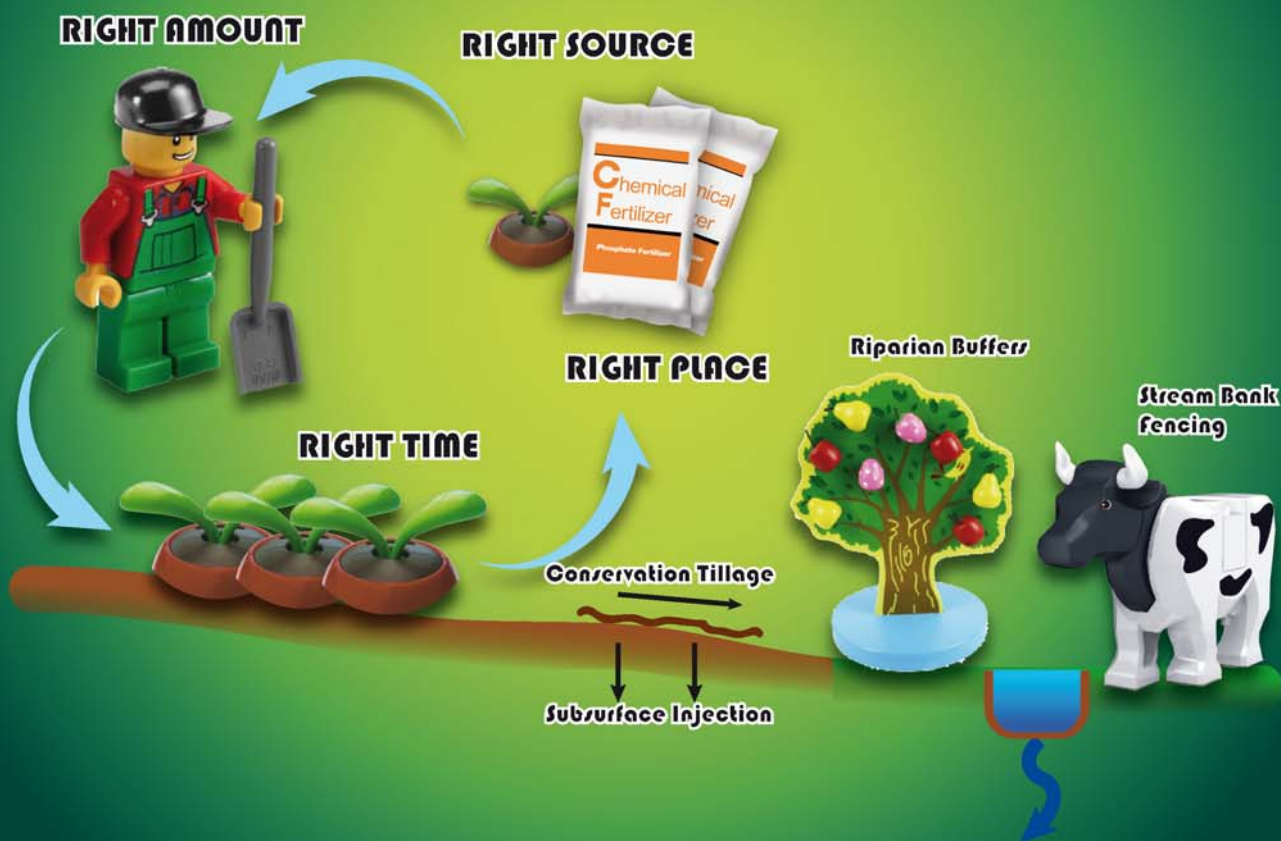


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# Characteristics of nanoparticles emitted from burning of biomass fuels

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## ABSTRACT

The characteristics of the particles of the smoke that is emitted from the burning of biomass fuels were experimentally investigated using a laboratory-scale tube furnace and different types of biomass fuels: rubber wood, whole wood pellets and rice husks. Emitted amounts of particles, particle-bound polycyclic aromatic hydrocarbons (PAHs) and water-soluble organic carbon (WSOC) are discussed relative to the size of the emitted particles, ranging to as small as nano-size (<70 nm), and to the rate of heating rate during combustion. Differential thermal analysis (DTA) and thermogravimetric analysis (TG) techniques were used to examine the effect of heating rate and biomass type on combustion behaviors relative to the characteristics of particle emissions. In the present study, more than 30% of the smoke particles from the burning of biomass fuel had a mass that fell within a range of <100 nm. Particles smaller than 0.43  $\mu\text{m}$  contributed greatly to the total levels of toxic PAHs and WSOC. The properties of these particles were influenced by the fuel component, the combustion conditions, and the particle size. Although TG–DTA results indicated that the heating rate in a range of 10–20°C did not show a significant effect on the combustion properties, there was a slight increase in the decomposition temperature as heating rate was increased. The nano-size particles had the smallest fraction of particle mass and particle-bound PAHs, but nonetheless these particles registered the largest fraction of particle-bound WSOC.

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## Introduction

Biomass energy is a solution to the increasing demand for renewable energy that can reduce carbon dioxide (CO<sub>2</sub>) emissions, which is necessary to address the issue of global warming.

However, the burning of biomass has a considerable regional and global impact on both the chemical properties of the atmosphere and the radiative balance of the Earth (Andreae and Merlet, 2001).

Although biomass fuel has carbon-neutral characteristics, biomass burning can be a significant source of ambient

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nanoparticles. The emission of fine particles in the atmosphere constitutes a serious concern for human health and contributes to photochemical smog. Fine particles play a particularly important role in health since this size distribution penetrates deeper than larger particles and causes damage to the most critical components of the respiratory system (Andrea, 2009). This is because the deposition site of inhaled particles changes with particle size, and the clearance time of the deposited particles varies depending on the deposition sites, leading to differences in toxicity, even for the same composition of particles. Therefore, it is necessary to determine the chemical composition of particles with respect to particle size, particularly for nanoparticles. In addition, how the characteristics of nanoparticles vary during the combustion process has not been investigated in detail.

Polycyclic aromatic hydrocarbons (PAHs) and water soluble organic carbon (WSOC) are typical components emitted during the burning of biomass (Mayol-Bracero et al., 2002; Furuuchi et al., 2006; Bignal et al., 2008; Tekasakul et al., 2008; Chomanee et al., 2009). Some PAHs are known to be carcinogenic (IARC, 1982), and exposure to high levels of PAHs has produced immunosuppressive effects (Leo, 2005). WSOC compounds are a matter of interest not only because WSOC in an aerosol form exerts climate effects such as contributing to cloud condensation nuclei generation (Redemann et al., 2001; Andreae and Merlet, 2001; Kundu et al., 2010), but because WSOC is a major environmental concern due to the presence of natural organic carbon in water that interact with trace amounts of pollutants when transferred into a natural water environment. Dissolved organic carbon has been recognized as a major carrier of trace metals and persistent organic contaminants that are relatively insoluble and immobile (Jonnalagadda and Nenzou, 1996). Furthermore, dissolved organic carbon in water is believed to be one of the important precursors for the generation of trihalomethanes and other chlorinated products (Tao, 1996), which have produced carcinogenic effects in various test organisms.

Investigation into the physicochemical properties of smoke particles from the burning of various kinds of biomass fuels have indicated that biomass combustion can be a significant source of fine particles and gaseous matter in ambient air, which then leads to environmental loads and serious health risks (Sippula et al., 2007), and most PAHs are associated with fine particles (Choosong et al., 2007; Furuuchi et al., 2006, 2007a,b; Tekasakul et al., 2008; Hata et al., 2009b).

Variations in the emission of smoke PAHs depend on the type of biomass fuel, the combustion process (Oanh et al., 2005), the biomass blending ratio, the excess air ratio, and on the moisture content of the fuel used in the combustion process (Chao et al., 2008; Chomanee et al., 2009). However, current understanding of the effects of the combustion parameters on PAH formation of particulate emissions from biomass combustion remains to be at the initial information stage and is rarely used either for source identification or for risk evaluation. In particular, information surrounding the physicochemical characteristics of smoke particles in the nano-size range is of particular importance in order to gain an understanding of the impact that burned biomass has on ambient air that can enter the respiratory system and affect the general health of an individual.

In the present study, a laboratory-scale electric tube furnace was used to examine the characteristics of particulate matters emitted from the combustion of different biomass fuels, rubber wood, whole wood pellets and rice husks—all examples of biomass fuels that are typically not used in Asian societies. The emitted amount of particles per unit of fuel mass was evaluated for different sizes of particles down to the nanosize range (<70 nm). Particle-bound PAHs and WSOC are discussed here relative to the contributions of different sizes of particles. The influence of the heating rate during combustion also is discussed.

## 1. Experimental

### 1.1. Tested biomass fuels and their thermal properties

The characteristics of biomass fuels are listed in Table 1, where the moisture content as well as the carbon, hydrogen, nitrogen and sulfur contents of biomass fuel were measured using a moisture analyzer HB43 (Mettler Toledo, Greifensee, Switzerland) and a CHNS analyzer vario III (Elementar Analysensysteme GmbH, Hanau, Germany).

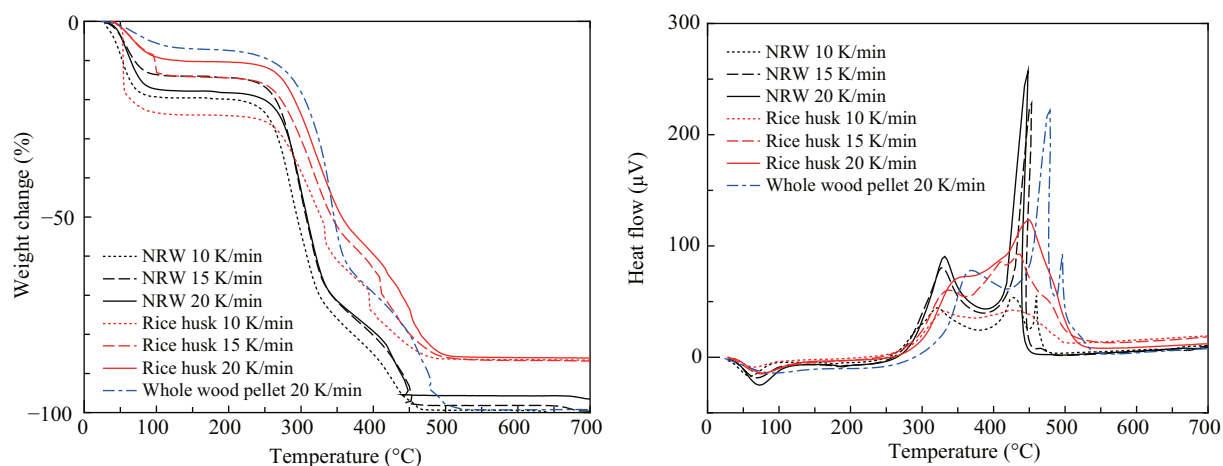
Fig. 1 shows the TG/DTA TG8120 (thermogravimetric/differential thermal combined analyzer, Rigaku, Tokyo, Japan) analysis results of 4.0–6.0 mg of tested biomass samples at different heating rates of 10, 15 and 20°C/min to determine the differences in reaction steps from weight loss and exothermic change between heating rates under a temperature program. The results showed that thermal decomposition started at 280°C, and pyrolysis was essentially completed at 500°C. Above that temperature, there was no further weight loss. As shown in Fig. 1, the TG curves shifted to higher temperatures as the heating rate increased. The results from the TG and DTA curves of the pyrolysis process appeared to be divided into 3 weight-loss phases during the heating process: drying at 150°C, charring at 200–350°C, and calcining at more than 350°C (Hu et al., 2000). Based on those results, the following combustion test temperature was set at 700°C, which was deemed to be high enough for combustion.

### 1.2. Experimental setup and procedure

A schematic diagram of the experimental setup is shown in Fig. 2. A selected amount of biomass fuel, was added to a tube furnace that consisted of a quartz glass tube (I.D. = 45 mm, O.D. = 50 mm, L = 500 mm) installed in an electric tube furnace that had a heating zone with a diameter of 50 mm and a length of 300 mm.

**Table 1 – Contents in tested fuel samples.**

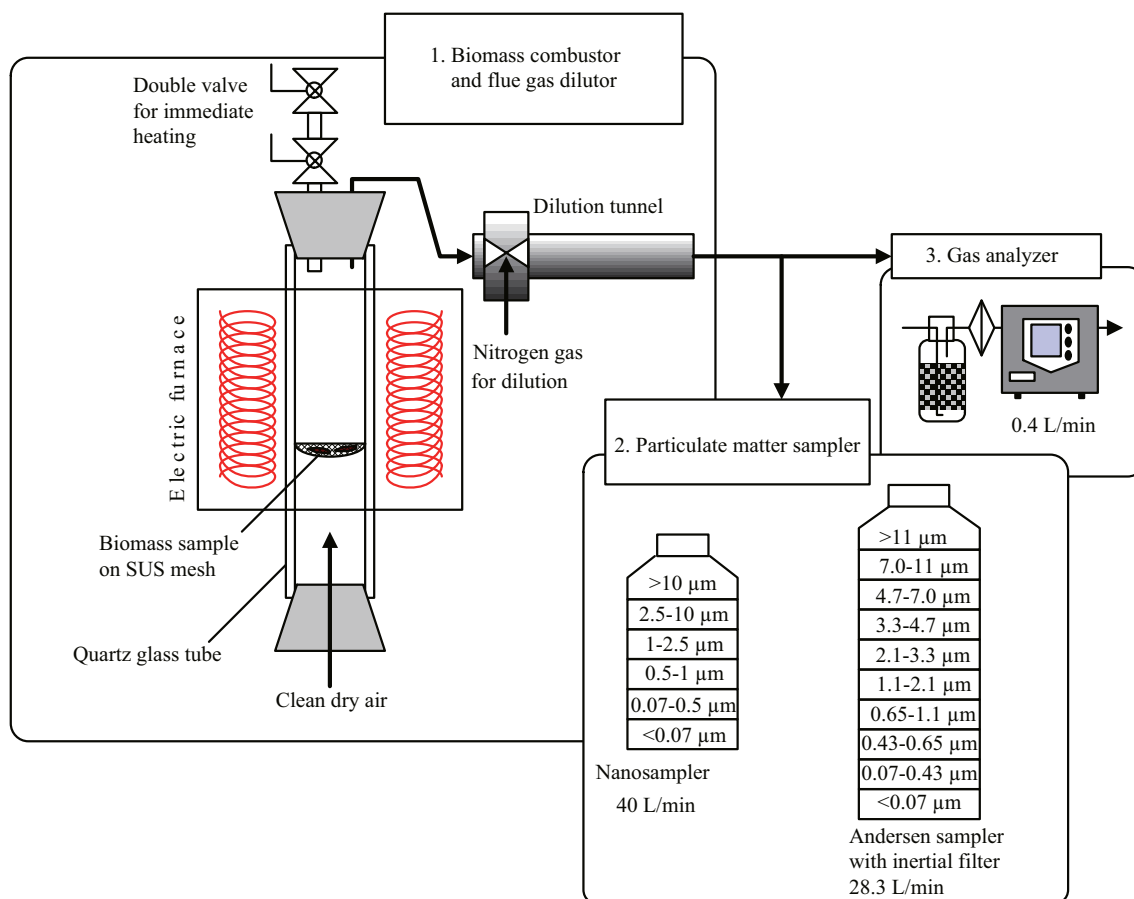
Sample	Component (%)				Moisture (%)
	C	H	N	S	
Natural rubber wood (NRW)	40	2	2	3	43.9 ± 4.3 (n = 6)
Rice husks	34	4	0.4	0	9.9 ± 0.5 (n = 5)
Whole wood pellets	47	7	1	0.3	11.0 ± 1.0 (n = 5)
n: sample numbers.					



**Fig. 1 – TG (a) and DTA (b) curves of biomass samples, natural rubber wood (NRW), rice husk and whole wood pellet with variations in heating rates. PAH: polycyclic aromatic hydrocarbons; WSOC: water-soluble organic carbon; DTA: differential thermal analysis; TG: thermogravimetric analysis; NRW: natural rubber wood; HEPA filter: high efficiency particulate air filter; ANIF: Andersen sampler with an inertial filter; HPLC: high performance liquid chromatography; PIC: product of incomplete combustion.**

To compare the heating processes of biomass, two different heating procedures were used for the combustion test of only whole wood pellets. Prior to the small tube combustion test, biomass pellets were carefully placed on

quartz combustion boats in the middle of the tube furnace for “Gradient heating”. The fuel samples were heated for 45 min until each reached 700°C and were maintained at 700°C for another 30 min. While the fuel samples were heated for



**Fig. 2 – Schematic diagram of the experimental setup.**

**Table 2 – Experimental conditions.**

Gas feeding conditions: Clean dry air 5 L/min	
Sampling conditions	Natural rubber wood and rice husks Andersen sampler with an inertial filter (ANIF), 28.3 L/min, 10 stages as shown in Fig. 2 (Whole wood pellets) Nanosampler (NS), 40 L/min, 5 stages as shown in Fig. 2
Heating conditions	
Gradient heating (all tested samples)	Setting temperature: 700°C Heating rate: 10, 15 and 20°C/min Holding time: 10 min
Immediate heating (whole wood pellets only)	Preheating temperature: 400, 500, 700, and 900°C

**Table 3 – Maximum CO and minimum O<sub>2</sub> concentrations relative to heating rates and heating conditions.**

Sample	Heating rate (°C/min)	CO <sub>max.</sub> (ppm)	O <sub>2-min.</sub> (vol.%)
Natural rubber wood (NRW)	10	456	16.9
	15	486	17.8
	20	576	17.6
Rice husks	10	962	16.3
	15	624	15.4
	20	642	11.3
Whole wood pellets	20	810	16.5
	Immediate heating	230	16.7

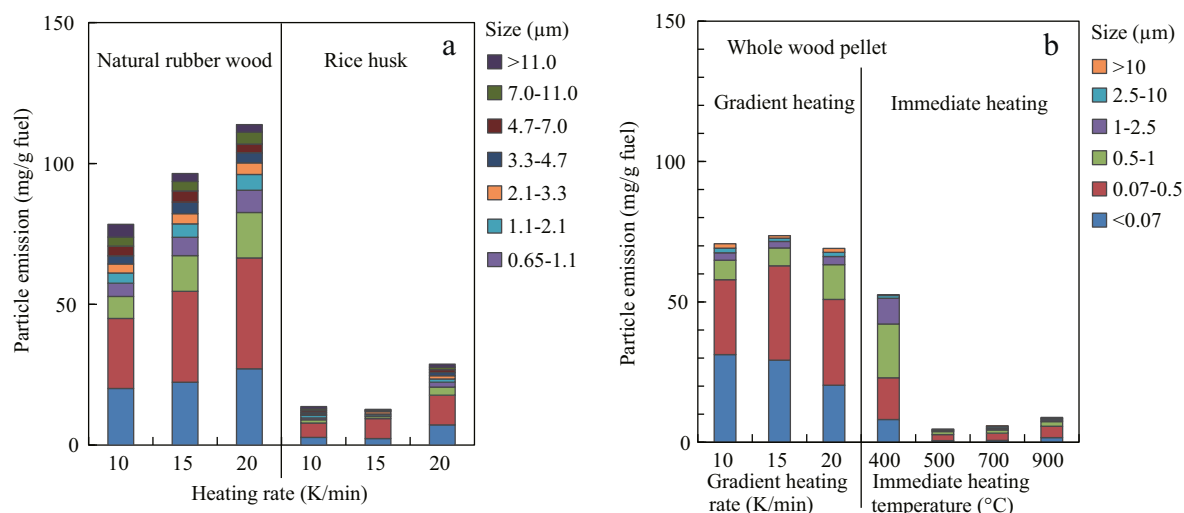
combustion, the air was cleaned via a HEPA filter and dried packed silica gel was fed at a constant flow rate via a mass flow controller MFC3650 (KOFLOC, Kyoto, Japan) into the furnace using an air pump. The flue gas from the furnace was diluted by mixing it with clean and dried nitrogen gas in a dilutor to reduce the temperature and the moisture content. In order to obtain a uniform dispersion of gas and smoke particles before the measurement,  $\phi$ 15-mm- and 750-mm-long stainless steel tube were used downstream from the dilution tunnel as a post-mixer.

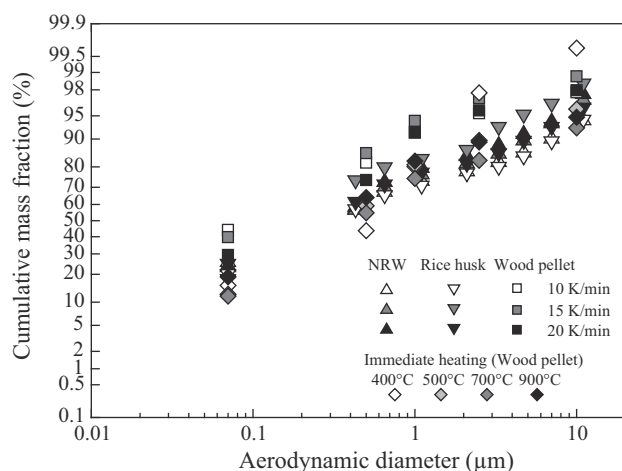
For an “Immediate heating test,” the biomass samples were kept in the tubes in a sealed area on top of the tube furnace until the tubes could be heated to 700°C. When this occurred, the fuel was dropped into the hot tube. In order to reduce the temperature and moisture content, the exhaust was diluted and cooled by mixing with a constant flow of nitrogen gas in a dilution tunnel (ID 18 mm, OD 20 mm, L 810 mm with an orifice diameter of 10 mm). The experimental conditions are summarized in Table 2.

Particulate matters were collected downstream from the diluter using an Andersen cascade impactor with an inertial filter stage (ANIF) attached between the 0.43  $\mu$ m stage and the backup filter (Hata et al., 2009a; Furuuchi et al., 2010) or using a Nanosampler (Furuuchi et al., 2010) that is capable of the offline analysis of TSP/PM<sub>10</sub>/PM<sub>2.5</sub>/PM<sub>1</sub>/PM<sub>0.5</sub>/PM<sub>0.07</sub>. The inertial filter for those samplers had a cutoff size of ca. 70 nm. Binder-less  $\phi$ 80 quartz fiber filters Pallflex 2500QAT-UP (Pall Corporation, Washington, New York, USA), prepared 48 h in a desiccator at room temperature (ca. 25°C) and ca. 50% RH, were used for the particle sampling through impactor stages. Concentrations of gaseous components, CO, CO<sub>2</sub>, O<sub>2</sub>, NO<sub>2</sub> and SO<sub>2</sub>, were continuously monitored by a gas analyzer PG-250 (Horiba, Tokyo, Japan) during the test run. Cut pieces of rubber wood, rice husks and whole wood pellets were used as test fuels. Fed amounts ranged from 0.1 to 0.5 g. To study the effect of operating conditions on the combustion and emission characteristics of biomass burned in a tubular furnace, the test runs were conducted at different heating rates and room temperature was adjusted to 10, 15 and 20°C/min. The experimental conditions are summarized in Table 2.

### 1.3. Analysis of emitted particles

Following the procedure established in a previous report (Furuuchi et al., 2006, 2007a,b), 16 particle-bound PAH compounds were analyzed: Naphthalene (Nap), Acenaphthene (Ace), Phenanthrene (Phe), Anthracene (Ant), Fluorene (Fle), Fluoranthene (Flu), Pyrene (Pyr), Benzo[a]anthracene (BaA), Chrysene (Chr), Benzo[e]pyrene (BeP), Benzo[a]pyrene (BaP),

**Fig. 3 – Particle mass emission per unit fuel mass for different size ranges relative to heating rates and heating conditions.**



**Fig. 4 – Emitted particle size distributions for tested biomass relative to heating rates and heating conditions.**

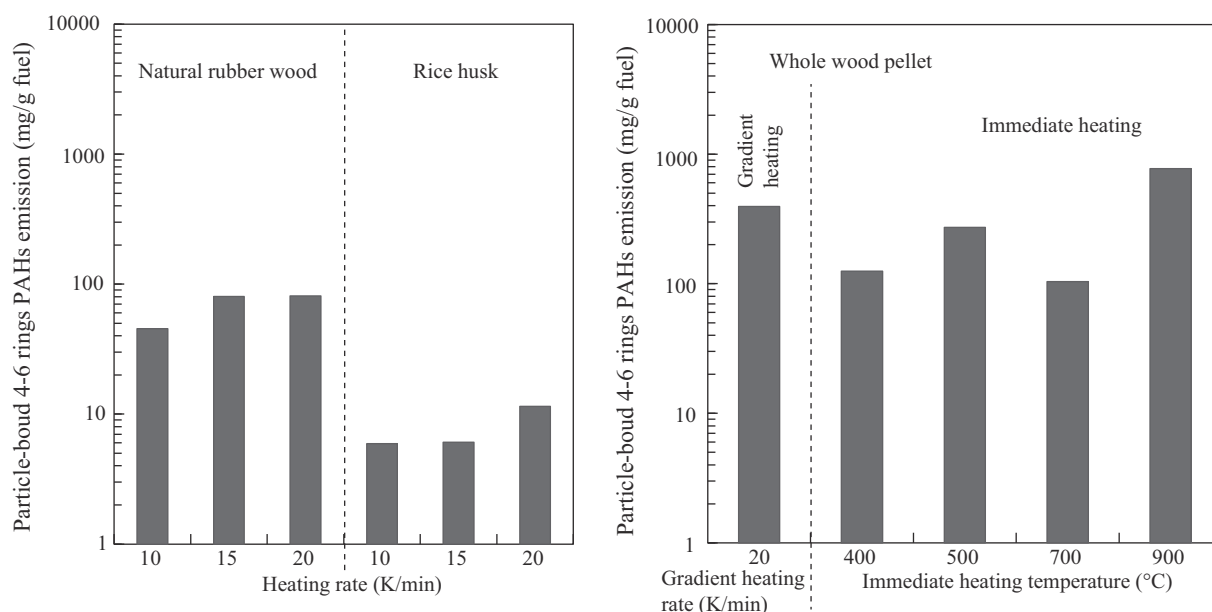
Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Dibenz[a,h]anthracene (DBA), Indeno[1,2,3-cd]pyrene (IDP), and Benzo[g,h,i]perylene (BghiPe). Filter samples cut into small pieces were ultrasonically extracted in ethanol:benzene (1:3, V/V) for 15 min. After adding 50  $\mu$ L of dimethyl sulfoxide (DMSO), the extracted solution was concentrated using a rotary evaporator NVC-2000 (EYELA, Tokyo, Japan). Extracted PAHs were analyzed using high-performance liquid chromatography (HPLC) L-2130/2200/2300/2485 (Hitachi High-Technologies Corporation, Tokyo, Japan) with a fluorescence detector and an Inertsil ODS-P reversed-phase column (particle size 5  $\mu$ m,  $\phi$ 4.6 mm diameter, 250 mm length) maintained at 70°C. The mobile phase was a mixture of acetonitrile and water with a gradient concentration mode of acetonitrile, and a flow rate of 0.4 mL/min. The timing of the fluorescence detector was set to detect at the optimum excitation and emission wavelengths for each PAH ranging

from 340 to 482 nm. The basic control method was programmed following the method established (Tang et al., 2005; Furuuchi et al., 2006, 2007a,b). The recovery efficiency was confirmed to be  $0.82 \pm 0.12$  ( $n = 3$ ) by adding a standard reagent (Accustandard, New Haven, Connecticut, USA, 0.2 mg/mL in  $\text{CH}_2\text{Cl}_2$ :MeOH (1:1)) to dissolved samples following the method established in previous reports (Tang et al., 2005; Furuuchi et al., 2006, 2007a,b). Once the filter blank values of the PAHs were confirmed to be small enough, then they were subtracted from analyzed values. Following the method established in a previous report (Yang et al., 2003), filter samples were cut into small pieces then ultrasonically extracted in 30 mL of ultra-pure water for 15 min. Extracts were then filtered using a 0.2- $\mu$ m nylon filter. The concentration of WSOC was determined using a total carbon analyzer TOC-V (Shimadzu Corporation, Kyoto, Japan). Filter blank values were subtracted from the analyzed amounts of samples.

## 2. Results and discussion

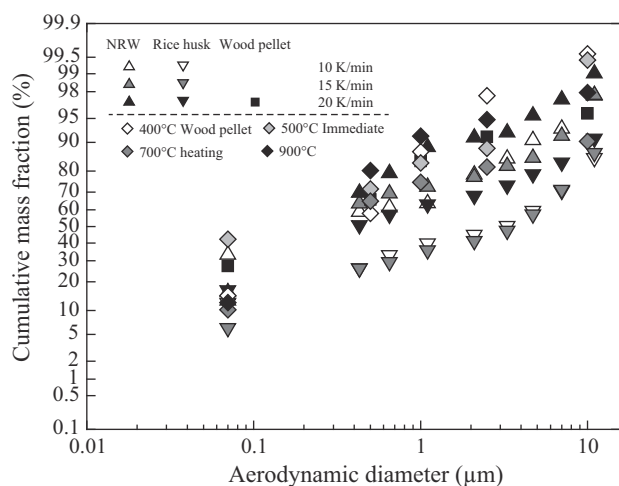
### 2.1. Gas components, mass concentration and size distribution of smoke particles

Table 3 indicates the maximum carbon monoxide and minimum oxygen concentrations relative to heating rates and heating conditions. Because particulate matters are Products of Incomplete Combustion (PIC), the concentration of the main gas components in the flue gas was monitored. The sensors had an insufficient amount of time to monitor the time change in detail, therefore, only the maximum and minimum concentrations are shown. CO concentration was lower than 1000 ppm for all tested conditions and  $\text{O}_2$  concentration was higher than 15 vol.% for most conditions. Concentrations of both  $\text{SO}_2$  and  $\text{NO}_2$  were less than 1 ppm at the background level.  $\text{CO}_2$  concentration was less than 0.3 vol.% for



**Fig. 5 – Particle-bound 4-6 ring PAH mass emission per unit fuel mass in  $\text{PM}_{0.07}$  relative to heating conditions.**





**Fig. 6 – Emitted particle-bound 4–6 ring polycyclic aromatic hydrocarbon size distributions for tested biomass relative to heating rates and heating conditions.**

all tested conditions. Based on the CHNS component analysis shown in Table 1, the amount of combustible air for tested samples ranged in 2.6–2.8 L/min. Because the air feed rate in the present study was fixed at 5 L/min, air was oversupplied so that the main factors of incomplete combustion were temperature and fuel conditions, as established by the following results.

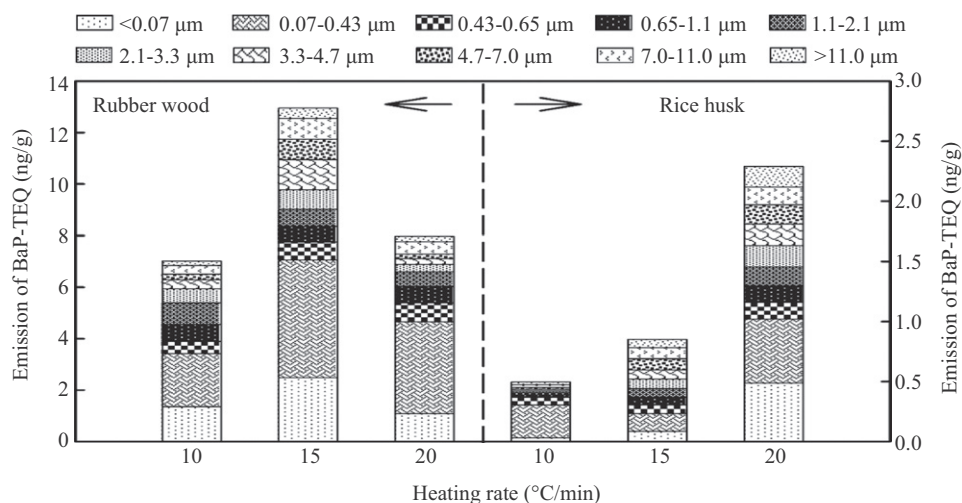
Fig. 3 shows total particle mass emissions per unit fuel mass calculated from the collected particle mass in the samplers and gas volume that was fed to the furnace. The results show that total particle emissions from combustion of the natural rubber wood and the rice husks increased with the heating rate, but results were not so clear for the whole wood pellets. The whole wood pellets showed significant differences in emitted particle mass based on two heating conditions. The rubber wood and wood pellets emitted significantly more particles than the rice husks, which indicated that biomass type affects the combustion exhaust particle emissions during “Gradient heating” more than moisture content does.

The cumulative size distributions of collected particles are shown in Fig. 4. The particle size distribution of smoke particles varied depending on both the fuel type and the heating rate. Nanoparticles (<70 nm) and ultrafine particles (0.07–0.43 μm) were present in rates of 20%–30% and 40%–50%, respectively, in an ambient aerosol. Nanoparticles (<100 nm) were estimated by interpolating the distributions to be 30%–50%, indicating an important contribution of nanoparticles from biomass burning.

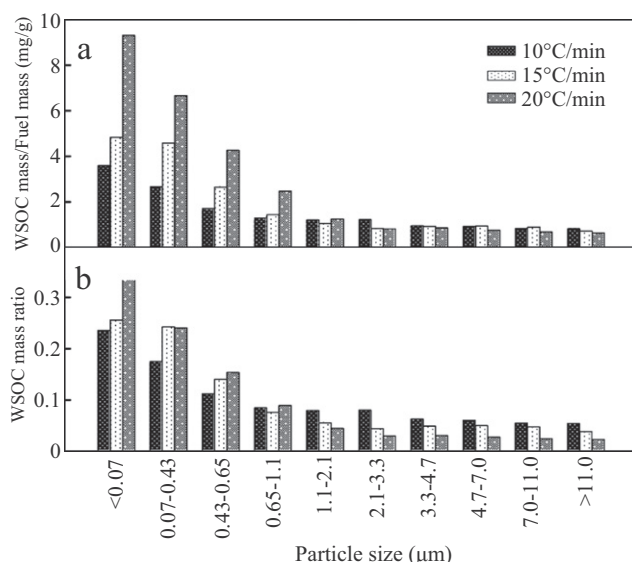
## 2.2. Particle-bound PAHs in emitted smoke

Figs. 5 and 6 show the emitted amounts of 4–6 ring PAHs per unit of fuel mass in nano-particles ( $PM_{0.07}$ ) and their size distributions, respectively. Rubber wood and whole wood pellet combustion had roughly 2 orders of magnitude higher PAH emissions than rice husks. The 4–6 ring PAHs did not simply follow the tendency of particles as indicators of incomplete combustion. The results in Fig. 6 show that the heating rate had a significant effect on PAH emissions, particularly with fine particle in the nano-size range. For all tested conditions, PAH emissions showed a similar trend as particle mass showed the highest emissions in the range of 0.07–0.43 μm, which was 50%–60% for >15 K/min, and 50%–60% for rice husk at 20 K/min. In most cases, nano-particles under 0.07 μm showed the second-highest emissions at 5%–40%. This tendency for size-fractioned PAH distribution was similar to particle size and other results from real biomass combustion seen in residential wood stoves (Tissari et al., 2008).

Fig. 7 shows the amount of PAHs in terms of the BaP toxic equivalent mass per unit fuel mass for each size range of particles measured at different heating rates. The largest Benzo(a)Pyrene Toxic Equivalence Quotient emission was observed at 15 K/min for rubber wood (12.9 ng/g fuel), which was within the range of the largest particle mass, or, 0.07–0.43 μm, and compared with 2.4 ng/fuel-g for rice husks at 20 K/min. Nanoparticles (<100 nm) contribute 16%–22% of total BaP-TEQ emissions, which is about half that of a given particle mass.



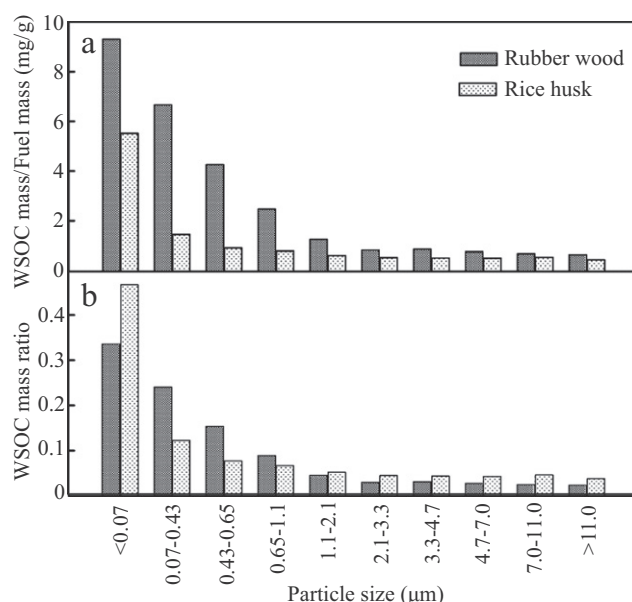
**Fig. 7 – Emissions of BaP-TEQ in each smoke particle size range for different heating rates.**



**Fig. 8 – Emitted amount of WSOC of rubber wood burned relative to heating rates and particle size range. (a) WSOC mass/fuel mass; (b) WSOC mass fraction (mg in size/mg in total).**

### 2.3. Particle-bound WSOC in emitted smoke

The emitted mass of WSOC per unit of fuel mass and mass ratios of WSOC between sized particles is shown in Fig. 8a and b for different heating rates. The WSOC increased as the particle size decreased and particles <70 nm made the maximum contribution of 30%–40% of total emissions: 20%–40% for nanoparticles (<100 nm), although PAHs and particle mass were highest in the 0.07–0.43 μm range. Fig. 9 shows the



**Fig. 9 – Comparison amount of WSOC from rubber wood and rice husks burned relative to particle size ranges: (a) WSOC mass/fuel mass (mg/g); (b) WSOC mass fraction (mg in size/mg of total).**

mass comparison of WSOC per unit of fuel mass and the mass ratios of WSOC among size-classified particles between smoke particles from rubber wood and rice husks. The results indicate that for WSOC, the biomass type had a significant effect on the amount of WSOC, as did the heating rate. The WSOC mass fraction was quite large (ca. 80%) for nanoparticles, suggesting the importance of nanoparticles as an ambient WSOC carrier. The dependence of WSOC on particle size was different from that of PAHs, which showed maximum emissions in the 0.07–0.43 μm range of the maximum mass ratio.

Large specific surface areas and the polarity of soot surfaces are related to ‘solubility’ (Chughtai et al., 1996) and may explain the larger fractions of gaseous WSOC that may be absorbed by nano-particles.

### 3. Conclusions

By sampling and analysis of exhaust gas particles from the laboratory-scale electric tube furnace combustion of solid biomass fuels, the following characteristics of nanoparticles were obtained: (1) more than 30% of smoke particles from the burning of gradient-heated biomass fuel was found to be within the nano-size range (<100 nm) on a mass basis. Particle emission characteristics varied by type of fuel and heating conditions; (2) particles smaller than 0.43 μm made a large contribution to PAHs, and, therefore, to their toxicity and to the fraction of WSOC. These conditions were influenced by the fuel component, combustion conditions and the range of particle size; (3) the largest fraction of particle-bound WSOC was found in the nano-size range, although this size of particle did not constitute the largest fraction of particle mass or particle-bound PAHs.

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### REFERENCES

- Andrea, D.A., 2009. Combustion-formed nanoparticles. *Proc. Combust. Inst.* 32 (1), 593–613.
- Andreae, M.O., Merlet, P., 2001. Emission of trace gases and aerosols from biomass burning. *Global Biogeochem. Cycles* 15 (4), 955–966.
- Signal, K.L., Landridge, S., Zhou, J.L., 2008. Release of polycyclic aromatic hydrocarbons, carbon monoxide and particulate matter from biomass combustion in a wood-fired boiler under varying boiler conditions. *Atmos. Environ.* 42 (39), 8863–8871.
- Chao, C.Y.H., Kwong, P.C.W., Wang, J.H., Cheung, C.W., Kendall, G., 2008. Co-firing coal with rice husk and bamboo and the impact on particulate matters and associated polycyclic aromatic hydrocarbon emissions. *Bioresour. Technol.* 99 (1), 83–93.

- Chomanee, J., Tekasakul, S., Tekasakul, P., Furuuchi, M., Otani, Y., 2009. Effects of moisture content and burning period on concentration of smoke particles and particle-bound polycyclic aromatic hydrocarbons from rubber-wood combustion. *Aerosp. Air Qual. Res.* 9, 404–411.
- Choosong, T., Furuuchi, M., Tekasakul, P., Tekasakul, S., Chomanee, J., Jinno, T., et al., 2007. Working environment in a rubber sheet smoking factory polluted by smoke from biomass fuel burning and health influences to workers. *J. Ecotechnol. Res.* 13 (2), 91–96.
- Chughtai, A.R., Brooks, M.E., Smith, D.M., 1996. Hydration of black carbon. *J. Geophys. Res.* 101 (D14), 19505–19514.
- Furuuchi, M., Tekasakul, P., Murase, T., Otani, Y., Tekasakul, S., Bai, Y., 2006. Characteristics of particulate emitted from rubber-wood burning. *J. Ecotechnol. Res.* 12, 135–139.
- Furuuchi, M., Bai, Y., Tekasakul, P., Otani, Y., Tekasakul, S., Choosong, T., 2007a. Effect of soft X-rays irradiation on PAHs concentration in sized smoke particles from biomass burning. *Proceedings of Asian Aerosol Conference AAC 2007*. Kaohsiung, Taiwan, pp. 116–117.
- Furuuchi, M., Murase, T., Tsukawaki, S., Hang, P., Sieng, S., Hata, M., 2007b. Characteristics of ambient particle-bound polycyclic aromatic hydrocarbons in the Angkor Monument Area of Cambodia. *Aerosp. Air Qual. Res.* 12 (1–2), 221–238.
- Furuuchi, M., Eryu, K., Nagura, M., Hata, M., Kato, T., Tajima, N., et al., 2010. Development and performance evaluation of air sampler with inertial filter for nanoparticle sampling. *Aerosp. Air Qual. Res.* 10 (2), 185–192.
- Hata, M., Fukumoto, M., Otani, Y., Bai, Y., Sekiguchi, K., Tajima, N., et al., 2009a. Performance of new samplers based on “inertial filter” technology applied for classification of ambient aerosol nano-particles. *Proceedings of the 6th Asian Aerosol Conference AAC2009*, Bangkok, AM-P-03.
- Hata, M., Morishita, K., Furuuchi, M., 2009b. Particulate and gaseous pollutants emitted from wood biomass combustion in small scale stoves. *Proceedings of the 6th Asian Aerosol Conference AAC2009*, Bangkok, AP-20.
- Hu, Y.C., Zhou, P.J., Qu, S.S., 2000. TG–DTA studies on wood treated with flame-retardants. *Holz Roh Werkst.* 58 (1–2), 35–38.
- International Agency for Research on Cancer, IARC, 1982. *Monographs, the rubber industry.*, p. 28.
- Jonnalagadda, S.B., Nenzou, B., 1996. Studies on arsenic rich mine dumps: I. Effect on the surface soil. *J. Environ. Sci. Health* 31 (8), 1909–1915.
- Kundu, S., Kawamra, K., Andreae, T.W., Hoffer, A., Andreae, M.O., 2010. Diurnal variation in the water-soluble inorganic ions, organic carbon and isotopic compositions of total carbon and nitrogen in biomass burning aerosols from the LBA-SMOCC campaign in Rondônia, Brazil. *Aerosol Sci.* 41 (1), 118–133.
- Leo, M.L., 2005. Polycyclic aromatic hydrocarbons, 3th ed. In: Audrey, E.M. (Ed.), *Chromatographic Analysis of the Environment*, Vol. 93. CRC Press, Boca Raton London, New York, pp. 555–616.
- Mayol-Bracero, O.L., Guyon, P., Graham, B., Roberts, G., Andreae, M.O., Decesari, S., et al., 2002. Water-soluble organic compounds in biomass burning aerosols over Amazonia 2. Apportionment of the chemical composition and importance of the polyacidic fraction. *J. Geophys. Res. Atmos.* 107 (D20), 8091. <http://dx.doi.org/10.1029/2001JD000522>.
- Oanh, N.T.K., Albina, D.O., Ping, L., Wang, X.K., 2005. Emission of particulate matter and polycyclic aromatic hydrocarbons from select cookstove–fuel systems in Asia. *Biom. Bioenerg.* 28 (6), 579–590.
- Redemann, J., Russell, P.B., Hamill, P., 2001. Dependence of aerosol light absorption and single-scattering albedo on ambient relative humidity for sulfate aerosols with black carbon cores. *J. Geophys. Res.* 106 (D21), 27485–27495 (27).
- Sippula, O., Hokkinen, J., Puustinen, H., Ylipirilä, P., Jokiniemi, J., 2007. Fine particle emissions from biomass and heavy fuel oil combustion without effective filtration (BIOPOR). *VT Working Papers*, p. 72.
- Tang, N., Hattori, T., Taga, R., Igarashi, K., Yang, X.Y., Tamura, K., et al., 2005. Polycyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons in urban air particulates and their relationship to emission sources in the Pan-Japan sea countries. *Atmos. Environ.* 39 (32), 5817–5826.
- Tao, S., 1996. Fractionation and chlorination of organic carbon in water from Yinluan River, Tianjin, China. *GeoJournal* 40, 213–217.
- Tekasakul, P., Furuuchi, M., Tekasakul, S., Chomanee, J., Otani, Y., 2008. Characteristics of pahs in particulates in the atmospheric environment of Hat Yai city, Thailand, and relationship with rubber wood burning in rubber sheet production. *Aerosp. Air Qual. Res.* 8 (3), 265–278.
- Tissari, J., Lyyranen, J., Hytonen, K., Sippula, O., Tapper, U., Frey, A., et al., 2008. Fine particle and gaseous emissions from normal and smouldering wood combustion in a conventional masonry heater. *Atmos. Environ.* 42 (34), 7862–7873.
- Yang, H., Li, Q.F., Yu, J.Z., 2003. Comparison of two methods for the determination of water-soluble organic carbon in atmospheric particle. *Atmos. Environ.* 37 (6), 865–870.



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