.98% of total PAHs). The inhalable particles (PM_{10}) were predominantly composed of 5-ring PAHs (55.67% of total PAHs) followed by 4-ring PAHs (27.58% of total PAHs) and 3-ring PAHs (14.98% of total PAHs) as shown in Figure 5(a). These results show that adsorption of low volatile PAHs onto particle surfaces was easier than that of high volatile ones due to their volatile property. This is closely related to the previous studies indicating that more than four aromatic rings readily adsorbed onto combustion aerosols [15-17]. However, only small amount of BgP, six-rings PAHs (0.41% of total PAHs) was observed. In this work, small amount of Nap attached on particles (1.36% of total PAHs) was detected even it is the most volatile PAHs. Several studies also found Nap in the particle phase [6, 36].

In this study, 16 PAHs were divided into four groups following the International Agency for Research on Cancer (IARC): BaP as carcinogenic to humans (Group 1), DbA as probably carcinogenic to humans (Group 2A), Nap, BaA, BbF, BkF, InP and Chr as possibly carcinogenic to humans (Group 2B), Acy, Ace, Flu, Phe, Ant, Fla, Pyr and BgP as not classifiable as to its carcinogenicity to humans (Group 3) [37]. The fraction of PAHs bound on ultrafine particles was mainly classified as Group 2B (79.59 % of the total mass), followed by Group 1 (10.85 % of the total mass), and Group 3 (9.56 % of the total mass) whereas Group 2A was not detected. The fraction of PAHs bound on fine particles was dominantly classified as Group 3 (46.56 % of the total mass) followed by Group 2B (37.01 % of the total mass), Group 1 (13.03% of the total mass), and Group 2A (3.40 % of the total mass), respectively. The fraction of PAHs bound on coarse particles was mainly classified as Group 2B (75.15 % of the total mass) followed by Group 3 (18.67 % of the total mass), Group 1 (6.18 % of the total mass) while Group 2A was not detected. The ratio of carcinogenic PAHs adsorbed on ultrafine particles was similar to that of coarse particles. The fraction of PAHs bound on the inhalable particles (PM_{10}) was primarily classified as Group 2B (53.45 % of the total mass) followed by Group 3 (33.86 % of the total mass), Group 1 (10.71 % of the total mass), and Group 2A (1.98 % of the total mass) as shown in Figure 5(b). The results reveal that some PAHs bound on particles during open burning of cable sheath were assessed as possible carcinogens to humans.
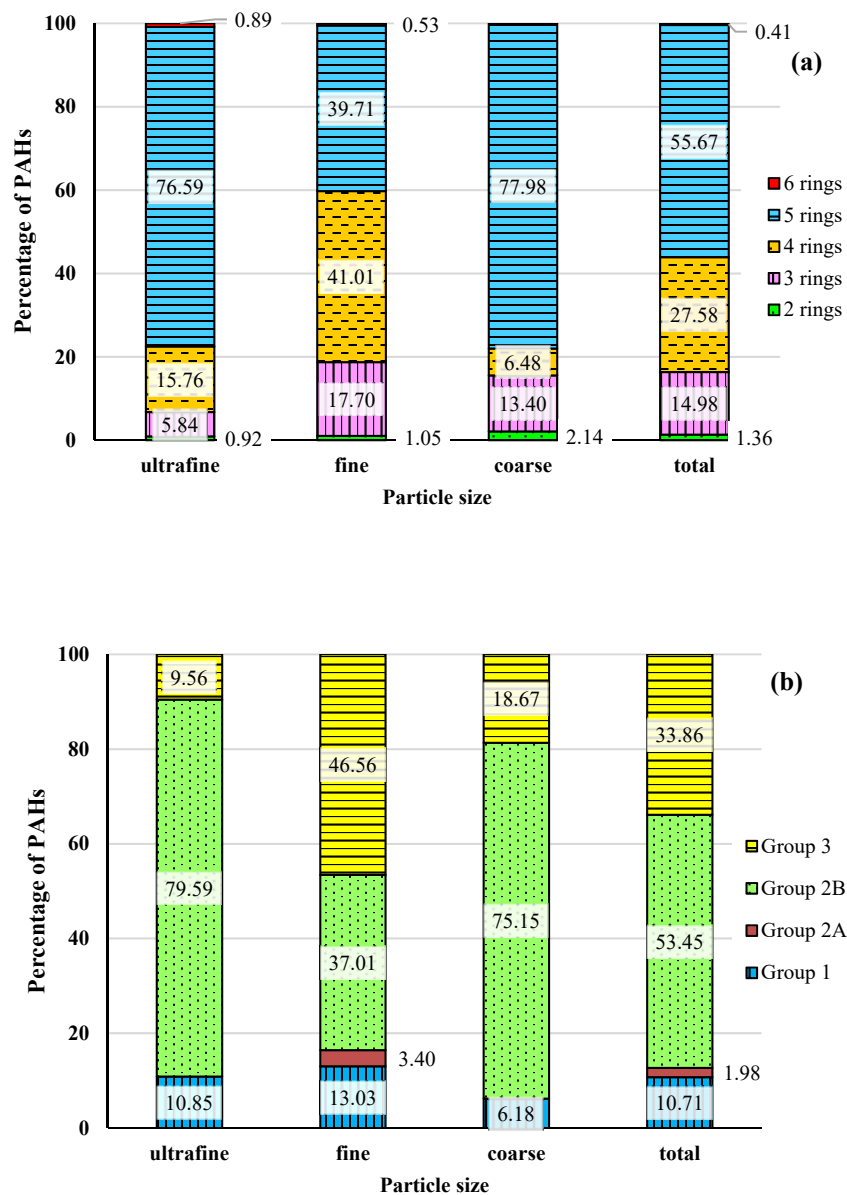
Figure 5. The ratios of 3-6 ring PAHs (a) and carcinogenic PAHs (b) adsorbed on particles during open burning of cable sheath

Table 2. Concentrations of 16 PAHs and B[a]P$_{eq}$ adsorbed on particles

Compounds	TEF	Cable sheath burning	
		PAHs (ng m^{-3})	B[a]P$_{eq}$ (ng m^{-3})
Nap	0.001*	85.71	0.08571
Acy	0.001*	29.99	0.02999
Ace	0.001*	126.54	0.12654
Flu	0.0005	55.88	0.02794
Phe	0.0005	662.78	0.33139
Ant	0.0005	68.67	0.03433
Fla	0.05	888.88	44.44423
Pyr	0.001	261.42	0.26142
BaA	0.005	231.86	1.15932
Chr	0.03	316.20	9.48589
BbF	0.1	309.43	30.94302
BkF	0.05	2,402.90	120.14512
BaP	1	670.24	670.23810
InP	0.1	n.d.	n.d.
DbA	1.1	124.11	136.52008
BgP	0.02	25.86	0.51719
Total		**6,260.47**	**1,014.35**
Lifetime lung cancer risk			**8.83E-02**

Note: TEFs derived from Larsen and Larsen [31] except TEFs* of Nap, Acy, Ace derived from Nisbet and Lagoy [30]
E-02 means additional cases per 100 people exposed.

Table 2 shows the concentrations of 16 PAHs bound on particles and BaP-equivalent during open burning of cable sheath. The concentrations of total PAHs and total B[a]P$_{eq}$ during wire burning were 6,260.47 ng m^{-3} and 1,014.35 ng m^{-3}, respectively. The B[a]P$_{eq}$ value during burning of cable sheath significantly exceeded 1 ng m^{-3} of the European Union's annual average standard [38]. The estimated lifetime cancer risk was 8.83E-02 (8.83 additional cases per 100 people exposed). The experimental results proposed that some PAHs released from open burning of cable sheath were established lung carcinogens resulting in high lung cancer risk, which is in consistent with the study of Wang *et al.* [19].

4. Conclusions

The ambient particles during open burning of cable sheath comprised coarse particles (49.59% of the total mass) and fine particles (38.96% of the total mass), and ultrafine particles (11.45% of the total mass). The particle size distributions were bimodal with one major peak in the coarse mode (5.8-4.7 μm range) and another minor peak in the accumulation mode (1.1-0.65 μm range). Total PAHs were dominantly bound on fine particles (58.23% of the total PAHs) followed by coarse

particles (30.30% of the total PAHs) and ultrafine particles (11.47% of the total PAHs), respectively. The inhalable particles mainly contained 5-ring PAHs (55.67% of total PAHs) followed by 4-ring PAHs (27.58% of total PAHs) and 3-ring PAHs (14.98% of total PAHs), respectively. Some of the PAHs were determined as possibly carcinogenic to humans (Group 2B). The average concentrations of 16 PAHs and $B[a]P_{eq}$ during open burning of cable sheath were $6,260.47$ ng m^{-3} and $1,014.35$ ng m^{-3}, respectively. The estimated lifetime lung cancer risk during wire burning was $8.83E\text{-}02$ (8.83 additional cases per 100 people exposed). The exposure of PAHs adsorbed on particles during open burning of cable sheath was potentially high risk for developing lung cancer.

5. Acknowledgements

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