

Characteristics of Atmospheric Particulate Matter and Metals in Industrial Sites in Korea

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Abstract

The distribution of metals in atmospheric particulates less than 10 μm was studied at a petrochemical refinery site and at a non-ferrous heavy metals industrial site in the city of Ulsan, South Korea in both the summer and fall seasons. The samples were collected with a high volume sampling system equipped with a 9 stage cascade impactor, which effectively separated the particulate matter into 9 size ranges. Total PM₁₀ was $59 \pm 14 \mu\text{g}/\text{m}^3$ in summer and $56 \pm 18 \mu\text{g}/\text{m}^3$ in fall at the petrochemical site whereas it was $52 \pm 14 \mu\text{g}/\text{m}^3$ in summer and $88 \pm 36 \mu\text{g}/\text{m}^3$ in fall at the non-ferrous heavy metals site. The particle size fractionation in less than 10 μm showed a typical bimodal distribution, with one peak corresponding to the particle size range of 1.1-4.7 μm and the other to the range of 4.7-10 μm . Five heavy metals (Ni, Cu, Zn, Pb, and Cd) were measured in the composite mixture of particulates (0.1-1.1, 1.1-4.7 and 4.7-10 μm). The heavy metals concentrations were found to be higher in the 1.1-4.7 μm fraction followed by 4.7-10 and 0.1-1.1 μm . Among the metals Pb showed particle size dependent whereas Zn was homogeneously mixed in all sizes. The obtained data are important for an estimation of level pollution with heavy metals in industrial sites.

Keywords: atmospheric particulate, heavy metals, seasonal variation

1. Introduction

Urban areas often produce atmospheric particulate that lead to numerous health hazards to both humans (Bünger, Krahl, Schröder, Schmidt, & Westphal, 2012; Dockery & Pope, 1994; Olsson & Benner, 1999) and ecosystems (Bell, Samet, & Dominici, 2004a, 2004b; Schwartz, Dockery, & Neas, 1996). Of these hazardous pollutants, particulate matter (PM) with aerodynamic diameter less than or equal to 10 μm (PM₁₀) are particularly important as they can carry many toxic elements within a complex mixture of anthropogenic and naturally occurring airborne particles. They are formed by two basic mechanisms: dispersion and condensation, including chemical reactions. The atmospheric particulates are a mixture of primary and secondary aerosols and usually have bimodal mass distributions (Bashurova, Koutzenogil, Pusep, & Shokhirev, 1991; Karanasiou, Sitaras, Siskos, & Eleftheriadis, 2007; Spurny, 1996). The last decade has seen a plethora of research documenting relations between particulate matter and respiratory diseases, cardiovascular mortality, morbidity and malignant lung diseases (Bräuner et al., 2007; Liu, Ying, Harkema, Sun, & Rajagopalan, 2013; Ostro, Broadwin, Green, Feng, & Lipsett, 2006; Risom, Møller, & Loft, 2005; Simkhovich, Kleinman, & Kloner, 2008). An increase in PM₁₀ by a concentration change of 10 $\mu\text{g}/\text{m}^3$ can yield 1% increase in overall mortality and 3-6% increase in deaths associated with respiratory disease (Kan, Wong, Vichit-Vadakan, & Qian, 2010; Ostro, Chestnut, Vichit-Vadakan, & Laixuthai, 1999; Ostro, Hurley, & Lipsett, 1999).

Metals are commonly found in atmospheric particles. While they can be present in almost all sizes of atmospheric particulate, in general, fine particulate carries higher concentrations of toxic metals than coarse particulate (Fang & Huang, 2011; Hieu & Lee, 2010). Metals associated with respirable particles have been shown to increase numerous diseases (Hu et al., 2012; Pandey et al., 2013). Metals in the urban atmosphere are frequently associated with specific pollutant sources, and these are often used as tracers in order to identify the source of atmospheric particulate (Chen et al., 2013; Duan & Tan, 2013; Wang, Bi, Wu, Zhang, & Feng, 2013;

Weckwerth, 2001).

The determination of particle sizes in total PM₁₀ and the metals associated with those particles may provide an insight into health hazards for different particle sizes. The coarser particles (aerodynamic diameter > 3 µm) are thought to be formed by high temperature combustion, crustal erosion and road dust resuspension. The finer fractions are believed to be principally emitted from anthropogenic sources including combustion, high-temperature industrial activities and automotive traffic (Dockery & Pope, 1994; Toscano, Moret, Gambaro, Barbante, & Capodaglio, 2011). Knowledge on the size of the particles and their relationship to metals is vital in understanding health hazards in the atmosphere and to develop efficient methods and technologies to tackle these problems.

Numerous studies have been undertaken to estimate PM₁₀ and its associated heavy metals in the atmosphere surrounding industrial areas across several continents (Chen, 2007; Lim, Lee, Moon, Chung, & Kim, 2010; Ochsenkühn & Ochsenkühn-Petropoulou, 2008; Razos & Christides, 2010). The dispersion and accumulation of particulate matter in any location is mainly affected by the existing sources, meteorological conditions and local topography. The local wind circulations may change the particulate matter's dispersion and accumulation within a very short distance and this in turn affects pollution levels in a small area from time to time. The city of Ulsan is one of the most industrial regions in South Korea. Previous studies have been undertaken to understand the particulate matter in residential areas in Ulsan (Hieu & Lee, 2010; Lee & Hieu, 2011). However, atmospheric particulate dispersion in industrial zones has received little attention.

The aim of this study was to investigate the atmospheric particulate and associated heavy metals in the summer and fall seasons at a petrochemical (PC) industry and a non-ferrous industry (NFI) sites. In order to study the differences observed in the relevant size distributions, cluster analysis was performed on the aerosol data. The examination of the relationship between the different particle sizes and metals in particles will help in identifying the formation, dispersion and accumulation of particles and metals in two seasons: summer and fall.

2. Method

2.1 Sample Collection

The two sites (PC and NFI) are located in an industrial zone in Ulsan, South Korea (Figure 1). Site PC is approximately 2 km north-west of site NFI. A total of 10 samples were collected from each site for each of two seasons (summer and fall) using a 9 stage cascade impactor (Environmental Tisch, USA). The samples were collected from the roofs of the industrial buildings, 21 m above the street level during summer and fall (2007). The impactors contained 9 filters (0-0.4, 0.4-0.7, 0.7-1.1, 1.1-2.1, 2.1-3.3, 3.3-4.7, 4.7-5.8, 5.8-9.0, and 9.0-10 µm) and were exposed to the atmosphere for 24 h to retrieve daily average PM samples. The samples collected on the filters were kept in desiccators for 48 h at laboratory temperature (22 °C) to minimize the effect of moisture on the filters before and after taking air samples. A gravimetric analysis was performed using a 5-digit microbalance (Mettler, Toledo) to measure the collected mass in each fraction. Meteorological data (temperature, wind speed, wind direction, humidity, cloud cover and precipitation) were collected from the Ulsan Metropolitan Government.

Flow rates were measured in the field using a manometer which sensed the pressure drop across the 9 stages of the impactor. The manometer was calibrated in the laboratory using a positive displacement flow rate meter. In the field, the flow rate was set at an initial value of 28.3 m³/h. At the end of the measurement (24 h), the corresponding value was found to be 28.2 m³/h, that corresponded to a deviation of less than 0.35%.

Quantification of the particles collected in cascade impactor for the nine fractions were based on the theoretical impaction curve diagrams of each stage of the ambient cascade impactor provided by the impactor manufacturer. Detailed PM mass fraction calculation processes are mentioned in Table 1. The cascade measurement was verified by the β-ray attenuation method at the same sampling site, measured by Ulsan Network of Air Pollution Monitoring (UNAPM). The PM₁₀ concentrations obtained in this study using the cascade impactor were significantly correlated ($R^2 = 0.88$) with the PM₁₀ concentrations measured.

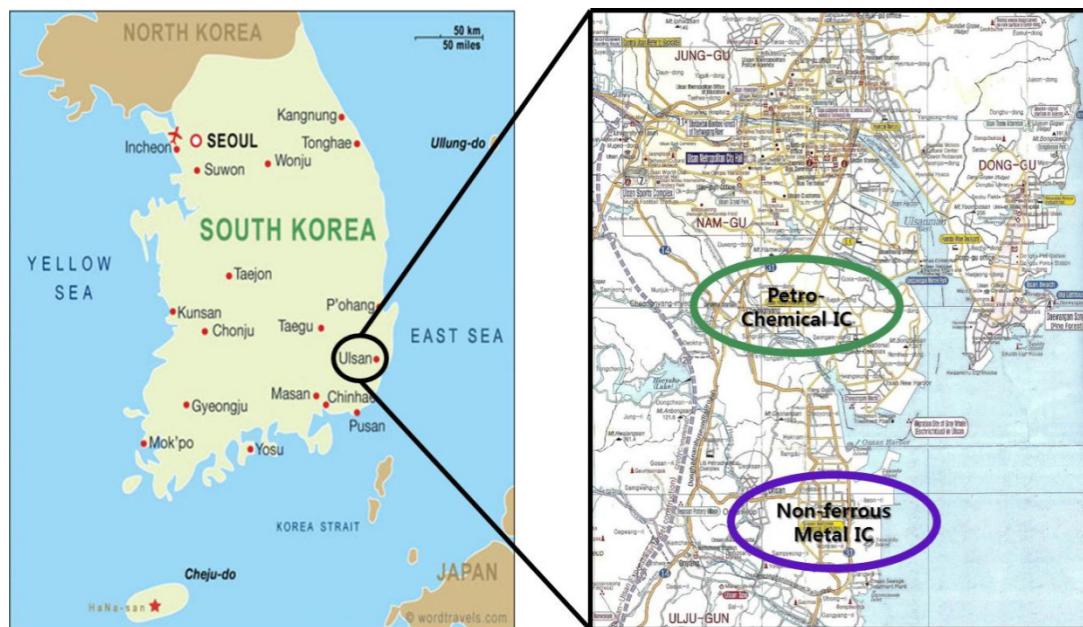


Figure 1. Ulsan map showing PC and NFI sampling sites

Table 1. PM mass calculation

Stage	Size range (μm)	Mass	Calculation formula
0	9.0-10.0	m1	$\text{PM}_{10} = m9 + m8 + m7 + m6 + m5 + m4 + m3 + m2 + 0.76 \times m1$
1	5.8-9.0	m2	
2	4.7-5.8	m3	
3	3.3-4.7	m4	$\text{PM}_4 = m9 + m8 + m7 + m6 + 0.84 \times m5 + 0.13 \times m4$
4	2.1-3.3	m5	
5	1.1-2.1	m6	
6	0.7-1.1	m7	$\text{PM}_1 = m9 + m8 + m7 + 0.44 \times m6$
7	0.4-0.7	m8	
Backup filter	<0.4	m9	

2.2 Meteorological Data Analysis

Meteorological data (wind speed and wind direction) were taken from the network station (located at the sampling site) for air quality monitoring in Ulsan. The windrose diagrams was plotted based on hourly data using Korean software for atmospheric dispersion modeling (Airmaster, version 2.0) provided by Korean Meteorological Administration.

2.3 Heavy Metals Analysis

Due to the low mass in each filter, a total of 9 samples were grouped into three composite samples (0-1.1, 1.1-4.7 and 4.7-10 μm). The composite mixture was dissolved in approximately 30 cc of 1:1 mixture of nitric acid (1.03 M) and perchloric acid (2.23 M) and extracted using an ultrasonic bath (50 kHz) for 1 hr (50% cycle). After extraction, the extract was filtered through glass fiber filters (GF75, Advantec) and diluted. The diluted extract was used to quantify heavy metals using Atomic Absorption Spectroscopy (Varian, AA 240). Five heavy metals (Ni, Cu, Zn, Pb, and Cd) were quantified.

Blank correction was done by measuring the back filters and membrane using the gravimetric method and heavy metals measurement used the method described above. The blank contribution to the total concentration was regularly under 15% for all the elements. Accuracy and precision (repeatability) were measured using Standard Urban Dust Reference Material (NIST, SRM-1648). The recovery was between 80-115%.

2.4 Statistical Analysis

Two statistical methods, Pearson correlation analysis and cluster analysis, were tested to measure the relationship among the variables (particle sizes and seasons). Pearson correlation “r” was investigated by comparing the “r” value. Pearson’s “r” is invariant to scale changes where the original data is transformed to standardized form, dividing the distance from the mean by the sample standard deviation. The properties of Pearson’s correlation coefficient lie between -1 to +1, with 0 indicates no relationship between pairs of variable sand follows “t” distribution with $n-2$ degrees of freedom, where n is the number of samples. To test the significance of correlation (r), where r differs from zero, the test statistic t_r is defined as:

$$t_r = \frac{r\sqrt{(n-2)}}{\sqrt{(1-r^2)}}$$

It was considered 1% level of significance (α) for $r \geq 0.9$ and 5% level of significance for $0.7 \leq r \leq 0.89$. We used a cumulative sum (CUSUM) method (Kamruzzaman, Beecham, & Metcalfe, 2011), to test whether there was an evidence of relative changes under the mean or not. The CUSUM at time n was calculated as:

$$C_t = \sum_{i=1}^n (x_i - \bar{x})$$

where x_i represent a sample size, \bar{x} is the mean of the sample of length n . It is noted that if consecutive values tend to lie below the mean, then C_t will have a negative slope and if consecutive values tend to lie above the mean then C_t will have a positive slope.

Cluster analysis (CA) was performed to classify particles sizes on the basis of the similarities of their concentration. Hierarchical cluster analysis, used in this study, assisted in identifying relatively homogeneous groups of variables (particles sizes), using an algorithm that starts with each variable in a separate cluster and combines clusters until only one is left. A dendrogram was plotted to assess the distance of the clusters formed (Aryal, Lebegue, Vigneswaran, Kandasamy, & Grasmick, 2009).

3. Results

3.1 Particle Size Distribution

The total PM_{10} ranges were $59 \pm 14 \mu g/m^3$ in summer and $66 \pm 18 \mu g/m^3$ in fall at the PC site and $52 \pm 14 \mu g/m^3$ in summer and $88 \pm 36 \mu g/m^3$ in fall at the NFI site. The result shows that PM_{10} concentration in fall was relatively higher than in summer. The result shows although the value did not exceed the daily average guideline $100 \mu g/m^3$ but exceeded annual average guideline $100 \mu g/m^3$ (Ministry of Environment, 2007). By the beginning of fall, the ambient temperature drops down. Increase in PM_{10} in with lowering atmospheric temperature has been reported in other cities (Aryal et al., 2008; Ho et al., 2006; Kulshrestha, Satsangi, Masih, & Taneja, 2009) and in Ulsan residential area also (Hieu & Lee, 2010; Lee & Park, 2010). Table 2 shows PM_{10} observed in other industrial sites across the world. The results show that the PM_{10} observed in this study at both sites were within the range of reported values in the literature (Fang et al., 2000; Karar, Gupta, Kumar, & Biswas, 2006).

Figure 2 shows PM_{10} distributionin both sites and seasons. The figure indicated that there was difference in PM_{10} in summer and fall on both sites not only in total but also in fractions (particle sizes). To understand the relationship among particles within sites among two seasons, we calculated Pearson Correlation “r”. Table 3 shows the correlation coefficient for particles (sizes) within sites and between sites.

Table 2. PM_{10} value observed in other industrial sites

Site	Features	Concentration ($\mu\text{g}/\text{m}^3$)
La Coruna, Spain (Beceiro-González, Andrade-Garda, Serrano-Velasco, & López-Mahía, 1997)	Industrial site	25
Taejon, Korea (Kim, Lee, & Jang, 2002)	Industrial site	72
Barcelona, Spain (Rodríguez et al., 2004)	Industrial site	33
Kolkatta, India (Karar & Gupta, 2006)	Industrial site	62-401
Tito Scalo, Italy (Ragosta et al., 2006)	Industrial site	24
Dunkirk, France (Alleman, Lamaison, Perdrix, Robache, & Galloo, 2010)	Industrial site	14-36
Basque country, Spain (Viana, Querol, Alastuey, Gangoiti, & Menéndez, 2003)	Urban area near to industry	34
San Joaquin valley, CA, USA (Chow et al., 1999)	Urban area near to industry	50

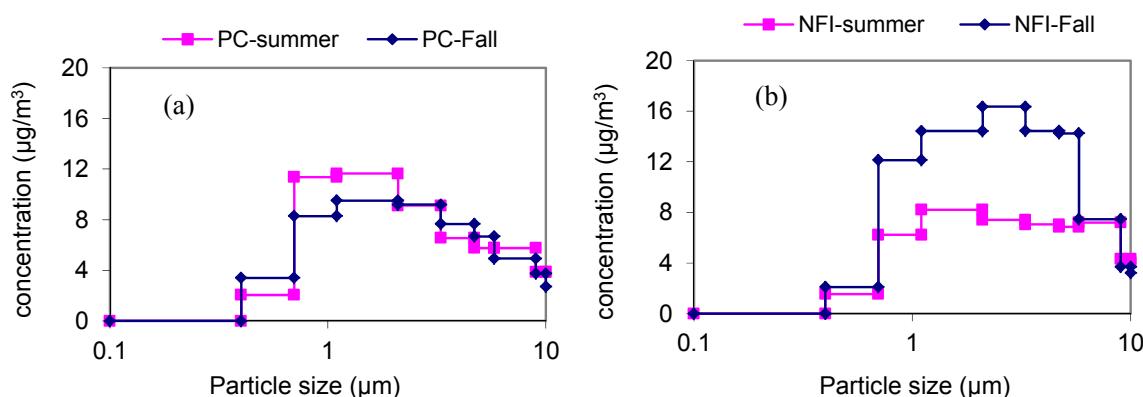


Figure 2. Particle size distributions for (a) PC and (b) NFI areas

Table 3. Relationship between particle sizes

	PC in Summer	PC in Fall	NFI in Summer	NFI in Fall
PC in Summer	1			
PC in Fall	0.91*	1		
NFI in Summer	0.77**	0.81*	1	
NFI in Fall	0.77**	0.94*	0.86*	1

*Coefficients are statistically significant at 1% level.

**Coefficients are statistically significant at 5% level.

The statistic showed that PM_{10} had significant correlation in PM_{10} between summer and fall (explain with what?) ($r = 0.91$ for PC site and 0.86 for NFI site with level of significance 1%). Among the sites (PC and NFI), the Pearson 'r' was 0.77 with level of significance were at 5%. Moreover, we observed significant mean difference (t statistics) and noticeable relative changes at $5.61 \mu\text{m}$ with $p = 0.000803$ for PC sites and at $4.47 \mu\text{m}$ with $p = 0.002903$ for NFI sites for summer and fall, when we used the cumulative sum method. Figure 3 shows relative change in particle sizes in PC and NFI in summer and fall. The solid line (vertical) shows where maximum noticeable change occurs in PM fractions in summer and fall.

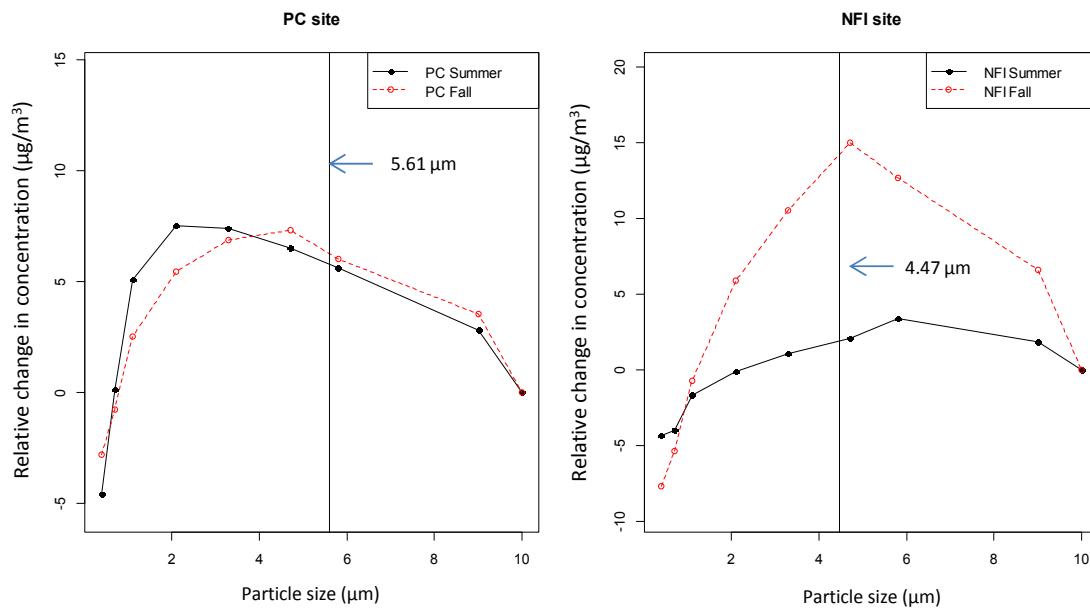


Figure 3. Particle size distributions for (a) PC and (b) NFI areas

Noticeable changes in particle size distribution for PC and for NFI in summer and fall season suggests that agglomeration of particles and/or intrusion of particles from surrounding areas may have occurred in fall. To understand the possible intrusion from surrounding sites, we plotted wind rose diagram for summer and fall (Figure 4). The wind direction in the summer is from southeast to northwest (sea to land). During fall, as the temperature drops down and the prevailing wind flows from north to south. Since NFI is at downstream of the industrial zone, two possible assumptions were thought for higher concentration of PM in NFI during fall: i) the northwesterly wind brought particulates from the upstream to NFI and ii) NFI produced more PM in fall.

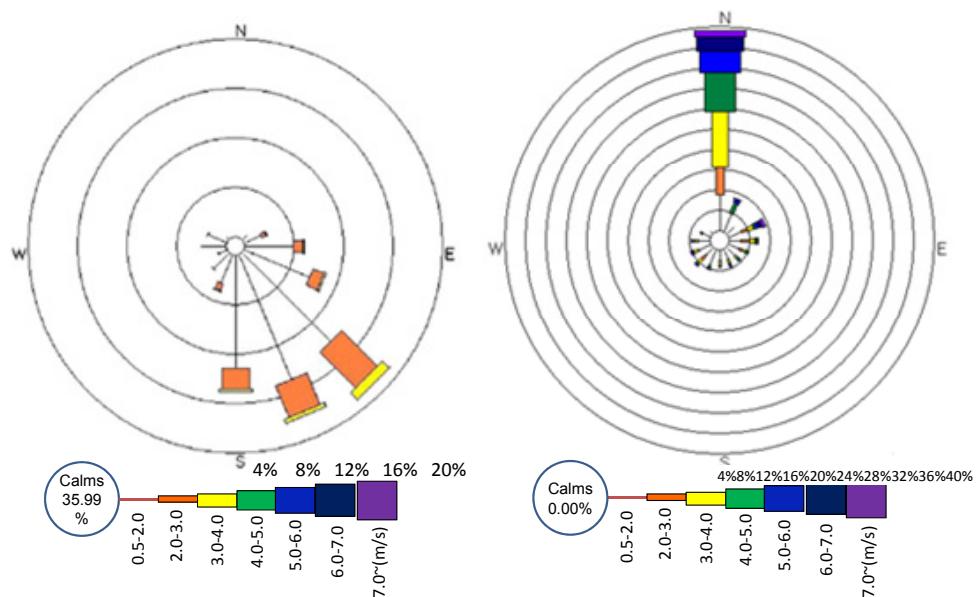


Figure 4. Windrose diagram at sampling site for summer (left) and fall (right)

To understand the particle size relationship, we plotted dendrograms using a cluster analysis. This method of analysis is described in (Aryal et al., 2009). Figure 5 shows dendrogram plots for particles collected at the PC and NFI sites in both summer and fall. The dendrograms show that the particle size distribution is mainly bimodal, with one cluster corresponding to the finer fraction 1.1-4.7 μm and the other cluster to the coarse mode 4.7-10 μm , shown by dotted boxes. Similar bimodal distributions have been reported earlier (Karanasiou et al., 2007; Toscano et al., 2011). Our observations show a close relationship between the ultrafine particles ($<1.1 \mu\text{m}$) and the coarser fraction ($>5.8 \mu\text{m}$). The relationship between ultrafine and coarse fractions suggests the possibility of similar sources and/or modes of release.

3.2 Heavy Metals Distribution in Particles

Five heavy metals (Ni, Cu, Zn, Pb, and Cd) were measured. Since the concentrations of the particles in the samples were low, composite samples were made by mixing nine different sizes to three fractions (10-4.7, 4.7-1.1 and 0.1-1.1 μm). Figure 6 shows the pseudo total heavy metals concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) in the total particulate (PM_{10}) calculated as mass fractions. Pseudo total heavy metal concentration refers to the sum of measurement of individual fractions. Out of total metals abundance, Zn abundance is more than 86%. It is seen that while the total particulate matter concentration increased in fall, the overall heavy metal content decreased except for Ni at NFI site. Similar and higher concentration of heavy metal contents in particulate matter in summer has been reported in literatures (Espinosa, Ternero Rodríguez, De La Rosa, & Sánchez, 2001; Gupta, Salunkhe, & Kumar, 2010; Querol et al., 2006; Razos & Christides, 2010; Shah, Shaheen, & Jaffar, 2006; Smichowski et al., 2004).

To know the possible reasons for high value of Zn and Ni in fall than summer we plotted the graph between fractional concentration and total concentration as shown in Figure 7. It can be assumed that if the sources for both seasons are similar then fractional distribution of metals with respect to total will also be similar whereas inclusion of surrounding sources changes the relationship to good or poor depending upon incursion amount. The graph showed that relationship between fractional metal concentrations with total are more linear in summer than in fall. It is to be noted that Zn concentration became slightly higher in fall than in summer (less than 2 fold) whereas Ni concentration increased almost 6 folds fold in fall than in summer in NFI. Table 3 shows the estimated fraction particle size effect and filled regression model. Due to small decrease in concentration for Zn R^2 value was relatively lower in fall than in summer. For Ni strong increase of its concentration, R^2 value changed significantly. This led us to conclude that Zn and Ni were added to the atmosphere in PC and NFI from surrounding during fall season.

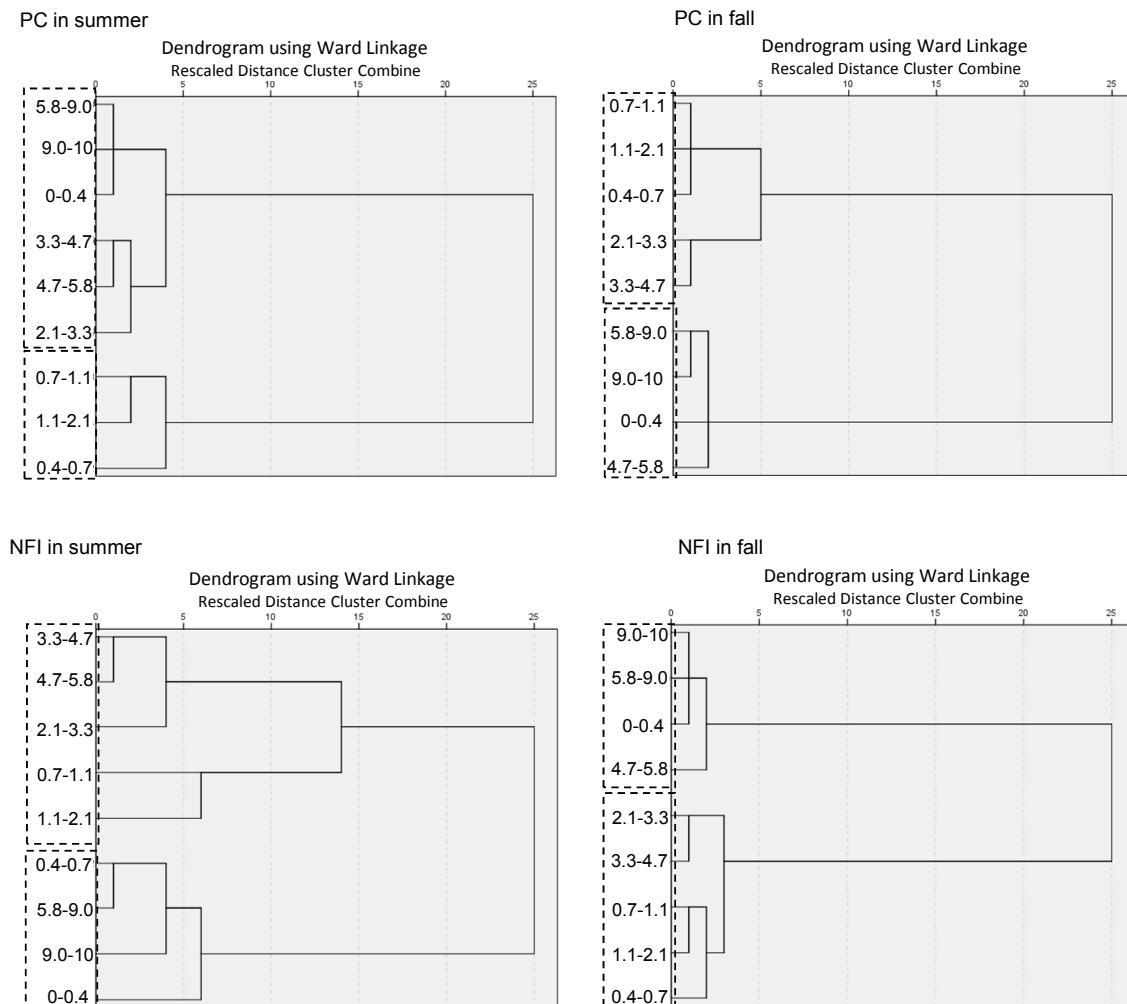
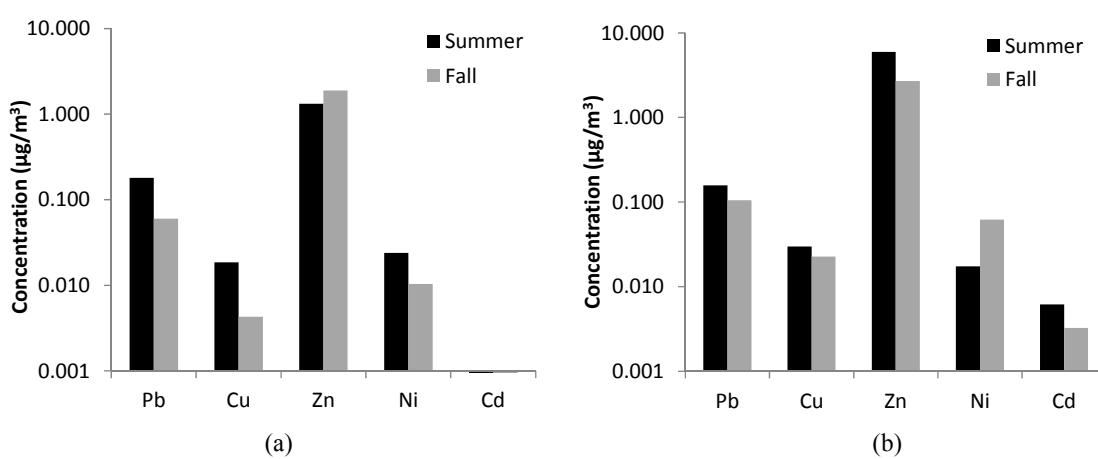


Figure 5. Particle size distributions and their relationships

Figure 6. Pseudo total metal concentrations ($\mu\text{g}/\text{m}^3$) in total particulate matter in (a) PC and (b) NFI

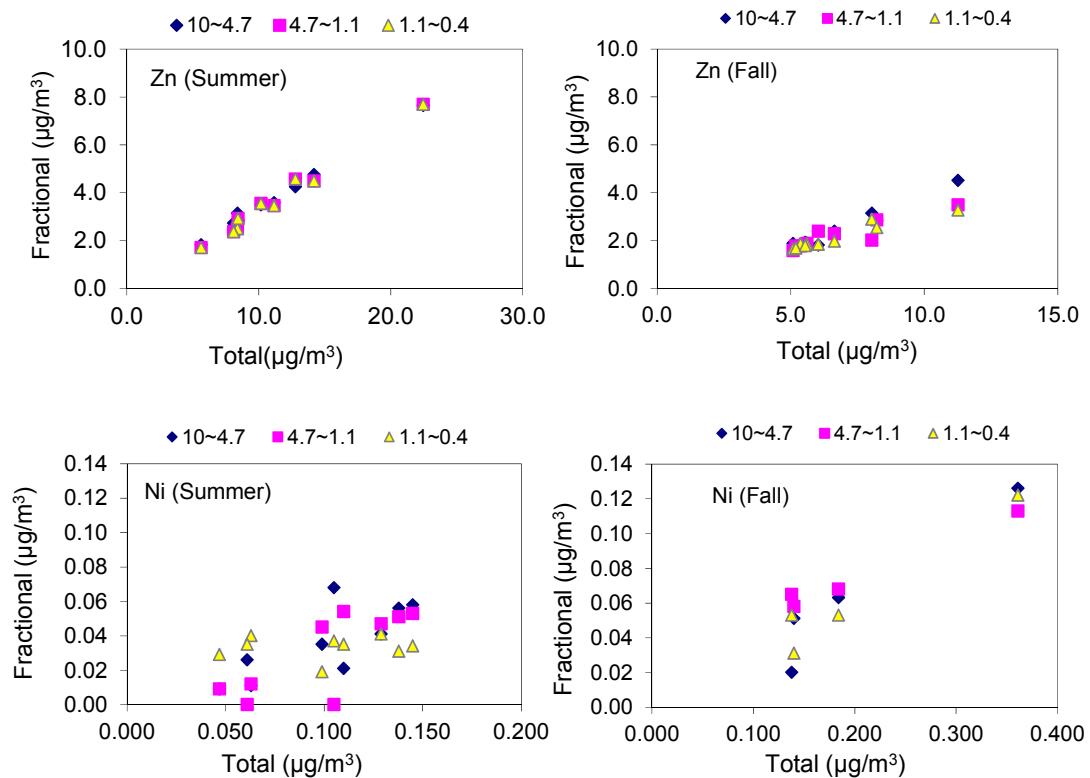


Figure 7. Relationship between fractional concentration with total for Zn (in PC) and Ni (in NFI)

4. Conclusions

Atmospheric particulate matter and associated heavy metals in less than 10 μm was investigated at two industrial sites in Ulsan, South Korea for two seasons were analyzed. The following conclusions can be drawn from this work:

Total PM₁₀ ranged from $59 \pm 14 \mu\text{g}/\text{m}^3$ in summer and $56 \pm 18 \mu\text{g}/\text{m}^3$ in fall for the petrochemical (PC) site whereas for the non-ferrous heavy metals (NFI) site it was around $52 \pm 14 \mu\text{g}/\text{m}^3$ in summer and $88 \pm 36 \mu\text{g}/\text{m}^3$ in fall. An increase of PM₁₀ concentration at the NFI site in fall indicated that this area possibly receives particulates from surrounding areas.

An increase of 0.4–5.8 μm particle size in fall at the NFI site indicates that the area may have received particulate matter from other areas.

The heavy metals concentration was relatively higher in the summer than in fall (except for Zn in PC and Ni in NFI) and the change in concentration was possibly due to contribution from surrounding sources.

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