Risk Assessment of Emitted Particle-bound Polycyclic Aromatic Hydrocarbons from Lignite-biomass Pelletization Burning: Size Distribution and Human Health Effects

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Abstract

Risk assessment of emitted particle-bound polycyclic aromatic hydrocarbons (PAHs) from lignite-biomass pelletization burning focusing on size distribution and human health effects are investigated. The particles matters and PAHs are experimented by a tube furnace and high performance liquid chromatography coupled diode array and fluorescence detectors (HPLC-DAD/FLD), respectively. The carcinogenic, mutagenic, and toxic potencies of PAHs are also discussed in relative to the size distribution of emitted particle PAHs. The results indicate that the carcinogenic, mutagenic, and toxic potencies of PAHs are likely to accumulate in the ultrafine particles finer than 0.65 µm. The ultrafine particles contribute the largest PAHs existences of carcinogenic, mutagenic and toxic substances; even though they register in the small mass fraction of PAHs. Moreover, the co-pellets burning can reduce the values of carcinogenic PAHs, carcinogenic equivalency quotients (BaP-TEQ), mutagenic equivalency quotients (BaP-MEQ), and toxic equivalency quotients (TCDD-TEQ) by more than 60 %.

Keywords: Polycyclic aromatic hydrocarbons, pelletization, burning, risk assessment, size distribution.

Introduction

Lignite is an abundant material which is globally used for energy generation. It is believed that lignite material consists of many hazardous elements especially a great source of polycyclic aromatic hydrocarbons (PAHs) that have been contaminated the environment during mining, transportation, storages and discards. Its combustion also releases a great amount of carcinogenic PAHs [1,2]. Meanwhile, biomass is a good solution to support the energy demand which can accompany with lignite due to a huge availability and a carbon neutral material. Rubber wood is a major fuel for drying the rubber sheets and generating the energy in Thailand [3-5]. Even though the rubber wood, functioned as biomass, is the renewable or zero-net carbon fuel, its combustion emits a huge value of particulate matters and its associated PAHs, especially the existences of carcinogenic and mutagenic PAHs [3,5,6]. A significant source of ambient particles and PAHs mainly cause from the fuel combustion.

The PAHs are the elements emitted from the incomplete burning or pyrolysis of fuels, which exist the complex aromatic structures [7]. The different PAHs characteristics can be influenced by the different fuel properties and combustion condition [7,8]. Wen *et al.* [9] reported on the coal combustion that the

PAHs seemed to be highly occurred at the low combustion air and temperature. The coal combustion at 350 ℃ with air flowrate of 0.3 L/min produced the highest concentration of PAHs of 352.98 μg/g fuel while the coal combustion at 950 ℃ with air flowrate of 1.2 L/min produced 0.34 μg/g fuel. The combustion was dominated by the light molecular weight PAHs (LWPAHs < 228 g/mol). The heavy molecular weight PAHs (HWPAHs > 228 g/mol) also increased when the temperature increased. On the other hand, Hata *et al.* [10] and Furuuchi *et al.* [3] reported that the combustion of biomass was the significant sources of ambient particles and particle-bound PAHs. The particle-bound PAHs strongly accompanied with the fine particles. An example, the biomass combustion released 7 - 700 mg/g fuel of 4 - 6 aromatic ring PAHs in $PM_{0.07}$. Moreover, Peng *et al.* [8] reported on the co-combustion of coal and municipal solid waste at the different temperatures. The municipal solid waste combustion and the coal combustion alone emitted the PAHs concentrations in range 0.11 - 10.05 mg/g and 0.18 - 3.62 mg/g, respectively. The coal combustion above 800 ℃ (2.56 mg/g) emitted the total PAHs 5 folds higher than at 700 ℃ (0.49 mg/g). Unlikely, the municipal solid waste combustion released the highest concentration of PAHs at 700 ℃ (10.05 mg/g). The co-combustion of municipal solid and coal could trade the total PAHs concentrations in range 0.12 - 2.4 mg/g at which the maximum PAHs concentrations occurred at 900 ℃. Most of mass concentration of PAHs was dominated by the 3 - 4 aromatic ring PAHs such as ANT and PHE, whereas toxicity degree of PAHs was dominated by the 5 - 6 aromatic ring PAHs such as BbF, BaP, and DahA [8,11]. Furthermore, it was reported that the compressed fuels could reduce the particulate matters and PAHs as compared to the uncompressed fuels [7,12].

The PAHs are the well-known harmful emissions that seriously affect to the human health. Some of PAHs are known to be carcinogenic, mutagenic, and teratogenic. The PAHs can accumulate in the ambient particles that travel around the workplaces or in the atmosphere. The coarse particles easily deposit in the workplaces while the fine particles can transport in the long distances which contaminate the water, soil, air or food by the atmospheric precipitation. It is realized that the finer particles, the more human health effects in the repository system depending on their size distribution and the location of deposited particles. The coarse particles (PM_{10}) can only deposit in nasal cavity or trachea while the fine particles ($PM_{2.5}$) can go to bronchus. The seriously damage of respiratory system is the ultrafine particles (PM_1) that can penetrate to the end of alveoli [7,10,13]. In addition, the mass concentration of PAHs is not enough to evaluate the risk assessment of the human health. Several risk assessments have been developed to equity the components of PAHs to benzo(a)pyene (BaP) or 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDD) basis. The carcinogenic, mutagenic, and toxic potencies can assess to possibility of lung and liver disease and even caner at the high doses. The use of risk assessment may permit a more accurate of human health effects [14-16]. Many approaches have been studied the PAHs emitted from biomass [3,5,10,17], coal [1,2,9], or their blend combustion [8,18]. However, the particle-bound PAHs related to the size distribution emitted from the pellet combustion are few addressed. Moreover, the risk assessments of carcinogenic, mutagenic and toxic potencies of PAHs are rarely evaluated in the load of human health risk. In particular, the information surrounding the particle-bound PAHs and its risk assessment is essential to gain the knowledge of impacts of biomass and lignite pellet combustion, that pollute to the atmosphere and respiratory system of human.

The aims of this present study are to investigate the risk assessment of emitted particle-bound PAHs from the lignite-rubber wood sawdust pelletization burning. The co-pelletization combustion and the PAHs analysis are conducted by a tube furnace and HPLC-DAD/FLD, respectively. The risk assessment that affects the human health is evaluated via the carcinogenic, mutagenic, and toxic potencies of PAHs. The size distribution of hazardous PAHs is also discussed.

Materials and methods

Pellets

Pellets are co-pelletized by lignite and rubber wood sawdust collected from Lampang province and Songkhla province, Thailand, respectively. The pellets are produced using a screw press with 10 %wt of starch as a binder. The dimension of pellets such as 100 % rubber wood sawdust (R), 50 % rubber wood sawdust and 50 % lignite (RL), and 100 % Lignite (L) result in 8 - 10 mm of diameters. Furthermore, the lengths of pellets are manually selected or cut in 3.5 - 4 of length-to-diameter ratio. As seen in **Figure 1**, the burning rate decreases in addition of lignite, whereas the pellet density increases. This result shows a good agreement with Jiang *et al.* [19]. However, the compressive strength is approximately 4.2 MPa of RL followed by 3.1 MPa of R and 2.4 MPa of L. Jiang *et al.* [19] reported that the high density and strength of sludge and biomass pellets would be decreased the burning rate by the release of volatile matter untighten the structures of pellets. This consequence may cause from the same components of sludge and biomass: lignin, hemicellulose and cellulose. In this study, the lignin inside the rubber wood sawdust can improve the bounding mechanism of aromatic ring structures / unburnt minerals of lignite by transformed itself to wax and oil [20,21]. It can be seen that the properties of biomass can enhance the density and compressive strength of lignite.
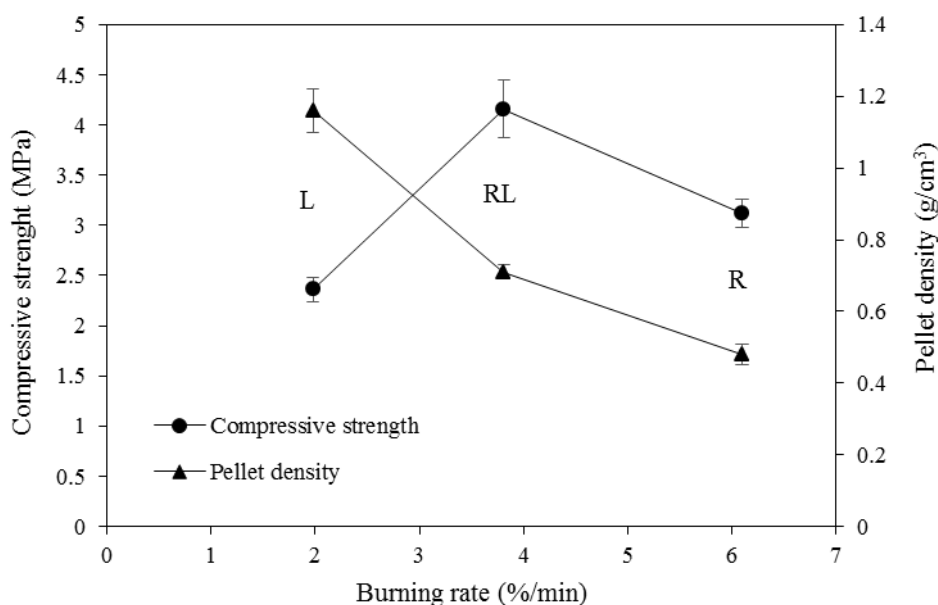

Figure 1 Characteristics of pellets basing on burning rate during devolatization stage of thermal analysis versus compressive strength and pellet density.

Characteristics of pellets are shown in **Table 1**, where C, H, O, N, and S are analyzed using CHNS-O analyzer model CE Instrument Flash EA 1112 series (Thermo Quest, Italy); moisture content is determined by ASTM D-3173; ash is determined using the removed ash of tube furnace as descripting in experimental set up; and heating value is examined by Dulong's equation.

Table 1 Characteristics of pellets (n = 3).

Components/pellets (%db.)	R	RL	L
Carbon, C	48.40	45.38	41.92
Hydrogen, H	6.46	5.38	4.18
Oxygen, O	42.93	35.65	26.42
Nitrogen, N	0.15	0.73	1.31
Sulfur, S	-	1.60	3.33
Moisture	5.62	5.38	6.99
Ash	1.05	12.73	25.76
Heating value, HHV* (MJ/kg)	17.95	16.84	15.46

* $HHV = 33.823C + 144.25(H - 0.125O) + 9.419S$

Experimental setup

A schematic diagram of combustion system is shown in **Figure 2**. Eleven pellets are randomly selected to burn in a tube furnace as descript in a previous study [22]. A quartz column is inserted to the furnace with dimensions: 0.06 m of diameters and 0.85 m of length. The pellets are placed on ceramics boats at a center of heating zone. The pellets are burnt for 5 times at 5 ºC/min from room temperature to 700 ºC. To ensure all pellets burnt out, the combustion is maintained for 20 min at 700 ºC. Furthermore, the combustion air, approximately 1.6 L/min which is controlled by 1 mm-diameter orifice meter connected with the u-tube manometer, is purged into the inserted quartz column of furnace. A dilutor is used to reduce the moisture and temperature of exhaust. The particulate matters are collected by Anderson cascade impactor using an initial filter (ANIF) at 28.3 L/min of flow rate. Quartz filters (D = 80 mm) treated for 72 h in a lightless desiccator (25 ºC, 50 % RH) are used in the cascade impactor. Moreover, the flue gas was tested by the Testo 350XL gas analyzer at 1.03 L/min of flow rate. The ash is removed and weighed when the furnace cools down to the room temperature.
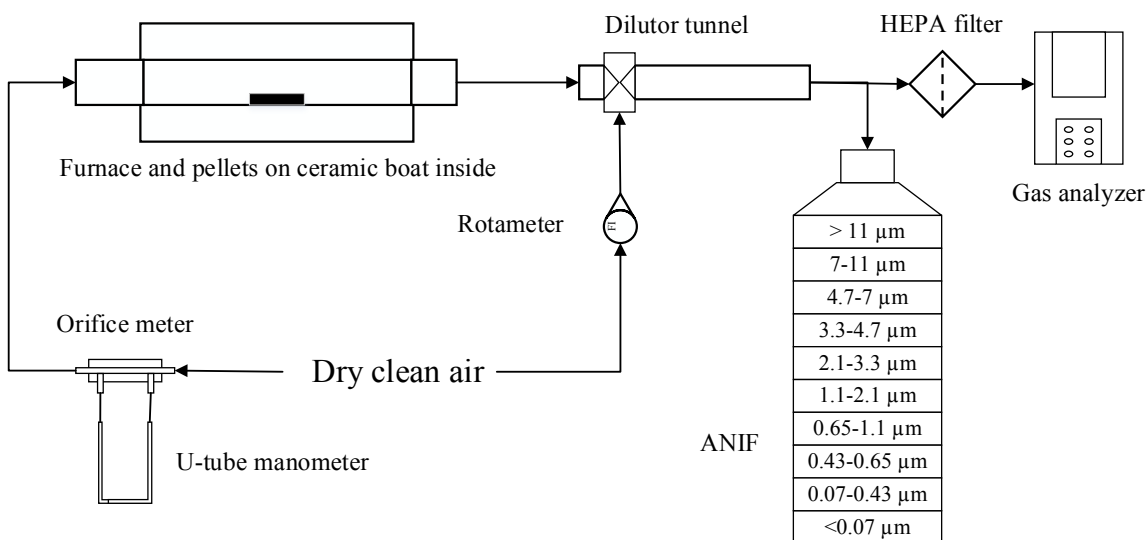

Figure 2 Combustion system for emission testing.

Particulate analysis

Before analyzing the PAHs, the filters chopped into small pieces are ultrasonically extracted in 20 mL of dichloromethane for 20 min twice. After that, a rotary evaporator is used to remove the dichloromethane after adding 50 µL of dimethyl sulfide. The extracted is kept in a freezer at 4±2 ºC for further PAHs analysis.

The 16 priority PAHs as displayed in **Table 2** are analyzed by HPLC-DAD/FLD as described in the previous study [22]. The PAHs analysis is operated in a gradient condition of acetonitrile and ultra-pure water. The mobile phases of 70 - 100 % acetonitrile are flowed into the system at 0.5 mL/min. Timing of FLD detector is set in accordance with selected excitation and emission wavelengths of each PAHs components from 280 - 433 nm, whereas timing of DAD detector is followed the gradient condition at 254 nm of wavelengths. To assure the analysis, the recovery and calibration curves are confirmed to be 72 - 138 % and 0.99 of r-square. In addition, the detection limit is defined by the lowest concentration of PAHs in which the HPLC system can detect in term of signal to noise ratio larger than 3. The detection limit of each component PAHs is 5 - 99.2 ng/mL. Meanwhile, the blanks are also prepared. The PAHs results are corrected by the analyzed PAHs subtracting the blank PAHs.

Table 2 Equivalency factors of 16 priority PAHs.

PAHs components	Abbr.	Rings	MW	BaP-MEF [14]	BaP-TEF [23]	TCDD-TEF [16]
Naphthalene	NAP	2	128.	-	0.001	-
Acenaphthylene	ACY	3	152	0.00056	0.001	-
Acenaphthene	ACE	3	154	-	0.001	-
Fluorene	FLU	3	166	-	0.001	-
Phenanthrene	PHE	3	178	-	0.001	-
Anthracene	ANT	3	178	-	0.01	-
Fluoranthene	FLA	4	202	-	0.001	-
Pyrene	PYR	4	202	-	0.001	-
Benzo(a)anthracene[***]	*BaA*	4	228	0.082	0.1	0.000025
Chrysene[***]	*CHY*	4	228	0.017	0.001	0.0002
Benzo(b)fluoranthene[***]	*BbF*	5	252	0.25	0.1	0.00253
Benzo(k)fluoranthene[***]	*BkF*	5	252	0.11	0.01	0.00487
Benzo(a)pyrene[***]	*BaP*	5	252	1	1	0.000354
Dibenz(a,h)anthracene[***]	*DahA*	5	278	0.29	1	0.00203
Benzo(g,h,i)perylene	BghiP	6	276	0.19	0.01	-
Indeno(1,2,3-cd)pyrene[***]	*IcdP*	6	276	0.31	0.1	0.0011

Abbr. = abbreviation, MEF = mutagenic equivalency factors, TEF = toxic equivalency factors,
MW = molecular weights (g/mol)
[***] Probable human carcinogenic according to U.S. EPA [24].

Results and discussion

Flue gas particulate matters and PAHs

Mass concentrations of flue gas, particulate matters, and PAHs are shown in **Table 3**. The mass concentration of particulate matters and PAHs strongly emit from rubber wood sawdust pellets combustion due to the high volatile matter of biomass [7,25]. The high volatile matter of biomass may cause the combustion flame and the reactivity unstable. It results in high concentration of CO [17,26].

Furthermore, the emission of SO_2 and NO_2 are low due to the low S and N contents in the pellets, respectively. In case of using the dilutor, the sensor of gas analyzer cannot detect the insufficient amount of CO_2 and NO_2. However, the SO_2 emission appears in the R combustion due to the equipment error and uncleanness of combustion column in the dead zones on each site of furnace.

Table 3 Summarize of flue gas, particulate matters and PAHs concentrations emitted from pellet combustion.

Emission	R	RL	L
CO (ppm)	1,979	1,705	1,024
CO_2 (ppm)	-	-	-
NO (ppm)	31	29	8
NO_2 (ppm)	-	-	-
SO_2 (ppm)	6	29	48
Particulate matters (mg/g fuel)	109	63	19
PAHs (µg/g fuel)	89	32	18

The size distribution of particulate matters emitted from the pellet combustion is shown in **Figure 3**. The results of all conditions are calculated from the total concentration of particles and volume of gas that purge into the furnace. The results present that the fine particles ($PM_{2.5}$) and ultrafine particles (PM_1) emit 60 - 90 and 30 - 80 % of the exhaust, respectively.
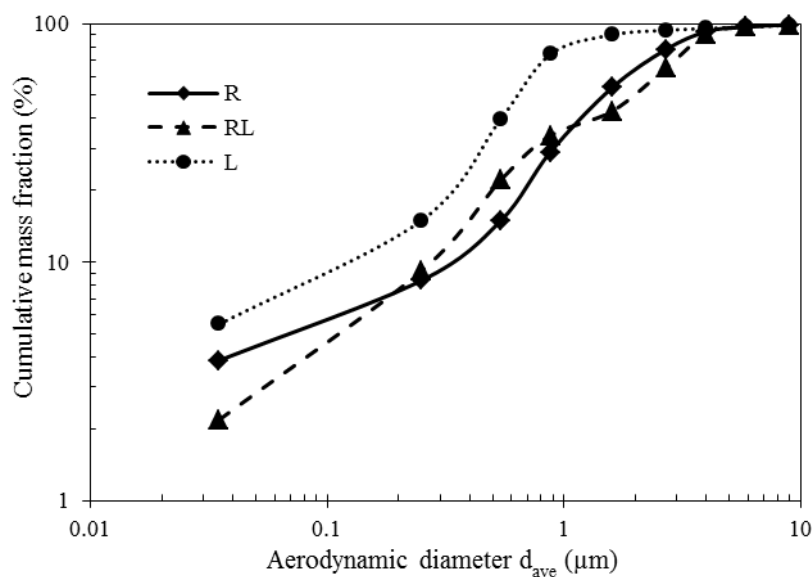

Figure 3 Size distribution of particulate matters emitted from pellet combustion.

Figure 4 indicates that the R and L combustion display single modal behaviors in a coarse mode and an accumulate mode, respectively. The RL combustion displays a bi-modal behavior because of the different behaviors of R and L particles. The peaks of R and L take place on 0.6 and 2.5 µm, respectively,

whereas the peak of RL is on both 0.6 and 2.5 µm. The fine particle PAHs might transport in the long distance and deposit in critical area of lung which increase more toxic condition as mentioned above. It can be concluded that the different properties of R and L can split the particle PAHs behavior of RL to the coarse mode.
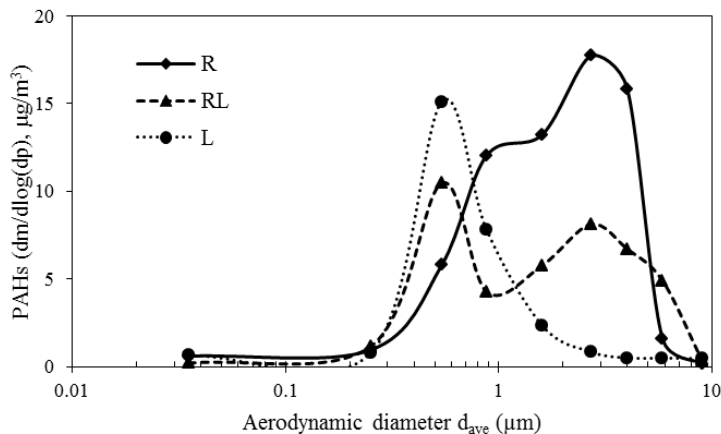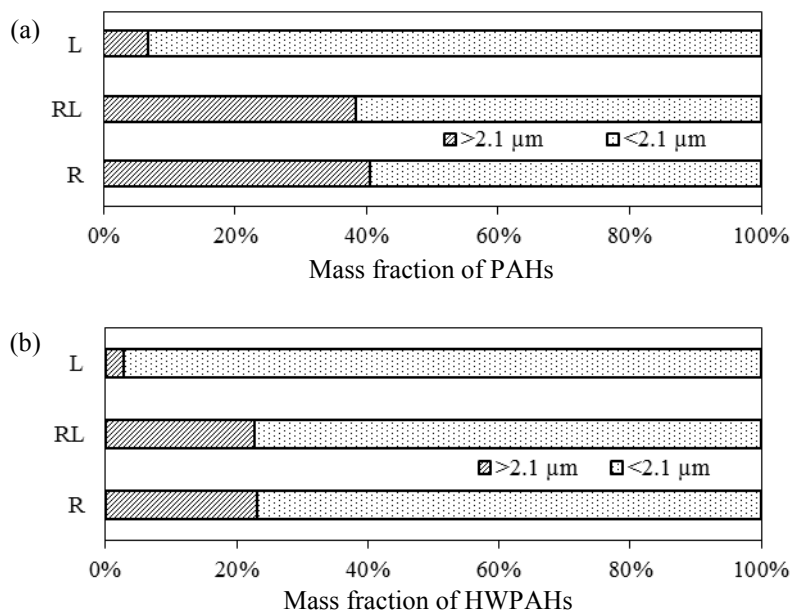

Figure 4 Size distribution of PAHs in each size range of particulate matters.
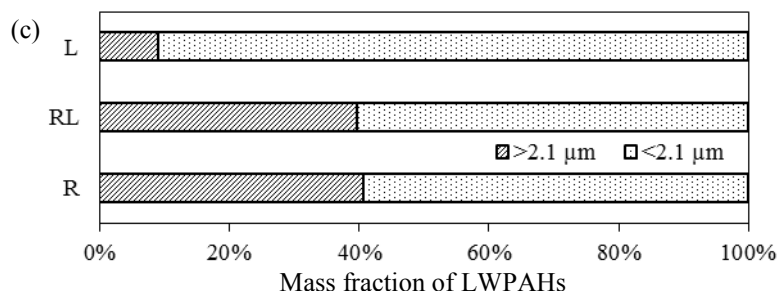
Figure 5 Mass fraction of (a) 16 PAHs, (b) heavy molecular weight PAHs, and (c) light molecular weight PAHs in particle size higher or lower than 2.1 μm.

Figure 5 indicates the mass fraction of PAHs, HWPAHs, and LWPAHs related the size separation of $PM_{<2.1}$ and $PM_{>2.1}$. The total mass concentration of PAHs of R simply emit as high as the total mass concentration of particulate matters following RL and L. The mass fraction of PAHs of R and L are abundant 60 % of $PM_{<2.1}$ while the mass fraction of PAHs of L are more than 90 % of $PM_{<2.1}$ (**Figure 5(a)**). In comparison between LWPAHs and HWPAHs, the LWPAHs in all conditions are pretty much higher than the HWPAHs similar to the study of Wen *et al.* [9]. However, the LWPAHs indicate in order of R > RL > L (11.67 - 87.91 μg/g fuel), while the HWPAHs indicate in order of R < RL < L (0.84-6.32 μg/g fuel). In addition, the HWPAHs are abundant more than 80 % of $PM_{<2.1}$ while the LWPAHs are similar trend to the PAHs in $PM_{>2.1}$ (**Figures 5(b) - (c)**). The increment of $PM_{<2.1}$ bound PAHs can also be found similar to the atmospheric particles [13] and solid fuel combustion in an open burning condition [7].
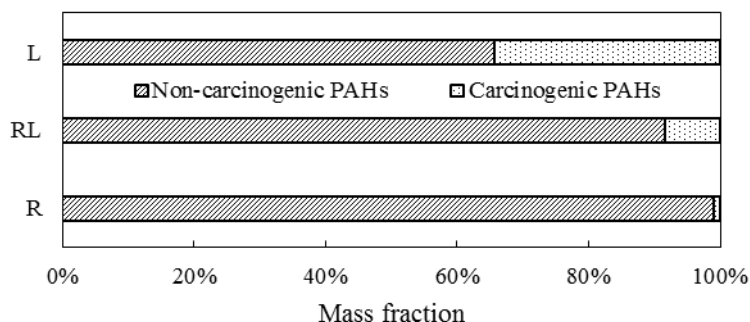

Figure 6 Mass fraction of carcinogenic and non-carcinogenic PAHs.

Figure 6 shows the mass fraction of carcinogenic and non-carcinogenic PAHs. The emitted mass fraction of carcinogenic PAHs is much fewer than the emitted non-carcinogenic PAHs. The carcinogenic PAHs present 30 % of L combustion, whereas lower than 10 % of RL and R combustion. Meanwhile, the carcinogenic PAHs of L combustion are greater than R combustion 7 folds of mass concentration. The contribution of carcinogenic PAHs or HWPAHs in the exhaust are more dangerous to the human health. Hence the risk assessment of PAHs is taken into account.

Size distribution of risk potency of PAHs
The risk potency stands for the harmfulness of human health. It is defined due to its chemical compounds and health effects [14-16]. The risk assessment of PAHs can hence be assessed by the

calculation of equivalent quotients of carcinogenic, mutagenic, or toxic potencies. The potencies are calculated using multiplying the concentration with the equivalent factors of each PAHs component (Eq. (1) - (3)).

$$BaP - TEQ = \sum (PAH_i \times BaP - TEF_i) \tag{1}$$

$$BaP - MEQ = \sum (PAH_i \times BaP - MEF_i) \tag{2}$$

$$TCDD - TEQ = \sum (PAH_i \times TCDD - TEF_i) \tag{3}$$

where $BaP - TEQ$, $BaP - MEQ$, and $TCCD - TEQ$ are carcinogenic equivalency quotients, mutagenic equivalency quotients, and toxic equivalency quotients which suppress the enzymatic activity of 7-etoksyresorufin O-deetylase in the liver cell, respectively. $BaP - TEF_i$, $BaP - MEF_i$, and $TCCD - TEF_i$ are BaP carcinogenic equivalency factors, BaP mutagenic equivalency factors, and 2,3,7,8-tetrachlorodibenzo-p-dioxin toxic equivalency factors for i individual PAHs, respectively. PAH_i is an i individual PAHs.

   Figure 7 and **Table 4** indicate the risk assessment of PAHs related to the size distribution of PAHs and total mass concentration. It is confirmed that the L combustion emits the massive hazard PAHs in comparison with the RL and R combustion (**Table 4**). The L combustion generates the HWPAHs or the 4 - 6 aromatic ring PAHs in the exhaust is relative to the high equivalent factors. It is simply that the PAHs of L combustion risk to the human health 13 folds of mutagenic potency, 4 folds of carcinogenic potency, and 75 folds of liver toxic potency in comparison with R combustion. On the other hand, the carcinogenic PAHs, BaP-TEQ, and BaP-MEQ of L and RL combustion accumulate in the ultrafine particles of 0.43 - 0.65 µm (**Figure 7**), which have met a good agreement with the literatures [3,5,10]. However, the carcinogenic PAHs, BaP-TEQ and BaP-MEQ of R combustion accumulate in the fine particles of 1.1 - 2.1 µm. It may cause from the rapid reactivity of volatile matters, the agglomeration, and the condensation of particles in the unstable medium condition [7,26]. The TCDD-TEQ in all conditions accumulates in the nano-particles lower than 0.07 µm because of the abundant components of BbF, BaP, and DahA in $PM_{0.07}$.

Table 4 Summary of human risk PAHs concentrations emitted from pellet combustion.

PAHs	R	RL	L
Carcinogenic PAHs (ng/g fuel)	837.35	2624.19	6159.31
BaP-MEQ (ng/g fuel)	32.08	140.47	409.90
BaP-TEQ (ng/g fuel)	101.65	133.57	375.23
TCDD-TEF (pg/g fuel)	10.82	175.81	815.52

   It can be concluded that the greater hazardous PAHs or molecular weight PAHs result in the stronger accumulation of PAHs in the fine particles. The great surface areas of the fine particles may permit the absorption of PAHs to accumulate on. Other reasons include the high vapor pressures and the various chemical compositions of HWPAHs that transfer the gas to particle phase [7].
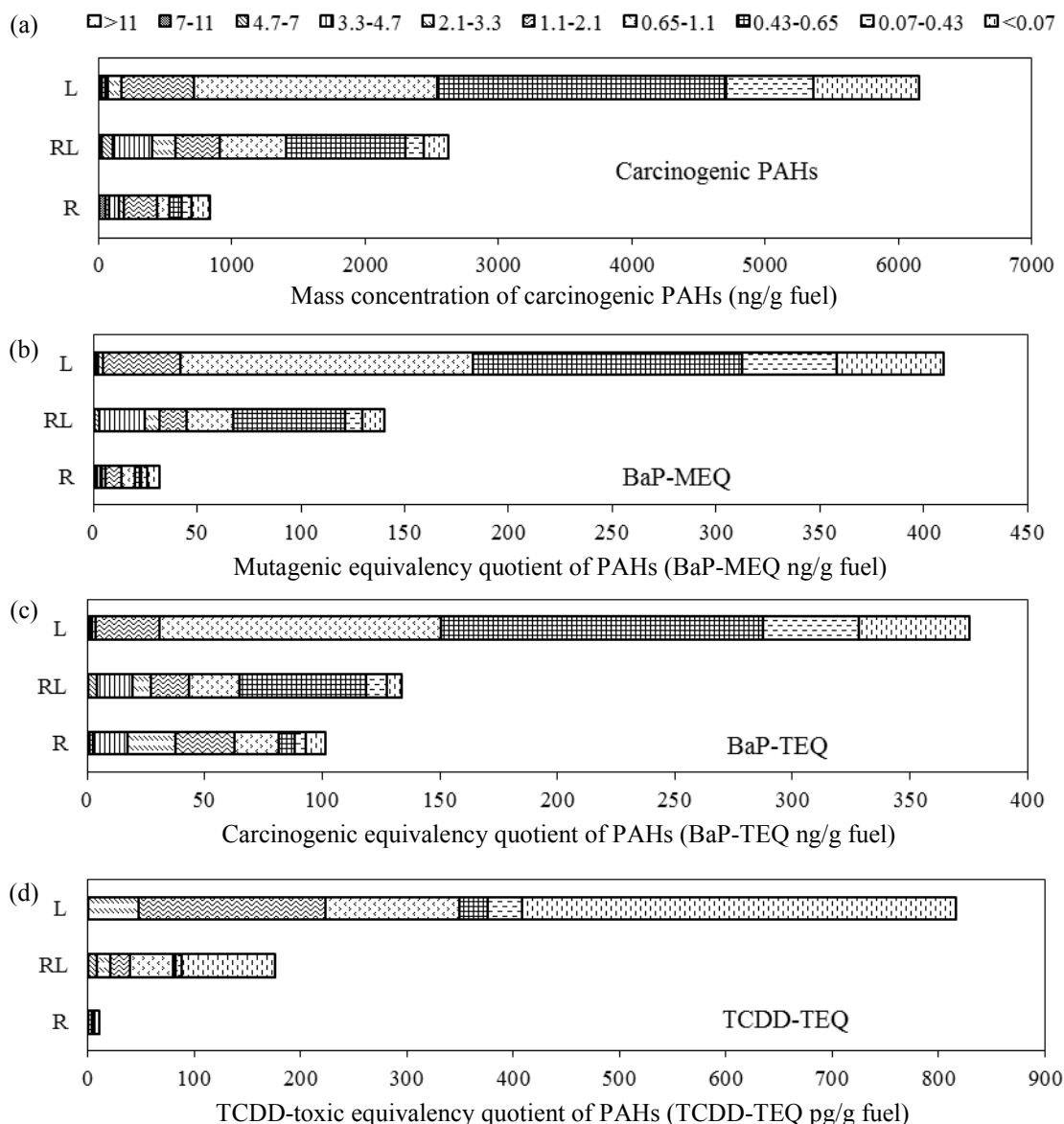
(a)   ☐>11 ■7-11 ▨4.7-7 ⊞3.3-4.7 ▨2.1-3.3 ▨1.1-2.1 ☐0.65-1.1 ▨0.43-0.65 ☐0.07-0.43 ☐<0.07

Figure 7 Risk assessment of PAHs: (a) mass concentration of carcinogenic PAHs (ng/g fuel), (b) Mutagenic equivalency quotient (BaP-MEQ ng/g fuel), (c) Carcinogenic equivalency quotient of PAHs (BaP-TEQ ng/g fuel), and (d) TCCD-toxic equivalency quotient of PAHs (TCDD-TEQ pg/g fuel).

Conclusions

The size distribution and risk assessment of human health effects of emitted particle PAHs from lignite-rubber wood sawdust pelletization burning are acquired that: (1) the L combustion causes seriously effects the human health as compared to the R and RL combustions even though the mass fraction of PAHs of L combustion is lower than R and RL combustions; (2) the RL combustion can reduce the values of carcinogenic PAHs, BaP-MEQ, BaP-TEQ, and TCDD-TEQ by more than 60 %; and

(3) the particle sizes smaller than 0.65 μm distribute the large fraction of PAHs and even the hazard PAHs and their toxicity. It is suggested that the pelletization burning between biomass and lignite should be operated instead of lignite burning alone in order to reduce the ultrafine or fine particles and the enrichment of hazard PAHs, which effect to the ambient air and human health.

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