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Comparison of physicochemical properties between fine ($PM_{2.5}$) and coarse airborne particles at cold season in Korea

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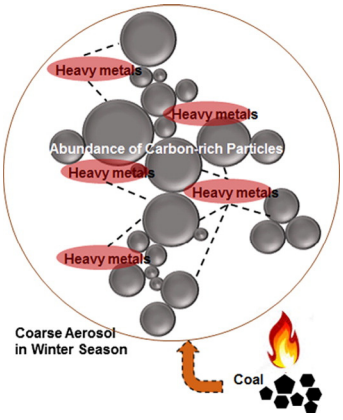
HIGHLIGHTS

- We characterized physicochemical properties of ambient aerosols at cold seasons.
- Surface chemistry and morphologies between fine and coarse particles were compared.
- Water solution soaked by $PM_{2.5}$ particles showed acidic condition.
- Spherical carbonaceous particles were frequently observed for coarse sample.
- Coarse particles contribute to conveyance of toxic contaminants into environments.

GRAPHICAL ABSTRACT



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ABSTRACT

Although it has been well-known that atmospheric aerosols affect negatively the local air quality, human health, and climate changes, the chemical and physical properties of atmospheric aerosols are not fully understood yet. This study experimentally measured the physiochemical characteristics of fine and coarse aerosol particles at the suburban area to evaluate relative contribution to environmental pollution in consecutive seasons of autumn and winter, 2014–2015, using XRD, SEM–EDX, XNI, ICP-MS, and TOF-SIMS. For these experimental works, the fine and coarse aerosols were collected by the high volume air sampler for 7 days each season. The fine particles contain approximately 10 μg m^{-3} of carbonaceous aerosols consisting of 90% organic and 10% elemental carbon. The spherical-shape carbonaceous particles were observed for the coarse samples as well. Interestingly, the coarse particles in winter showed the increased frequency of carbon-rich particles with high contents of heavy metals. These results suggest that, for the cold season, the coarse particles could contribute relatively more to the conveyance of toxic contaminants compared to the fine particles in the study area. However, the fine particles showed

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Morphology
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acidic properties so that their deposition to surface may cause facilitate the increase of mobility for toxic heavy metals in soil and groundwater environments. The fine and coarse particulate matters, therefore, should be monitored separately with temporal variation to evaluate the impact of atmospheric aerosols to environmental pollution and human health.

1. Introduction

Atmospheric aerosols have recently received attention in Korea due to the increase of fine particulate matter ($PM_{2.5}$) in atmospheric environments, and their human health risk. $PM_{2.5}$ is primarily emitted to the air through combustion and high temperature processes of biomass and fossil fuels so that the increase of $PM_{2.5}$ particles are associated with an industrialization and urbanization processes (Colbeck, 2008; Lakhani and Kumari, 2012). In particular, the Korean peninsula has been geographically affected by Asian Dust storm originated from neighboring countries such as China and Mongolia (Chun et al., 2008). The rapid industrial growth of those countries could significantly deteriorate air quality in Korea (Wang et al., 2005; Kim et al., 2012; Lee et al., 2013). It has been recognized that $PM_{2.5}$ typically contain abundant hazardous materials including heavy metals and organic compounds because of their smaller particle size with greater surface areas compared to the coarse airborne particulates (Pope and Dockery, 2006; Pöschl, 2005).

One of the main components in $PM_{2.5}$ particles is carbonaceous aerosol (i.e., a mixture of organic carbon and black carbon) that plays an important role on hazardous material conveyance, human health, and climate change. In urban areas, for example, the carbonaceous aerosols account for ≥50% of total mass concentrations in ambient aerosols (Kanakidou et al., 2005; Murphy and Pandis, 2009; Seinfeld and Pandis, 2006; Zhang et al., 2007). In addition, the carbonaceous aerosols are particularly sensitive to heterogeneous chemical reactions (Chungtai et al., 2002), which may control the fate and transport of toxic metals and carcinogenic organic chemicals such as polycyclic aromatic hydrocarbons (PAH) and nitro-PAHs (Zhu et al., 2001).

Human exposure to $PM_{2.5}$ and carbonaceous aerosols was strongly related to the increase of lung and cardiovascular diseases through inhalation of ambient air due to their ultrafine size and toxicity (McDonald et al., 2004; Metzger et al., 2004; Pope and Dockery, 2006; Yttri et al., 2009; Samet et al., 2005). Furthermore, the carbonaceous aerosols are the third largest contributor to the global warming following carbon dioxide and methane (Ramanathan and Carmichael, 2008; Yttri et al., 2009), and act as cloud condensation nuclei which lead to the formation of atmospheric brown clouds (Lohmann and Feichter, 2005; Ramanathan and Carmichael, 2008).

Although several previous studies were conducted to evaluate air quality, health effects, and source identification associated with atmospheric particles in Korea, their studies focused mainly on spring season due to frequent events of the Asian Dust storm (Kim et al., 2007; Heo et al., 2009; Lee et al., 2013, 2015). However, airborne particles in the cold season could contain more carbonaceous materials because of the increase of fossil fuel usage. The China, for example, is the world's largest consumer of coal for residential heating in the winter (Zhang et al., 2012). The coaly particles transported by wind to the neighboring regions could adversely affect atmospheric environment related to air pollution (Zhang et al., 2012; Zhang et al., 2013). Therefore, the goal of this study is to characterize the physical and chemical properties of ambient aerosols at the cold seasons in the suburban area of Korean. The suburban area was regarded as relatively non-contaminated area compared to the metropolitan cities such as Seoul and Daejeon in Korea. The specific objectives are to compare the physiochemical characteristics between the fine ($PM_{2.5}$) and coarser particles, and to evaluate the relative contribution and the effect of each aerosol on environmental system.

2. Materials and methods

2.1. Sampling

Sampling was conducted at the roof of the Isotope Building in Korea Basic Science Institute (~10 m height from surface) located at Ochang, northwest Cheongju City (Figure A1 in the Appendix A. Supplementary data [36°43'N 127°26'E]). The Ochang is a small town of Cheongju City with about 47,000 inhabitants in the area of 80 km^2. The land has been used as 28% industrial, 20.6% public, 18.7% green, 14% residential, 13.4% R&D, and 5.3% commercial business areas around the sampling site (www.ochangcmc.or.kr). Climate of the town reflects a continental climate with evident four seasons, which is affected by Monsoon in the summer and Siberian anticyclone during the winter. Annual mean temperature, precipitation, and relative humidity are 12.5 °C, 1239.1 mm, and 67.7%, respectively, based on the historical records through 1981–2010 (Korea Meteorological Administration (KMA), 2013).

Atmospheric aerosols were collected two periods (22–29 Oct. 2014, and 9–16 Jan. 2015) to monitor 7-day averages, respectively, using the 230 Series High Volume Cascade Impactors (Tisch Environmental., USA) at a rate of 1.13 $m^3\ min^{-1}$. A single stage high volume impactor was utilized to collect $PM_{2.5}$ aerosol samples over ADVANTEC QR-100 quartz fiber filters (Toyo Roshi Kaisha, Japan). The coarser airborne particles compared to $PM_{2.5}$ were accumulated by the glass fiber slotted collection substrates (TE-230-GF, Tisch Environmental) in the high volume air sampler during the sampling periods. All filters were precombusted at 900 °C (quartz), and 500 °C (glass) for several hours in a furnace before sampling processes. Both of the sampled $PM_{2.5}$ and coarse particles were analyzed to determine the physicochemical morphology, components, and compositions. The detailed experimental methodology was described in following sections.

2.2. Physical characterization

X-ray diffraction (XRD) analysis was performed to determine the mineral components in the $PM_{2.5}$ and coarser particle samples. The SmartLab XRD (Rigaku, Japan) was used with Cu Kα radiation at 40 kV and 200 mA ($\lambda = 1.54050°$). The diffraction patterns were obtained from 3 to 65° 2θ with a fixed time of 1 min per 1° 2θ.

Field-Emission Scanning Electron Microscope (FESEM: SUPRA 55VP, Carl Zeiss, Germany) was used to observe the morphologies for the $PM_{2.5}$ and coarse particle samples, after the samples were coated with platinum. The elemental analysis was conducted to specify the morphologies using the equipped Energy Dispersive X-ray Spectrometer (EDS) system (Bruker). The accelerating voltage for the electron microbe was 10 kV under high vacuum pressure during operation.

In addition, the X-ray Nano Imaging (XNI) was collected for the samples on 7C XNI beamline of Pohang Light Source (PLS-II) operating with the 3.0 GeV and 400 mA stored-beam-current parameters. A liquid-nitrogen-cooled silicon (111) double-crystal monochromator was used to acquire the monochromatic X-ray beam from the high flux X-rays of the undulator source. Each aerosol particulate was dispersed on polyimide film with a proper thickness, and mounted on a three-axis piezo-driven scanning stage. The X-ray nano images were obtained through detector system comprising a scintillator crystal and optical ×20 microscope with a CCD (4096 × 4096 pixels of 9 μm size). More detailed information on the 7C XNI beamline was described in previous literature (Lim et al., 2014).

2.3. Chemical characterization

The concentrations of major cations, heavy metals, and rare earth elements were determined for the $PM_{2.5}$ and coarse particle samples. The filters containing aerosol particles were placed in Teflon digestion vessel for acid treatment. Each sample was treated with ~5 mL acidic mixture of HNO_3:HF:$HClO_4$ (volumetric ratio of 4:4:1) at 150 °C for 24 h to decompose the samples. The 20 mL of 1% HNO_3 solution was added to the individual batch after drying. The aliquot was analyzed using inductively coupled plasma atomic emission spectrometry (ICP-AES: Optima 8300, Perkin-Elmer, USA) for Al, Ca, Cr, Cu, Fe, K, Mg, Mn, Ti, V, Zn, and mass spectrometer (ICP-MS: Thermo Elemental X5, Thermo Scientific, UK) for trace and rare earth elements, respectively.

Additionally, the atmospheric aerosol samples were soaked into ~15 mL of Milli-Q water to determine major anionic concentrations. The samples were placed in the oven at 90 °C for 3 h to extract water-soluble matter, and they were cooled down at ambient temperature. After 48 h, the aqueous phase was filtered to remove impurities using a 0.45 μm PVDF syringe filter. Portable pH meter (Orion Star A211, Thermo Scientific) was used to measure pH value in each sample. The major anions (F^-, Cl^-, Br^-, NO_2^-, NO_3^-, SO_4^{2-}, PO_4^{3-}) were analyzed by DIONEX ICS-2100 Ion Chromatograph system (Thermo Scientific) equipped by AG18 and AS18 for the guard and analytical column, respectively.

The surface chemistry of the $PM_{2.5}$ and coarse particle samples was characterized by a time-of-flight secondary ion spectrometer (TOF-SIMS: TOF-SIMS V, ION-TOF GmbH, Münster, Germany). A bismuth primary ion beam impacted the surface of atmospheric aerosol particles deposited on the filter to collect the ion spectrum of emitted secondary negative and positive ions. The operating voltage of the Bi_3^+ ion beam is 30 keV with target current of 0.6 pA. The target area is $100 \times 100\ \mu m^2$ for the TOF-SIMS analysis.

3. Results and discussion

3.1. Atmospheric particle constituents

The $PM_{2.5}$ and coarser particle samples collected in the autumn (Oct. 2014) showed black (2.5Y2.5/1 [Munsell Soil-Color Charts]), and grayish brown (2.5Y5/2) color, respectively. In the winter (Jan. 2015), the $PM_{2.5}$ samples maintain comparative color, black (GREY1 2.5/N), while the color for coarser ones became thicker to very dark grayish brown (2.5Y3/2). Analysis of the black color in $PM_{2.5}$ at both autumn and winter samples revealed approximately 10 μg C m^{-3} consisting of ~90% organic, and ~10% elemental carbon regardless of the seasonal variation.

Fig. 1 indicates the results of XRD analyses for the fine and coarse particle samples. The coarse particles exhibited high intensity of mineral components suggesting that the formation of coarse particles is strongly related to fragmentation of soils and rocks. The dominant mineral group was quartz and feldspar, and minors were carbonates, gypsum, chlorite, and mica. In contrast, the salts of ammonium nitrate (NH_4NO_3) were observed more frequently for the $PM_{2.5}$ sample compared to the mineral particulate matter. The fine particles are predominantly formed by heterogeneous chemical reactions through incomplete combustion and gas-to-particle conversion in the atmospheric environments (Ian Colbeck, 2008). In particular, the NH_4NO_3 is one of secondary-produced components from the oxidation of NOx emissions in the suburban area (Heal et al., 2012).

The pH of water solution soaked by the $PM_{2.5}$ and coarser samples indicated slightly acidic and basic conditions, showing as pH = 4 and 8, respectively. The alkaline pH value for the coarser samples could be affected by Ca-rich mineral components as buffer materials. The measured anionic concentrations in the water soluble fractions of $PM_{2.5}$ were approximately 3 to >10 times greater than those of the coarser samples (Fig. 2). The anions of nitrate, sulfate, and chloride increased up to a factor of 3–8 in the winter season. The higher atmospheric concentrations of sulfur and nitrogen dioxide in the 2014–2015 winter season at the study area (Table A1) could support the abundance of sulfate and nitrate, because the formation of sulfate and nitrate is associated with the oxidation processes of sulfur and nitrogen dioxide in the atmosphere (Heal et al., 2012). These results agreed comparatively to the XRD data. The increase of chloride seems to cause the mixture of sea water spray and/or contaminants to the study area.

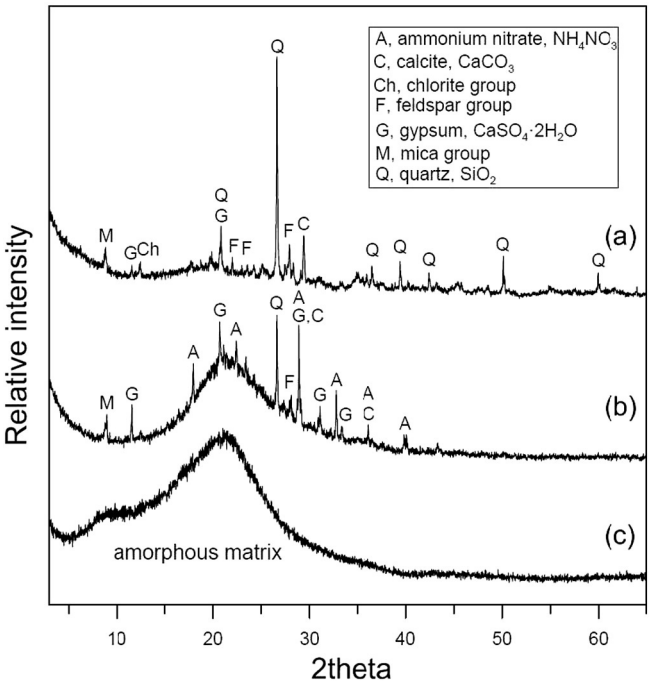

Fig. 1. The XRD results for (a) coarse particles, (b) $PM_{2.5}$, and (c) quartz fiber filter using the autumn samples.

3.2. Surface chemistry

The elemental distribution of positive ions detected on a surface of 100×100 μm area for the $PM_{2.5}$ and coarse particles were shown by TOF-SIMS images in Fig. 3(a) and (b). Potassium and sodium ions showed relatively greater intense distribution (i.e., bright region) in the lateral surface of $PM_{2.5}$ compared to other elemental ions, while high intense peaks for K, Na, Mg, Ca, Fe, Al, and Si were observed for the coarse particles. The intensity of spatial distribution for potassium and sodium could be associated with sulfates and nitrates in the $PM_{2.5}$ samples. The coarse particles consist of more naturally occurring earth elements including K, Na, Mg, Ca, Fe, Al, and Si compared to the $PM_{2.5}$ particles. The chemical and spatial composition was comparatively

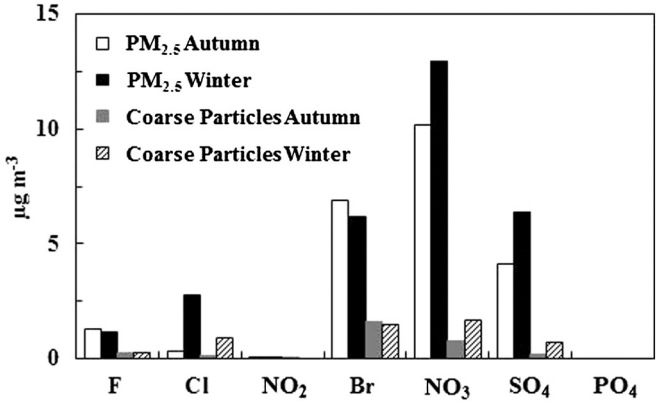

Fig. 2. Anionic concentrations in water soluble fraction of the aerosol samples.

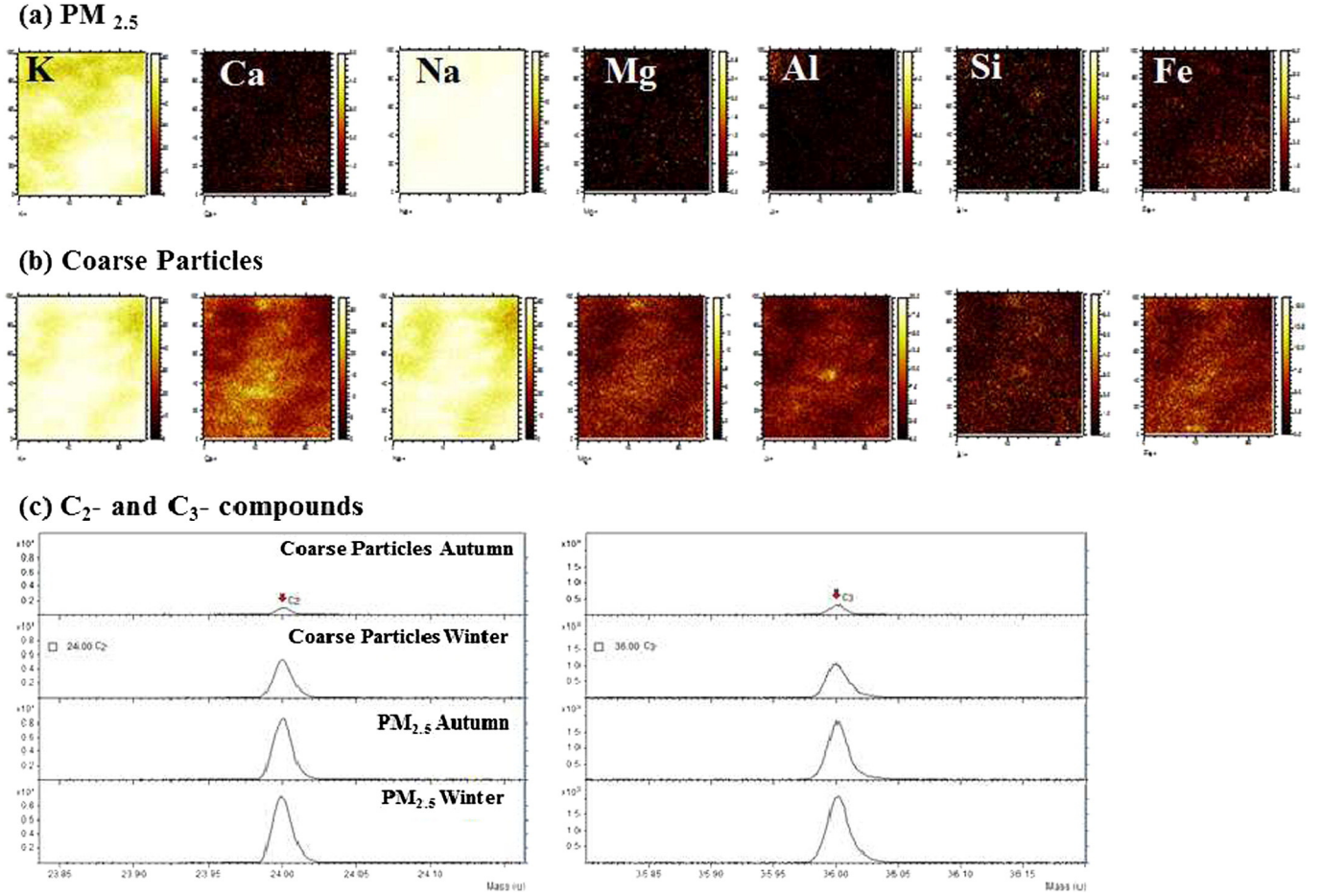

Fig. 3. TOF-SIMS images for elemental distribution on the surface of (a) $PM_{2.5}$ and (b) coarse particle samples collected in the winter season. Fig. 3(c) exhibits C_2- and C_3-compound peaks for each aerosol sample with seasonal variation.

identical for both $PM_{2.5}$ and coarse particle samples collected at autumn or winter season.

In addition, high intensity of the C_2- and C_3-compound peaks were observed for the $PM_{2.5}$ samples regardless of seasonal variation, suggesting abundant presence of aliphatic and aromatic hydrocarbons such as organic chemicals and black carbon materials (Fig. 3(c)). The coarse particles, interestingly, exhibited the increase of peak intensity for C_2- and C_3-hydrocarbons for the winter sample.

3.3. Morphology

Information on surface morphology determined at the laboratory is required to provide microphysical state of ambient aerosols with the chemical composition. The morphology of both $PM_{2.5}$ and coarse particle samples showed complicated structures including irregular, spherical, cluster, and chain-like shape particles (Figs. 4 & 5). However, the coarser sample contains more crystalline particles (e.g., mineral particles) compared to abundant amorphous shape in the $PM_{2.5}$ sample. The mineral particles have irregular shape with a sharp edge suggesting that these particles are mainly originated from geological sources such as soils and resuspended soil dust (Li et al., 2010). Predominant contents of elemental Si and O with Al, Fe, Ca, Mg, and K indicated that most of minerals are silicate and aluminosilicates such as quartz and feldspar, which is consistent with the XRD results. In addition, a certain amount of carbon was detected on the surface of minerals implying the adsorption of carbonaceous matter on their surface (Fig. 5).

In the case of $PM_{2.5}$ samples, the elemental composition was abundant following the order of carbon, oxygen, and nitrogen (Figure A2). The higher fractions of carbon in $PM_{2.5}$ sample indicated the relative abundance of carbonaceous materials such as black carbon (i.e., soot), tar balls, and fly ashes. On the other hand, the spherical particles were frequently observed with the size fraction of ~3 to <10 μm diameter in the SEM images of the coarse particle sample (Fig. 4). In particular, the relative number of these spherical particles increased slightly in the coarse particle samples collected at the winter season. A spherical-shape particle is typically formed through high-temperature combustion processes including wild biomass burning, coal combustion, and metallurgical emissions (Xie et al., 2005; Yue et al., 2006; Kim et al., 2012).

The elemental compositions of the spherical single and chain-like particles for the coarse particle sample are predominantly carbon and oxygen with aluminosilicates, potassium, and calcium (Fig. 5). These airborne particles might be originated from limestone in coal combustion process at local industries and/or thermal power plant (Yue et al., 2006). The fact that northwesterly wind is dominant in winter at the study area (Table A2) provides potential inflow of coal-derived fly ash from a power plant located west of the study area near the Yellow sea or China. Relative abundance of chlorine in the winter samples indicated mixing effects of sea salts with the ambient aerosol. Additionally, Lee et al. (2015) reported that atmospheric pollutants including a certain amount of coal fly ash were transported from China to Korea based on the Pb isotopic results.

In contrast, one of main carbonaceous materials for the $PM_{2.5}$ sample was soot that is characterized by aggregated clusters with small particle diameter of <1 μm (Kocbach et al., 2005) (Fig. 5). Soot aggregates are typical aerosol components which are formed by vehicle exhausts (Shi et al., 2003; Frank and Herbarth, 2002). The soot particles are usually embedded into the host materials such as sulfate and/or other organic matter (Adachi et al., 2010). The secondary formed aerosol components

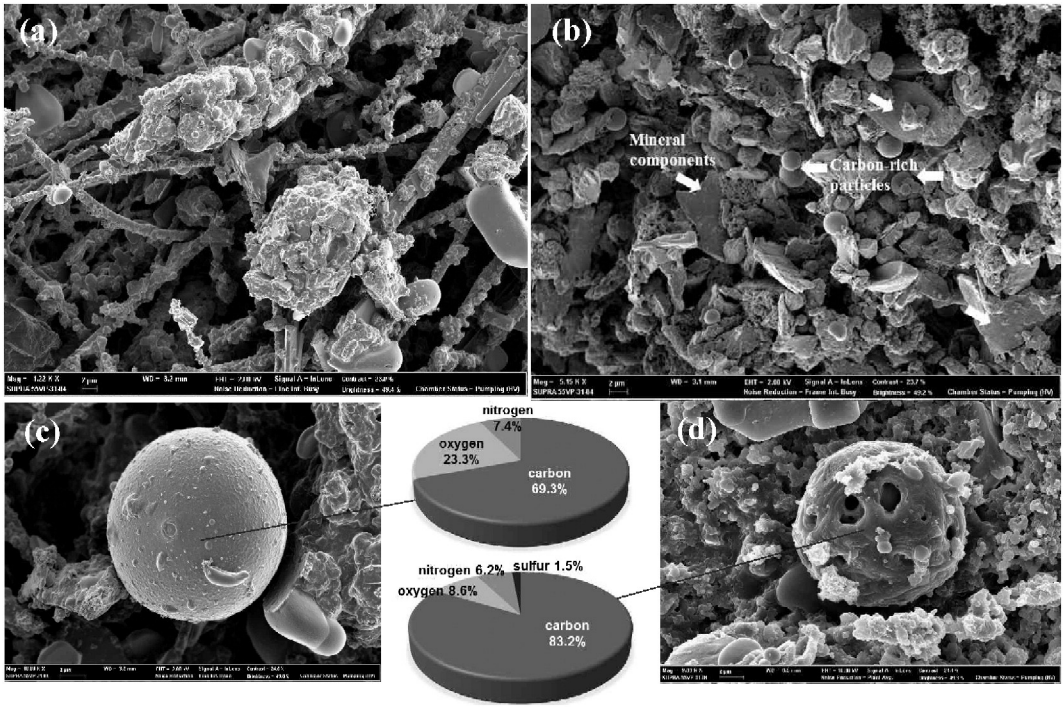

Fig. 4. Representatives of typical SEM images for this study: (a) $PM_{2.5}$, (b) coarse particle sample, (c) carbon-rich particle, and (d) carbon-rich particle with degassing holes formed by high temperature processes.

including ammonium nitrate may play an important role on the aggregation of soot particles in our samples.

3.4. Heavy metals

The relative contribution of fine and coarse particles to metal conveyance was computed by a ratio of the measured elemental concentration for each particle sample and summed total concentration, assuming that the studied fine and coarse particles represent total aerosol particles. The $PM_{2.5}$ particles contributed significantly to heavy metal conveyance showing approximately >50% of total concentrations for Fe, Cu, Mn, Pb, Zn, Ni, Cr, Co, As, Cd, Sr, Rb, Sn, and Sb in the autumn samples (Fig. 6(a)), although the elements of Ni, Cr, Co, As, Cd, Sn, and Sb were present as a trace below 10 ng m^{-3} (Table A2). The earth constituents in crust

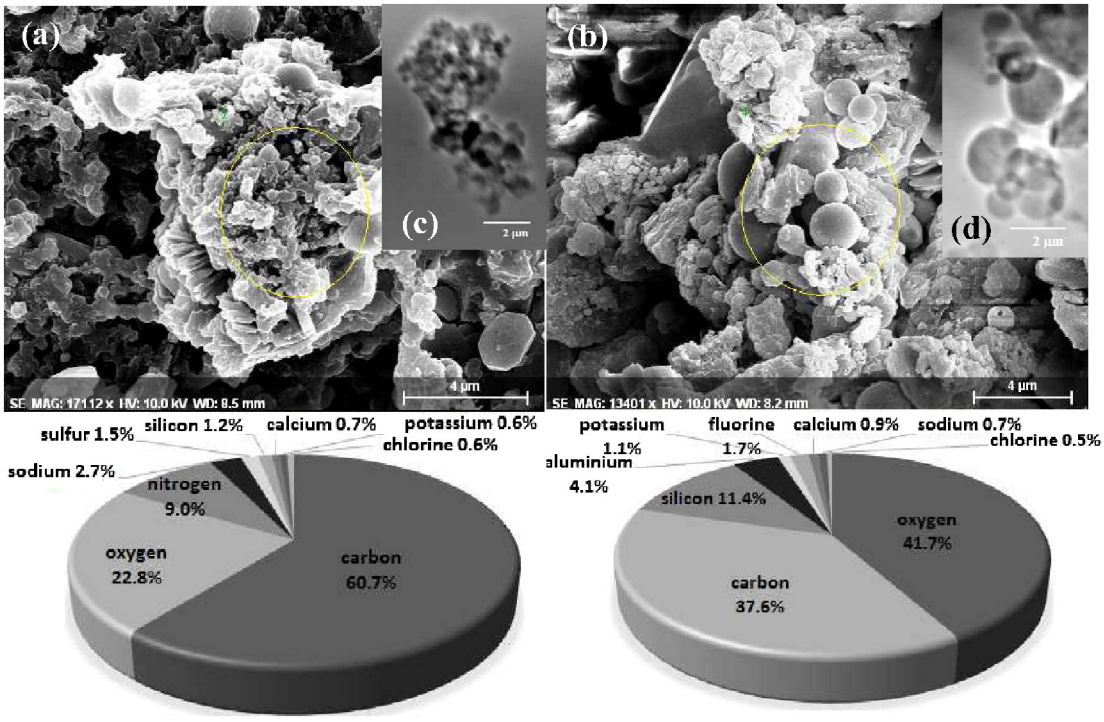

Fig. 5. The SEM images for (a) $PM_{2.5}$ and (b) coarse particles with carbon-rich components (middle of image) in the winter samples. The XNI images of (c) and (d) showed soot-like aggregates and chain structures, respectively.

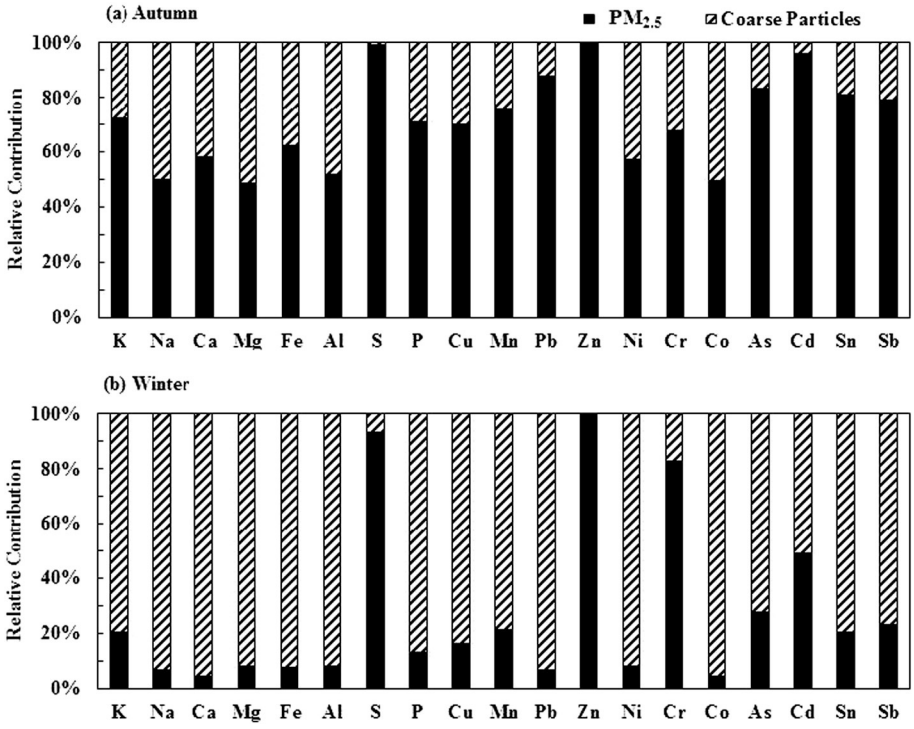

Fig. 6. Relative contribution of $PM_{2.5}$ and coarser airborne particles to metal conveyance: (a) autumn, and (b) winter.

and sea spray are comparatively rich for the coarse airborne particles, which is consistent to the surface chemistry based on the TOF-SIMS analysis. Relatively high proportion of Cr, Co, and Ni (>30–50%) with abundance of mineral phases in the coarse particles provides additional evidence that these elements are derived from the crustal origins (Lee et al., 2013).

The winter-sampled $PM_{2.5}$ particles, in contrast, contain lower mass concentrations for most of the measured elements rather than the coarse particles (Fig. 6(b)). Only sulfur and zinc elements were strongly related to the $PM_{2.5}$ particles regardless of seasonal variation. The rich sulfur contents (400–1800 ng m^{-3}) indicate abundant fine components formed by gas-to-particle conversion processes in the atmosphere. A main source for the zinc concentrated to fine particles (20–100 ng m^{-3}) could be local traffic exhaust emissions in the study area rather than tire wear and re-suspended road dust because of their size fractions (Councell et al., 2004; Thorpe and Harrison, 2008). The great mass concentrations of heavy metals except zinc in the coarse particle sample imply that coarse particles act potentially as an important medium to convey heavy metals to ecological and environmental systems. We posit that the abundant carbon-rich particles may contribute significantly to the metal contents in the coarser fractions of the winter samples.

3.5. Environmental implication

The water solution soaked by the $PM_{2.5}$ sample, interestingly, showed acidic condition, but slightly alkaline for the coarse particles. Deposition of abundant $PM_{2.5}$ particles to soil environment may be concerned with soil acidification in a Korean local area, because the atmospheric concentrations of $PM_{2.5}$ could be increased gradually through industrialization and urbanization processes in the neighboring countries. The acidic condition facilitates increasing solubility and mobility of toxic heavy metals, and causes environmental hazards in ecological system.

4. Conclusions

The physical and chemical properties of ambient fine and coarse aerosols were determined in the autumn and winter seasons at the Korean suburban area. The $PM_{2.5}$ particles contain abundant organic carbon fractions with sulfates and nitrates for both autumn and winter samples. The main components of coarse particles are naturally occurring earth constituents such as suspended soil dust and rock fragments. In particular, spherical-shaped carbonaceous materials (i.e., black carbon, fly ash, tar ball) were also observed for the coarse particles, and more frequently observed for the winter sample than autumn one. High contents of heavy metal in the coarse particles collected in the winter could be associated with these carbonaceous materials.

This study showed the relative importance of fine and coarse particles as conveyance medium of heavy metals at the cold season. The ambient airborne aerosols must be properly monitored according to spatial and temporal characteristics with meteorological conditions to reduce environmental risk in a local area.

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Appendix A. Supplementary data

The Supplementary data included Figure A1. Sampling location, Figure A2. SEM images with EDX analysis for (a) $PM_{2.5}$, (b) Coarse particle sample (winter), Table A1. Daily average weather conditions during sampling time, and Table A2. Measured elemental concentrations for fine and coarse particles. The Supplementary data associated with this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.10.021

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