SIZE DISTRIBUTION AND CONTRIBUTION OF PARTICLES FROM RICE STRAW OPEN BURNING TO THE ATMOSPHERE IN HANOI

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Abstract. Atmospheric ultrafine, accumulation mode and coarse fractions collected at representative rice straw open burning (RSOB) areas in Ha Noi were investigated to identify characteristics of mass - size distribution and contribution of particles emitted from rice straw (RS) burning to the atmosphere. Two sampling campaigns were conducted from 2018 to 2019. The first one was conducted at Quoc Oai (QO_RS burning site) and Tay Mo (TM_no rice field site). The second one was conducted in two periods at Dong Anh site: RS burning period and non-RS burning period. In the RS burning period, PM_{1.2.5} showed the highest fraction among all collected particles at both sampling campaigns, while PM_{2.5.10} was the most abundant in non-RS burning period and no rice field site. The average mass concentration of PM_{2.5} (particulate matter with an aerodynamic diameter < 2.5 μm) in RS burning period and non-RS burning period were 79.7 ± 46.5 μg m⁻³ and 65.2 ± 21.9 μg m⁻³, respectively at Dong Anh site. Those values were 90.9 ± 33.2 μg m⁻³ at the QO_RS burning site and 71.9 ± 29.3 μg m⁻³ at the TM_no rice field site. The proportion of fine particle (PM_{2.5}) at both sampling campaigns were considerably higher in RS burning period as compared to non-burning period and no rice field site, while the concentration of ultrafine particle (PM_{0.1}) and coarse particle (PM_{10}) were similar between two periods and two sites. This result provides better understanding on size distribution and contribution of fine particles from open RS burning to the atmosphere in Ha Noi, which is an useful information for the environmental managers to control RSOB in Ha Noi as well as in Viet Nam.

Keywords: rice straw open burning, ultrafine particles, fine particles, size distribution, atmosphere.

Classification numbers: 3, 3.4, 3.4.5.

1. INTRODUCTION
Air pollution has recently received much attention due to its alarming level in many parts of the world. Air pollution derived from a wide range of sources including natural source and human activities such as fossil fuel combustion, i.e., natural gas, coal and oil, power industrial processes and motor vehicles [1]. Pollutants in atmospheric environment can be divided into two groups: particles and gases. Among air pollutants, atmospheric particles have been recognized to be important to air quality, health effect and have been studied intensively. Numerous epidemiologic studies have reported increase in human health risk (mortality, morbidity, respiratory and cardio) associated with exposure to atmospheric pollutants [2, 3]. The health effects of airborne particles depend strongly on their size, specific surface area, number, and chemical composition, such as their heavy metal contents, polycyclic aromatic hydrocarbons and their derivatives [4, 5].

Particulate matter (PM) in the atmosphere is a mixture of particles that vary in origin, composition, and size. Larger particles are derived primarily from soil and other crustal materials, meanwhile fine particles are derived chiefly from combustion of fossil fuels in processes such as transportation, manufacturing, power generation and also from biomass burning. The particle size and also chemical components of PM are important for evaluating their influence on air pollution and human health. The smaller particles pose a higher risk to human health. Fine particles (those with aerodynamic diameters equal to or less than 2.5 µm) are serious health concern because they can be penetrated most deeply into human respiratory system and the lung also [2, 4, 6]. In particular, ultrafine particles, also known as nanoparticles (NPs, aerodynamic diameters < 0.1 µm) have a greater tendency to be deposited in the pulmonary alveoli than do fine particles. This is because although the contribution of NPs to mass concentration is low, their contribution to number concentration and surface area is higher than that of larger particles. Therefore, NPs may act as important carriers of toxic compounds into the pulmonary alveoli and more harmful than PM$_{2.5}$ at the same mass concentration [7]. Particle mass concentrations in the atmosphere, indoor air, and industrial environments are regulated by environmental quality standards. Positive correlation between particle mass concentrations and adverse effects on human health have been established by epidemiologic studies [3, 8].

Recently, air quality in Ha Noi has been significantly decreased and has been reported in many information channels including regional and national media. Rice straw burning at field after harvest has been recognized as one of the important sources in agriculture activities which contribute to decreased air quality in Ha Noi. Although Ha Noi is the capital, the center of cultural, economic and political activity of the country, suburban areas around Ha Noi include residential areas intermixed among the agricultural area like Dong Anh, Gia Lam, Thanh Tri, Quoc Oai and thus enhancing rice straw burning impacts in the area. According to Viet Nam Statistics Portal 2018 [9], planted area for paddy rice was 1040.7 thousand ha for Red River Delta and 179.5 thousand ha for Ha Noi. The paddy rice cultivation area was concentrated in suburban areas of Ha Noi. As a result, large amounts of RS are produced after harvesting. Despite the fact that there are many governmental activities to minimize RS burning situation, open RS burning after harvest is still a common practice in Ha Noi as well as in Viet Nam due to the large amount of RS produced. In addition, open RS burning is considered by the farmers as a low cost and simple way to clean the field quickly for next cultivate season. This practice produces a considerable amount of pollutants into the atmosphere, the major compounds being CO$_2$, CO, PM and other organic compounds that can affect to visibility and air quality in Ha Noi [10, 11].
Numerous studies on size distribution and mass concentration of particles in the atmosphere have been conducted [12 - 18]. In Viet Nam, several studies focused on mass concentration of nanoparticles and fine particles, carbonaceous components of nanoparticles at urban locations in Ha Noi [19, 20]. Tran et al. [21] assess the air quality in school environment in Ha Noi focus on the mass-size distribution and found that ultrafine particles accounted for around 15 - 20 % wt/wt while fine particles (PM$_{2.5}$) made up almost 80 % wt/wt of PM$_{10}$. Furthermore, the accumulation mode (0.1 - 1 µm) in ambient air quality accounted for a relatively large percentage of PM$_{10}$, ranging from 33.4 % to 54.2 %. Up to now, the information on mass concentration of atmospheric particles in agriculture biomass as well as RS burning season and effect of RS burning to atmosphere in Ha Noi has been observed [10, 22, 23]. There has been no report on measurement of size distribution and mass concentration of different size particles in RSOB season in Ha Noi. This study was conducted to investigate size distribution of atmospheric particles in RS burning season and examine contribution of particles from RS burning season to the atmospheric environment in Ha Noi. Consideration of the contribution of RSOB to the atmospheric particles helps to better understand the emission characteristics of RSOB regarding to size distribution of particles.

2. MATERIALS AND METHODS

2.1. Sampling sites

Ha Noi is an economic and industrial center of Viet Nam, with 8.5 million inhabitants in the area of about 3,359 square km as of 2019. Being influenced by the Southeast Asia monsoon regime, the climate is tropical and humid. Winter, from November to March, is not so cold (average temperature 16 °C) and relatively dry, while summer, from May to September, is hot (average temperature 33 °C) with plenty of precipitation. The average annually humidity is 79 % and rainfall is 1,800 mm a year.

This work selected two representative sites with RS open burning in sub-urban areas of Ha Noi (Dong Anh and Quoc Oai) and one site far from RS open burning area (Tay Mo) to collect atmospheric particles. Two sampling campaigns were conducted from 2018 to 2020. Detail of sampling locations was described in Figure 1. The first sampling campaign was carried out at two sites: QO_RS burning site and TM_no rice field site. QO_RS burning site was set at Ngoc My, Quoc Oai with the latitude and longitude of 20°59'19"N - 105°37'42"E. This burning site is located within the residential area, which was bounded around by paddy rice fields. TM_no rice field site was set at Tay Mo (latitude and longitude is 20°59'56.8"N 105°44'49.0"E). This site is also located within the residential area, which is far about 13.5 km from QO_RS burning site and without paddy rice fields bounded around like QO_RS burning site. The sampling at these two sites was carried out over 6 consecutive days of RS burning season at the same time (started at 7:00 am on 10/11/2018 and finished at 6:30 am on 16/10/2018) using two samplers parallel. The second sampling campaign was conducted at Dong Anh site (the latitude and longitude of 21°10'13.6"N - 105°53'39.0"E) in two periods (RS burning period and non- RS burning period). This sampling was conducted over 7 consecutive days during RS burning period (from 11 to 18 November 2019) and 7 consecutive days during non-burning period (from 5 to 12 January 2020). The starting time and stopping time for one day were the same with the first sampling campaign. All sampling instruments were placed on the rooftop of a residential building 8 - 11 m above ground level. Results from the survey showed that the proportion of RS subjected to open field burning in summer-autumn crop (harvest from October to November) and winter-spring
crop (harvest in June) in suburban areas of Ha Noi, especially in Dong Anh and Quoc Oai, was about 70 % and 40 % of the total RS production, respectively [5].

Figure 1. Sampling locations in Ha Noi.

2.2. Sample collections and analysis

The airborne particles with different particles size fractions were collected on quartz fiber filters (2500 QAT – UP, 55 mm diameter, Pallflex, CT, USA) using Nanosampler II (Kanomax Japan Inc.) with a design air flow rate of 40.0 L min⁻¹. The Nanosampler consists of four impaction stages that remove particles with cut-off diameters of 10, 2.5, 1.0 and 0.5 μm by impaction onto the quartz fiber filters comprising each impaction stage. After these four impaction stages, particles pass through a cartridge packed with stainless steel fibers to collect particles larger than 0.1 μm. Finally, nanoparticles (< 0.1 μm) are collected uniform on to a 55 mm quartz fiber filter (2500 QAT – UP, Pallflex, CT, USA) in the last stage of Nano sampler. Totally, 6 different size fractions of airborne particles were collected including: larger than 10 μm (PM₁₀), between 10 and 2.5 μm (PM₂.₅₋₁₀), between 2.5 and 1.0 μm (PM₁₋₂.₅), between 1.0 and 0.5 μm (PM₀.₅₋₁), between 0.5 and 0.1 μm (PM₀.₁₋₀.₅), and smaller than 0.1 μm (PM₀.₁). Detail of each stage in Nano sampler was described in Figure 2.
Figure 2. Nano sampler II.

Twenty six of 24-h integrated samples were collected in two sampling campaigns. Flow rates of the sampler and meteorological parameters, including wind direction and velocity, temperature and humidity, were measured during sampling. Before sampling, filters were heated in an oven at 450 °C for 8 hr to remove volatile substances, and then placed in an isothermal box for cooling. The filters were kept in desiccators at room temperature within 48 hr and weighed before and after sampling by 6 digital weigh Mettler Toledo XS205 in Lab at Vietnam National University of Agriculture. Each filter was wrapped in aluminum foil and put in a sealable plastic bag and stored in a refrigerator at – 20 °C until analysis.

3. RESULTS AND DISCUSSION

3.1. Mass concentration of particles in RS burning period

This work identified the mass concentration of 6 various size particles in RS burning period at two sites (Dong Anh site and Quoc Oai site). In general, among 6 particle fractions, PM$_{1.0-2.5}$ was the most dominant in RS burning period at both sites with the average value of 46.3 ± 28.8 μg/m$^3$ and 44.6 ± 20.7 μg/m$^3$ at Dong Anh site and Quoc Oai site, respectively, followed by PM$_{2.5-10}$ and PM$_{0.5}$, The smallest proportion was nanoparticles (PM$_{0.1}$) at DA_RS burning site with the mass concentration of 6.5 ± 2.2 μg/m$^3$ and PM$_{0.1}$ at QO_RS burning site with the average level of 7.7 ± 2.3 μg/m$^3$. The concentration of nanoparticles at QO_RS burning site was 11.5 ± 3.9 μg/m$^3$. Despite its low mass concentration, nanoparticles have been recognized to be more important and harmful than the larger size particles such as PM$_{2.5}$ at the same mass concentration [20, 21]. Considering the mass concentration of fine particles, the mass concentration of PM$_{2.5}$ was 79.7 ± 46.5 μg/m$^3$ and 90.8 ± 33.2 μg/m$^3$ at DA_RS burning site and QO_RS burning site, respectively. In RS burning season, the mass concentration of PM$_{2.5}$ always exceed the Vietnamese standard (QCVN 05-2013) several times and much higher than the limit values recommended by WHO (25 μg/m$^3$) for 24 hours exposure.

Figure 3. Concentration of particles in RS burning period at two sampling sites.

Figure 3a shows the mean values and standard deviations of each particle fraction in each sampling site.
Figure 3b shows the concentration of PM$_{2.5}$ by days at two sampling sites.

The difference of PM concentration by days during sampling campaign associated with the number of RSOB piles, locations of RSOB piles and meteorological factors including wind direction and wind velocity. Wind roses at both sites were showed in Figure 1. The maximum level of PM$_{2.5}$ was observed at DA_RS burning site at the first day of sampling campaign (173 μg/m$^3$), where RSOB happened in most of surrounding fields and the wind direction blow burning smoke directly to the sampling site. This result was the same level with the value reported from the previous research, which was conducted in Ba Vi region [22]. In the second and the third day of sampling campaign at DA site, the rain at night and early morning make PM concentration has reduced dramatically. Even in the rainy day, RSOB still happened on the daytime. The difference of prevailing wind direction in rainy days (N and NNE) and the remain days (SE and SSE) also affect to the measured results. Wind direction and location of RS burning field was difficult to control. The minimum value of PM$_{2.5}$ (22.3 μg/m$^3$) can be seen in the day with the heavy rain at Dong Anh site. Fluctuation of PM concentration by days was observed similarly at QO_RS burning site. Although there are many factors related to the concentration of atmospheric particles, RS open burning may affect to the mass concentration of particles in the local atmosphere in our sampling campaigns.

### 3.2. Variation of particles mass concentration and size distribution between RS burning site and no-rice field site

The first sampling campaign (described in section 2.1) tried to compare the difference of mass concentration and size distribution of particles by space. The mass concentration of individual size particles at QO_RS burning site and TM_no rice field site was presented in Figure 4a. The most dominant particle was PM$_{1.0-2.5}$, followed by PM$_{2.5-10}$ and PM$_{3-10}$ in QO_RS burning site, while the most abundant particle in TM_no rice field site was PM$_{2.5-10}$, followed by PM$_{1.0-2.5}$ and PM$_{3-10}$. In this experiment, the mass concentration of most of size fraction at QO_RS burning site was higher than that at TM_no rice field site, except PM$_{3-10}$, its concentration was not various between two sites. This result suggests that RS burning smoke can emit inhale particles with an aerodynamic diameter less than 10 μm (PM$_{10}$). This particle has been reported that can be penetrated to the human respiratory and lung system. Many previous studies found that fine particles are originated from combustion of fossil fuel such as domestic cooking, transportation, manufacturing, agriculture biomass burning, etc. [4, 6, 24]. This result was appropriate with the fact of two sites. In QO site, RS open burning was observed frequently in harvest season, then it may affect to higher mass concentration of small particles in the atmosphere. TM site with no rice field around is far from QO site might not be affected by RSOB from QO site, although TM site located at downwind of QO site during sampling campaign.

Figure 5 showed the average proportion of each particle size fraction in total collected particles (in percentage) at sampling campaign. The size fraction of particle between 1.0 and 2.5 μm (PM$_{1.0-2.5}$) accounts the major proportions of mass concentration, with average level of 29 % of the total mass in QO_RS burning site. The value of this fraction has dropped to 26 % of the total mass in TM_no rice field site. At the site with no rice field around and far from RSOB, the result presented the dominant particle was PM$_{2.5-10}$ as compared to other particles (27 % of the total mass). The average percentage of PM$_{2.5}$ and PM$_{10}$ of the total mass at QO_RS burning site was 59.7 % and 84.6 %, which was slightly higher than those at TM_no rice field site (54 % and 81 %), respectively.
3.3. Variation of particles mass concentration and size distribution between RS burning period and non-burning period

The mass concentration of individual particles in two periods (burning period and non-burning period) was showed in Figure 4b. Rice harvest season in the north of Viet Nam usually lasts 2 - 3 weeks depending on the maturity of rice. Hence, RSOB happens scattered during harvest season and its affect can last for several weeks to month. Then, we collected particles for non-burning period when RSOB was completely finished. Among six size fractions, the mass concentrations of PM$_{1.0-2.5}$, PM$_{0.5-1.0}$ and PM$_{2.5-10}$ were considerably different between two periods, with higher values in RS burning period than those in non-burning period. The mass concentration of nanoparticles was similar between two periods with the average level of 6.5 ± 2.2 μg/m$^3$ and 6.0 ± 1.8 μg/m$^3$ in RS burning period and non-burning period, respectively. The remain fractions including PM$_{0.1-0.5}$ and PM$_{3.0}$ were almost in the same level between two periods. The higher level of PM$_{1.0-2.5}$, PM$_{0.5-1.0}$ and PM$_{2.5-10}$ fraction implicated the higher level of total atmospheric particles in RS burning period. In totally, the average concentration of PM$_{2.5}$ in RS burning period increased by 22% as compared to that in non-burning period at sampling campaign. Similar trends on the mass size distribution of particles were observed in two sampling campaigns (Figure 4a and Figure 4b).

As the same profile in the first sampling campaign, the fraction of PM$_{1.0-2.5}$ occupies the major proportions of mass concentration, with average level of 35% of the total mass (Figure 5). The value of this fraction has decreased to 29% in DA_non-burning period. Without RS burning, the particle fraction between 2.5 and 10 μm (PM$_{2.5-10}$) was dominant among 6 fractions. The average percentage of PM$_{2.5}$ and PM$_{10}$ of the total mass in RS burning period was 60% and 90%, which was slightly higher than those in non-burning period (56% and 87%), respectively. Higher contribution of fine particles from RS open burning in harvest season suggests that open burning of RS can be considered as one of significant emission sources to Ha Noi atmosphere. The result was appropriate with the previous studies that emissions from the burning agriculture crop residues were dominated by fine particles [16]. Keshtkar and Ashbaugh [25] has been found a bimodal mode dize distribution for rice straw burning, one peak in the accumulation mode (0.1 μm < d$_{ae}$ < 1.8 μm, in which d$_{ae}$ was aerodynamic diameter) and another peak in the coarse mode (d$_{ae}$ > 1.8 μm).
There were no significant differences in percentage of the smallest fraction (PM$_{0.1}$) between the two periods and two sites, accounting for around 5 - 7 % of the total mass. Particle formation in the nucleation mode (< 0.1 μm) has been documented for aerosols from vehicle emission, coal combustion and wood burning [26]. Venecek et al. [27] found that on-road gasoline and diesel vehicles made significant contributions to regional PM$_{0.1}$ in all 39 cities in United States. Nghiem et al. [28] also found that traffic contributed the biggest part of nanoparticles (46.28 % of five major emission sources to the PM$_{0.1}$) in Ha Noi. In this study, two sampling sites located in rural areas and far about 300 - 500 m from the inter-communal road with sparse traffic. Low transportation density and regular fuel burning in domestic activities was negligible in the experiments. Furthermore, there were no special events during sampling campaigns, the noticeable difference was RS burning activity. Then the stabilization of nano particle concentration in the current experiments suggests PM$_{0.1}$ was not important emission source from RS open burning in our case. For inhaled particulates matter, the contribution of PM$_{10}$ to the total mass varied from 81 - 90 %, in which, PM$_{2.5}$ contributed from 54 to 60 % of the total mass. Taking into consideration the proportion of PM$_{2.5}$ to PM$_{10}$, the results showed that PM$_{2.5}$ accounted for about 64 - 70 %, which is smaller than the values reported from Tran et al. [21] (about 80 %), where the research investigated indoor and outdoor particles in school environment. The higher proportion of PM$_{2.5}$ to PM$_{10}$ in the previous study related to higher contribution of ultrafine particles from traffic as a significant emission source. In this study, RS open burning contributed to the level of PM$_{1.2.5}$ fraction as the predominant particles. The contribution of this fraction in RS burning period leads to the higher level of PM$_{2.5}$ as compared to that in non-burning period. Higher level of PM$_{2.5}$ imply a significant threat to the human health. Hence, it is necessary to control RS open burning after harvest to reduce its impact to the local and regional air quality and also improve visibility in the area.

![Figure 5. Average size distribution of particles in two sampling campaigns.](image)

### 4. CONCLUSIONS

The work presented the mass concentration and size distribution of particles at two representative RS burning areas in Ha Noi. Two sampling campaigns was carried out to compare the variation of size distribution and mass concentration of particles between two periods (during
RS burning period and after RS burning period) and between two sites (within RS burning area and far from RS burning area). The results showed that, the concentration of PM_{2.5} in most sampling days exceeded the limit values recommended by Vietnamese standard and was much higher than the limit values by WHO for 24-h exposure. In general, among 6 particle fractions, PM_{1.0-2.5} was the most dominant fraction in RS burning period at both sites, while the most abundant particle in non-burning period and at TM_no rice field site was PM_{2.5-10}. The average proportion of fraction PM_{1.0-2.5} accounts about 29 - 35% and the average percentage of PM_{2.5} was 60 - 90% of the total mass in RS burning period. These values were slightly higher in RS burning period as compared to that in non-burning period. The mass concentration and proportion of PM_{0.1} and PM_{10} was not so much different between two periods and two sites. PM_{0.1} concentration accounted for around 5 - 7 % of the total mass. This result suggests that, nanoparticles is not so considerable fraction with high mass concentration in RS burning season, but PM_{1.0-2.5} was the most dominant fraction on mass concentration. This fact leads to the higher contribution of PM_{2.5} level from RS burning season to the local atmosphere and reduce air quality of the regional area. This result provides the insight into the Ha Noi air quality in term of size distribution and mass concentration of particles, which affected by RS open burning in the harvest season.

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Size distribution and contribution of particles from rice straw open burning to the atmosphere in HN


