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Research Article

Chemical Composition and Size Distribution of Aerosol Particle in High Polluted Periods

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ABSTRACT In this study, PM_{10} and $PM_{2.5}$ mass concentrations were investigated in Busan Metropolitan City on high pollution episode periods. PM_{10} and $PM_{2.5}$ were collected from December 2013 to January 2015. The aerosol samples were collected during Asian dust season and high polluted periods in winter respectively. The collected samples were analyzed by some techniques such as comparison of mass concentration, correlation analysis, chemical composition and particle size distribution. Mass concentration of PM_{10} in spring season ranged from 62.6 μg/m^3 to 131.5 μg/m^3. High PM_{10} and $PM_{2.5}$ concentration on spring were contributed mainly by Asian-Dust. In wintertime, PM_{10} and $PM_{2.5}$ high concentrations were almost contributed by anthropogenic source such as house heating and stagnant meteorological conditions. But, $PM_{2.5}$ mass concentration did not show any difference according to sampling sites. SEM-EDS results showed that high polluted periods have different chemical composition elements such as Al, Ca, Mo, Zn in spring and Cu, Cr, Mn, Ti, Zr in winter. Particle number distribution on Asian dust season showed that particle counts were increased to the factor of 2 in the size of 5 μm , the size of 3 μm was increased to the factor of 2, the size of 1 μm was increased the factor of 1.6 respectively. On the other hands, particle counts in the size of 1 μm and 0.5 μm were decreased to the factor of 1.6 and 0.3 μm was considerably decreased to the factor of 3.5. Finally, it is possible to establish the proper PM pollution control strategy to evaluate levels of high PM_{10} and $PM_{2.5}$ pollution periods, chemical composition and size distribution by detail analysis.

KEY WORDS Mass concentration, Chemical composition, Particle counts, Particle size distribution, High polluted Periods, PM_{10} and $PM_{2.5}$

1. INTRODUCTION

In general, high concentration pollutions for PM_{10} and $PM_{2.5}$ appeared mainly from wintertime to next year spring for every year. The high concentration for PM pollution lasted one or two days and above more than two days when it was occurred. WHO warned that $PM_{2.5}$ could affect man's both lungs and heart including premature death, increased respiratory symptoms, aggravated bronchial asthma. WHO technical report revealed that ambient air $PM_{2.5}$ policies affect 60–80% of the urban population exposure $PM_{2.5}$ and outdoor air is responsible

for 40–70% of total of the total population exposure to that (WHO, 2013). Recently, Ministry of Korea Government revised annual standard of $PM_{2.5}$ to its concentration from 25 to 15 μg/m^3 on March 2019. But, most of citizens experienced that $PM_{2.5}$ pollution is more severe before revising annual standard. This is the reason that the reduction strategies for $PM_{2.5}$ have focused on average annual concentration. But, average concentration for $PM_{2.5}$ and its frequency were increased in high polluted period by year and year. The previous work reported that the frequency of the PM_{10} concentration over 50 μg/m^3 has decreased while that less than 20 μg/m^3 has changed little (Kim and Yeo, 2019). It was concluded that the frequency between 20 and 50 μg/m^3 has increased. Other research showed that source contribution of $PM_{2.5}$ in Republic of Korea was investigated during KORUS-AQ campaign (Choi *et al.*, 2019). This research suggested that Chinese contributions are up to ~68% of $PM_{2.5}$ in surface air in South Korea. Park and Kim (2016) reported characteristics of water-soluble inorganic species in PM_{10} and $PM_{2.5}$ at two coastal sites during spring in Rep. of Korea. It was founded that ion concentrations of SO_4^{2-}, NH_4^+ and NO_3^- in both PM_{10} and $PM_{2.5}$ are high about two times.

The objective of this study was to evaluate PM_{10} and $PM_{2.5}$ mass concentration comparison in high polluted periods at Busan Metropolitan City. It is important to know the range of mass concentration of PM_{10} and $PM_{2.5}$ in high polluted periods to establish control strategy. Also, chemical composition by SEM-EDS to identify aerosol sources for PM_{10} and $PM_{2.5}$ in high polluted periods were reviewed. This study presents particle size distribution which was investigated by LPC counts in same periods. The data show particle number counts change which was observed in a special size range of high polluted period.

2. EXPERIMENTAL

2. 1 Experimental Method and Periods

In this experiment, Fig. 1 shows the sampling point which is located at 5 floor of Donga University at Sahagu. This university was surrounded by small mountains with maximum height of 497 m. In the west side of the point, Nak-Dong River flows from north to south side of sea. In the north side, about 4–5 km of the sampling point, Sasang Industrial Complex is located. This industrial complex has iron manufacture, nonferrous

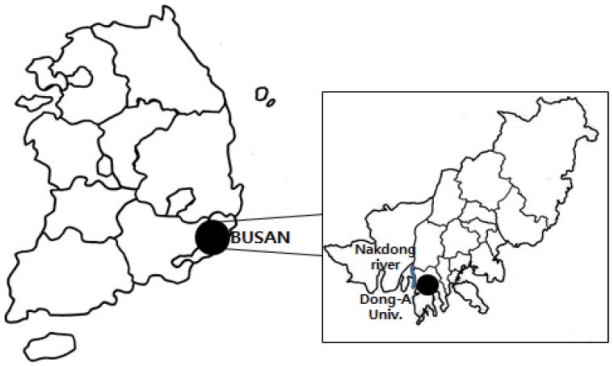

Fig. 1. Location of the sampling point.

Table 1. Aerosol sampling conditions.

Sampling site	Busan, Donga Univ. N35.116 E128.968	
Sampling period	Dec. 2013~Feb. 2015	
Sampling intervals	Depends on weather condition	
Sampling number	PM_{10}	$PM_{2.5}$
	20	20

manufacture, parts-component industry. In wintertime, this sampling point is strongly impacted by these industrial sources. In contrast to summertime, sea-salt concentration is high because of impact by southeast wind direction. In this study, sampling period was planned to 2 years from Dec. 2013 to Feb. 2015. Sampling condition was controlled by meteorological conditions such as high concentration pollution period and Asian dust season. Table 1 indicates aerosol sampling conditions for PM_{10} and $PM_{2.5}$.

2. 2 Aerosol Sampling Method

High concentration PM_{10} and $PM_{2.5}$ samples were collected by URG cyclone sampler (URG-2000-30 ENB PM_{10}, URG-2000-30EH $PM_{2.5}$) individually. This URG cyclone sampler consist of 3 stage filter pack and sampling flow late is 17.6 L/min. Nuclepore filter (Whatman Φ47 mm, pore size 0.2 μm) was used for collecting PM_{10} and $PM_{2.5}$ samples. This nuclepore filter is suitable for aerosol sampling because of hydrophilic and low cohesiveness with protein. Sampling time was set to 4 hours intervals basically, the time was controlled by ambient aerosol concentration. The collected aerosol samples were saved and dehumidified in desiccator on 24 hours and weighted its mass by digital electronic balance (M2P, SATORIUS Co.). Particle

Table 2. Specification of SEM-EDS.

Model	SEM	EDS
	JSM-6700F	OXFORD instrument 7421
Production company	JEOL (JAPAN)	OXFORD (U.K)
Resolution	1.0 nm (at 15 kV), 2.2 nm (at 1 kV)	30 mm,138 eV at Mn Ka
Magnification	25 to 19,000 (LM mode), 100 to 650,000 (SEM mode)	
Accelerating voltage	0.5 to 30 kV	
Electron gun	Cold cathode field emission type	
Alignment	Electromagnetic deflection system	
Objective lens	Strongly excited conical lens	
Specimen chamber	Large chamber for 200 mm specimen	
Light element detector resolution		Crystal: Silicon Window: Super ATW Detectable element: 5B to 92U 7.5 liter liquid nitrogen dewater

counts were monitored by Lase Particle Counter (LPC, TF500, KANOMAX Co.). This laser particle counter is possible to measure particle counts at the size of 5 range (0.3 μm, 0.5 μm, 1.0 μm, 3.0 μm, 5.0 μm). As the particle pass through the laser light, the particle blocks the laser light scattering and loss of light is detected by a photo detector if light scattering is used.

2. 3 Sample Analysis

It is important to know aerosol chemical composition to identify emission sources. It was used to identify emission source by PM_{10} and $PM_{2.5}$ chemical composition data which were determined by SEM-EDS. SEM-EDS is one of the excellent analysis tools for physical and chemical analysis of multiple aerosol samples to detect trace elements economically. Aerosol samples were analyzed by FE-SEM for image data and obtained chemical composition data by EDS (Energy Disperse X-ray Spectrometer). Test sample was coated with Pt by vacuum evaporation through cutting the size of 5 mm × 5 mm. Table 2 summarized FE-SEM analysis conditions, individual aerosol particle was observed by SEM with microscopic magnification of 500 to 10000.

3. RESULTS AND DISCUSSION

3. 1 Comparison of High Polluted PM_{10} and $PM_{2.5}$ Concentration Data

In this study, high concentrations were established at 80 μg/m^3 (PM_{10}) and 50 μg/m^3 ($PM_{2.5}$) individually. The reason is that good level of PM_{10} was ranged the concentration from 30 μg/m^3 to 80 μg/m^3. But severe level of PM_{10} starts with the concentration from 80 μg/m^3 by Korea Ministry of Environment 6 stage action guideline. But there is no guideline about $PM_{2.5}$ on severe stage. Therefore, 50 μg/m^3 for 24 hr average concentration (2015 standard) was used for the severe level for high concentration of $PM_{2.5}$. Fig. 2 shows PM_{10} and $PM_{2.5}$ mass concentration on high polluted periods in spring and winter, which were sampled at the point of this study and 4 sites in Busan air monitoring stations separately. Aerosol samples in high polluted periods were collected from March to May 2014 in spring season, and sampled two times from Dec. 2013 to Feb. 2014 firstly, from Jan. 2015 to Feb. 2015 secondly in winter. In this study, PM_{10} mass concentration in high polluted season of spring ranged from 62.6 μg/m^3 to 131.5 μg/m^3, average concentration was 80.4 μg/m^3. The data from other 4 sites in Busan air monitoring stations showed PM_{10} average concentration were ranged from 134.9 μg/m^3 to 153.5 μg/m^3, and indicated 1.67–1.9 times higher than the data of this study. This is contributed by the site of this study is located on the 150 m high from the ground level of traffic road. Because of coarse aerosol particles are reduced by its count concentrations to compare with roadside. $PM_{2.5}$ mass concentration in high polluted season of spring ranged from 45.8 μg/m^3 to 127.8 μg/m^3, and average concentration was 75.2 μg/m^3. The data from other 4 sites in Busan air monitoring stations show $PM_{2.5}$ average concentration were ranged from 72.2 μg/m^3 to 75.7 μg/m^3, $PM_{2.5}$ mass concentration were not showed any difference of their concentrations to compare with $PM_{2.5}$ data with other sites.

In contrast to winter, PM_{10} mass concentration ranged

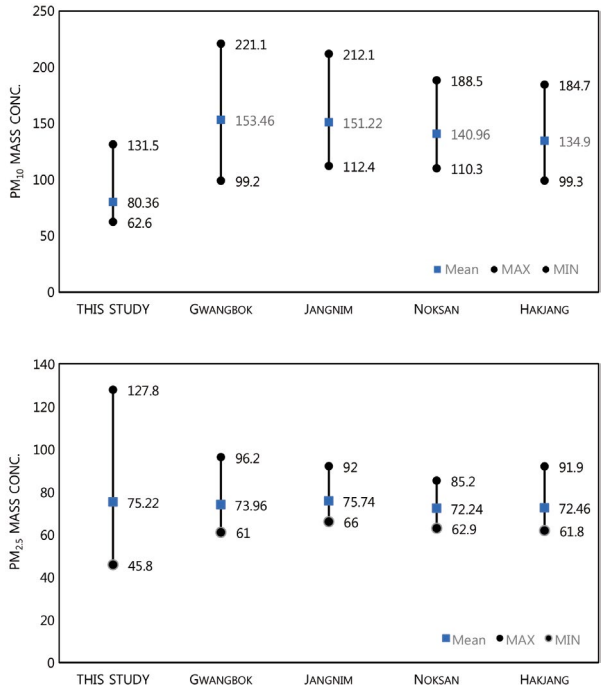

Fig. 2. PM_{10} and $PM_{2.5}$ mass concentration on high polluted season at 5 points (spring).

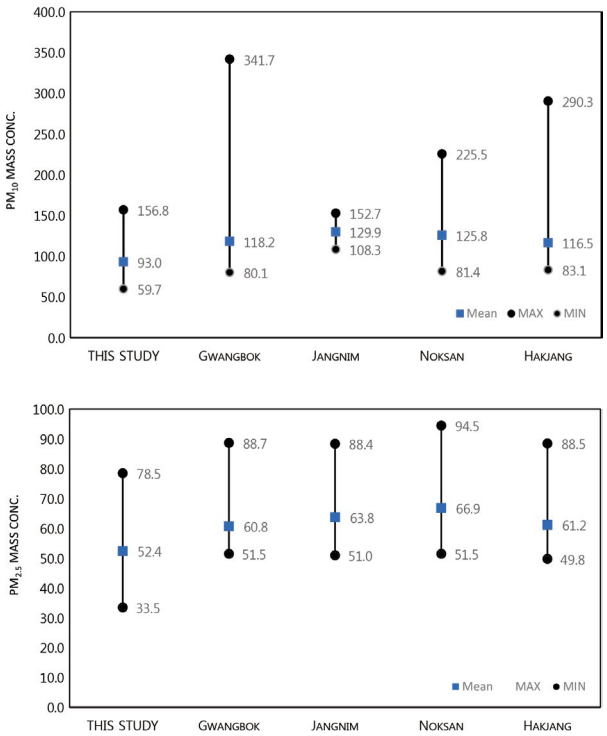

Fig. 3. PM_{10} and $PM_{2.5}$ mass concentration on high polluted season at 5 points (winter).

from 59.7 μg/m^3 to 156.8 μg/m^3 of this study, average concentration was 93.0 μg/m^3. Other monitoring sites data showed PM_{10} mass concentration in high polluted season of winter ranged from 116.5 μg/m^3 to 129.9 μg/m^3. In this study, $PM_{2.5}$ mass concentration in high polluted season of winter ranged from 33.5 μg/m^3 to 78.5 μg/m^3, and average concentration was 52.4 μg/m^3, other monitoring sites data showed $PM_{2.5}$ mass concentration in high polluted season of winter ranged from 60.8 μg/m^3 to 66.9 μg/m^3. $PM_{2.5}$ did not show any difference for mass concentration according to sampling sites. This result was contributed by meteorological factors of no winds and stable condition at all sites of the city in winter.

3.2 Chemical Composition on High Polluted PM_{10} and $PM_{2.5}$ Concentrations

Chemical composition data are very useful tools to source identification. To determine chemical elements, SEM-EDS analysis was used for determining in collected aerosol samples. SEM-EDS analysis was conducted to PM_{10} and $PM_{2.5}$ about 30 samples, the chemical composition data was obtained 10 times in one filter repeatedly by scatter electron beam radiation. Target elements for SEM-EDS analysis are 22 elements like as Al, Br, Ca, Cl, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, P, Pb, Si, Ti, V, Zn, Zr. Fig. 4 indicates high atomic (%) elements in order such as Si, Fe, Ca, Cl, Al, Mg, Na, K in spring, Si was higher 2 times than winter data in specially. This result can be explained that the Si, Fe, Ca were almost included in Asian dust in spring, and combined with other elements Cl, Mg, Na which were generated from sea-salt particles. Fig. 5 shows the different type elements pattern of Fe, Zn, Ti, Cu, S, Si, these elements mainly come from fossil fuel incineration facility in winter. Sulfur is good indicator of fossil fuel incineration. SO_2 can be emitted from incineration sources and converted to sulfate in fine particles. Hsu *et al.* (2016) reported that health-related particulate metals for PM_{10} and $PM_{2.5}$ were related to the traffic emission and coal combustion sources in winter. In Beijing, elevated concentrations of elements and water-soluble inorganic ions were founded on haze days for both PM_{10} and $PM_{2.5}$ (Gao *et al.*, 2015). He *et al.* (2015) reported impacts of biomass-burning on aerosol properties of a severe haze event over Shanghai to Korean Peninsula.

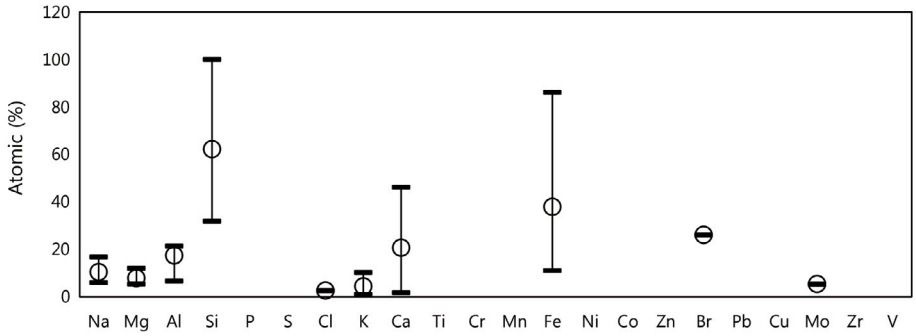

Fig. 4. Result of elemental composition by SEM-EDS in spring.

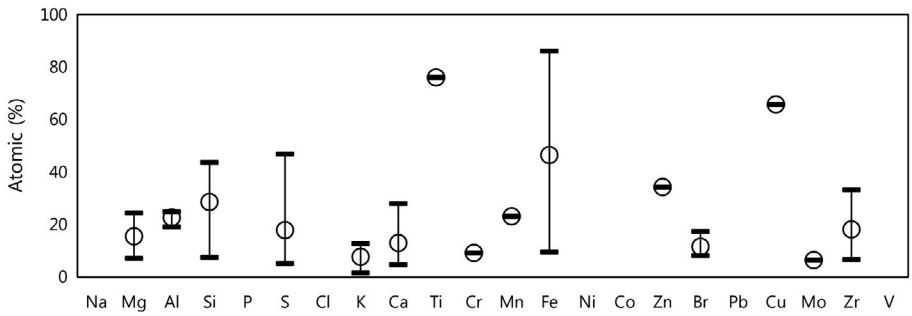

Fig. 5. Result of elemental composition by SEM-EDS in winter.

3.3 Particle Size Distribution of High Polluted Periods

Fig. 6 indicates particle counts measured by LPC collected on Asian dust period and non-Asian dust period. Particle counts were increased in size over than 3 μm and 5 μm. The particle counts (3 μm) were measured to 251, 224, 226, 279 on non-Asian dust period, it was increased to 533 about two times on Asian dust period. Also, the particle counts (5 μm) were measured to 73, 71, 76, 82 on non-Asian dust period, it was increased to 167 about two times on Asian dust period. Ma and Choi (2007) reported the number concentrations were increased in the size range of $PM_{2.5\text{-}5.0}$ and $PM_{5.0}$ during Asian Dust Period in April 2005. These results can be explained that Asian dust carried massive desert sands larger than 1 μm to sampling point by winds. This tendency was found inversely at the particle size below 1 μm. In particle size of 0.3 μm, the particle counts were measured to 219338, 346129, 410075, 346192 at non-Asian dust period. And the particle counts were decreased to 94754 on Asian dust period. In similar, the size of 0.5 μm, particle counts were measured to 15633, 16809, 24830, 16734, it was decreased to 11321 at Asian dust period. Jeon *et al.* (2000) reported particle counts measured by OPC also decreased too much in the range of 0.3 μm and 0.5 μm on Asian dust period. Kim *et al.* (2008) reported that some elements including cadmium showed its fine-fraction counterpart a contrary pattern with more enhanced values during Non-Asian Dust (2.60 ± 1.64 ng m^{-3}) than Asian Dust (1.60 ± 0.65 ng m^{-3}). Park and Kim (2006) also reported particle size shifted towards larger particles compared to smaller particles during the Asian dust period. It was supposed that particles size smaller than 1 μm can adhere to large particles through transportation from Asian deserts. In general, in the range of particles size below 0.1 μm, the gaseous pollutants and fine particles were conversed to new particles by homogeneous and heterogeneous reactions. It was assumed that the reaction did not occur that the AD event was not the main route of the Cd transport into Korea, or slowly progressed because of not too much gaseous pollutants and fine particles, this result showed that 50–70% reduction of total particles counts in this range.

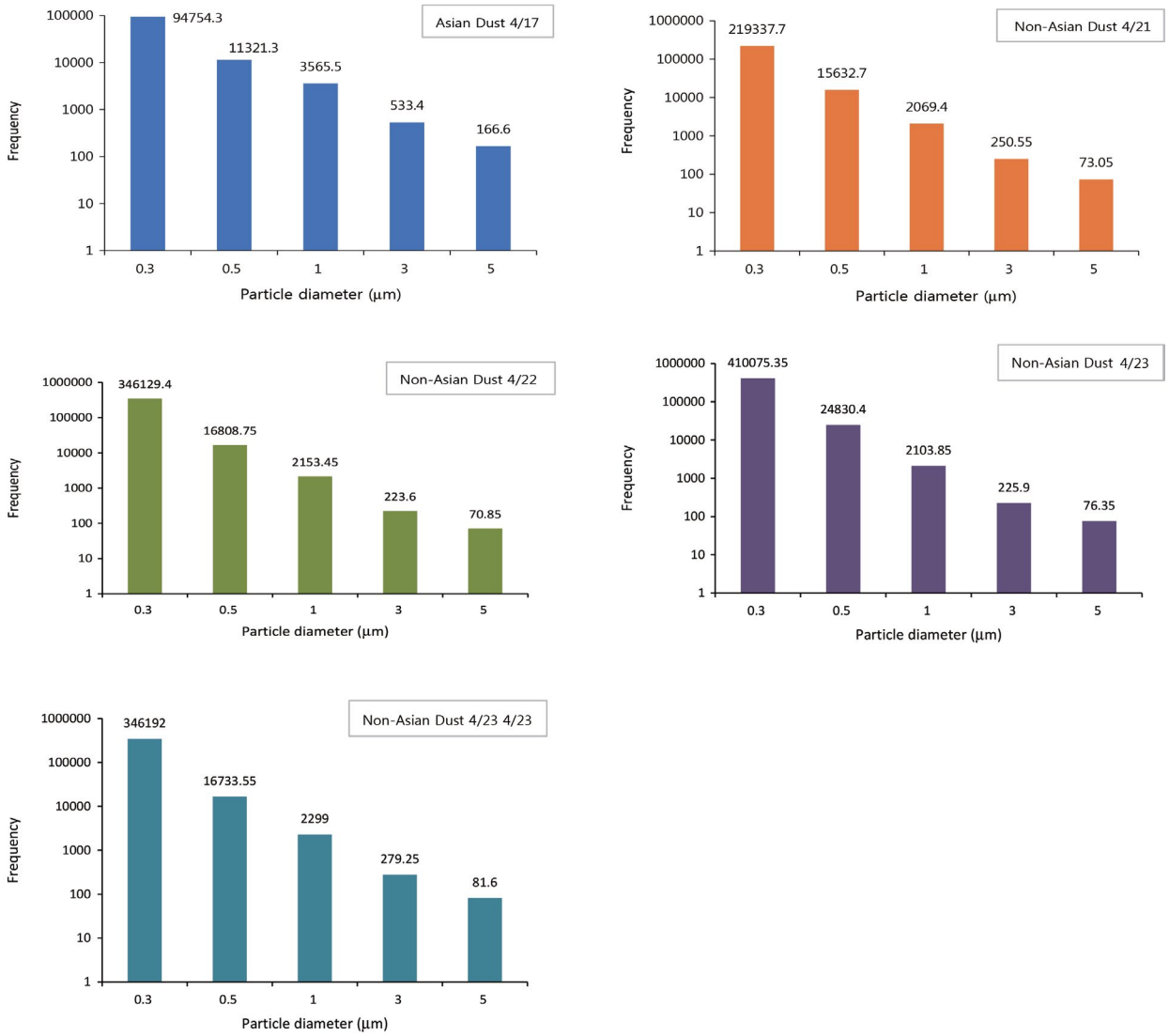

Fig. 6. Particle Counts by LPC collected on Asian dust period and non-Asian dust period.

4. CONCLUSIONS

We have evaluated mass concentration, chemical composition and size distribution of aerosol particles in high polluted periods of spring and winter. In comparison with PM_{10} and $PM_{2.5}$ mass concentration data, Asian dust increased both PM_{10} and $PM_{2.5}$ mass concentration in high polluted periods of spring. But, $PM_{2.5}$ did not show any difference for mass concentration according to sampling sites. This result was caused by meteorological factors of no winds and stable condition at all sampling sites of the city in winter. SEM-EDS analysis was used to determine metal elements in collected PM_{10} and $PM_{2.5}$ samples. Chemical composition data shows that crustal elements such as Si, Fe, Ca, Cl, Al, Mg, Na, K were abundant in aerosol samples, and which were originated from Asian dust in spring. The data shows that the different type elements pattern of Fe, Zn, Ti, Cu, S, Si, these elements mainly come from fossil fuel incineration facility in winter. Particle counts were increased in the size of 3 μm and 5 μm in Asian dust periods. Adversely, particle counts in the size less than 1 μm (0.3 μm, 0.5 μm) decreased too much in same periods. It seemed that particles size smaller than 1 μm can adhere to large particles by coagulation effects through transportation from Asian deserts.

It is important to consider particle size distribution by measured particle counts in high polluted periods of PM_{10} and $PM_{2.5}$. Because of adverse health effects of fine particles, it is a good index to check particle counts in high polluted PM_{10} and $PM_{2.5}$ periods for increasing of particle counts in size less than 1 μm.

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