Asian Journal of Atmospheric Environment
Vol. 11, No. 2, pp. 122-130, June 2017
doi: https://doi.org/10.5572/ajae.2017.11.2.122
ISSN (Online) 2287-1160, ISSN (Print) 1976-6912

Chemical Properties of Indoor Individual Particles Collected at the Daily Behavior Spaces of a Factory Worker

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ABSTRACT

The main purpose of the study was to clarify the properties of individual particles collected at each behavior space of a factory worker. The samplings of size-segregated ($PM_{2.1-1.1}$ and $PM_{4.7-3.3}$) indoor particles were conducted at three different behavior spaces of a factory worker who is engaged in an auto parts manufacturing plant (i.e., his home, his work place in factory, and his favorite restaurant). Elemental specification (i.e., relative elemental content and distribution in and/or on individual particles) was performed by a micro-PIXE system. Every element detected from the coarse particulate matters of home was classified into three groups, i.e., a group of high net-counts (Na, Al, and Si), a group of intermediate net-counts (Mg, S, Cl, K, and Ca), and a group of minor trace elements (P, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, and Pb). The results of EF for $PM_{4.7-3.3}$ in home indicated that several heavy metals were generated from the sources within the house itself. An exceptional feature shown in the individual particles in workplace is that Cr, Mn, and Co were clearly detected in both fine and coarse particles. Cluster analysis suggested that the individual coarse particles ($PM_{4.7-3.3}$) collected at the indoor of factory were chemically heterogeneous and they modified with sea-salt, mineral, and artificially derived elements. The principal components in individual coarse particles collected at restaurant were sea-salt and mineral without mixing with harmful trace elements like chromium and manganese. Compared to the indoor fine particles of home and restaurant, many elements, especially, Cl, Na, Cr, Mn, Pb, and Zn showed overwhelmingly high net-counts in those of factory.

Key words: Indoor air pollution, Individual particle, Chemical property, Micro-PIXE, Element, Factory worker

1. INTRODUCTION

Although individual variation exists, people spend most of their day indoors. A recent study found that Americans and Canadians spent an average of 90% and 93% of their life indoors, respectively (Pretty *et al.*, 2005). Spending so much of our time inside can cause huge harmful effects on our health and wellbeing. Many studies show that the indoor air in the average home can be up to 100 times more polluted than outdoor air (Rebecca *et al.*, 2004; Mooney and Nicell, 1992). The worst indoor air quality is manily caused by almost airtight buildings do not provide sufficient ventilation to remove indoor air contaminants. The diversification of our cultural life is one of the biggest contributors to indoor pollutants such as pets, tobacco smoke, furniture, fireplaces, ceiling coverings, building materials, household cleaning products, humidifiers, cockroach waste, and so on.

For workers, meanwhile, the work environment is a very important factor for their health status. There have been numerous studies showing the relationship between the particular occupational diseases and work environment. Bahrami and Mahjub (2003) compared lung function among workers in stone-grinder factories emitting high concentration of silica particles and reported the prevalence of respiratory symptoms in stone-grinders was higher than other workers. Chronic exposure to cement dust has been reported to lead to greater prevalence of various clinical conditions by Manjula *et al.* (2013). Kraus *et al.* (2001) pointed out that tungsten and cobalt in a plant producing hard metals might be responsible for both fibrogenic and carcinogenic effects.

So far, numerous studies on the indoor air quality for gaseous and particulate materials have been reported (Kanatani *et al.*, 2014; Hunt and Johnson, 2012; Kagi *et al.*, 2007; Sakai *et al.*, 2004). However, in the case

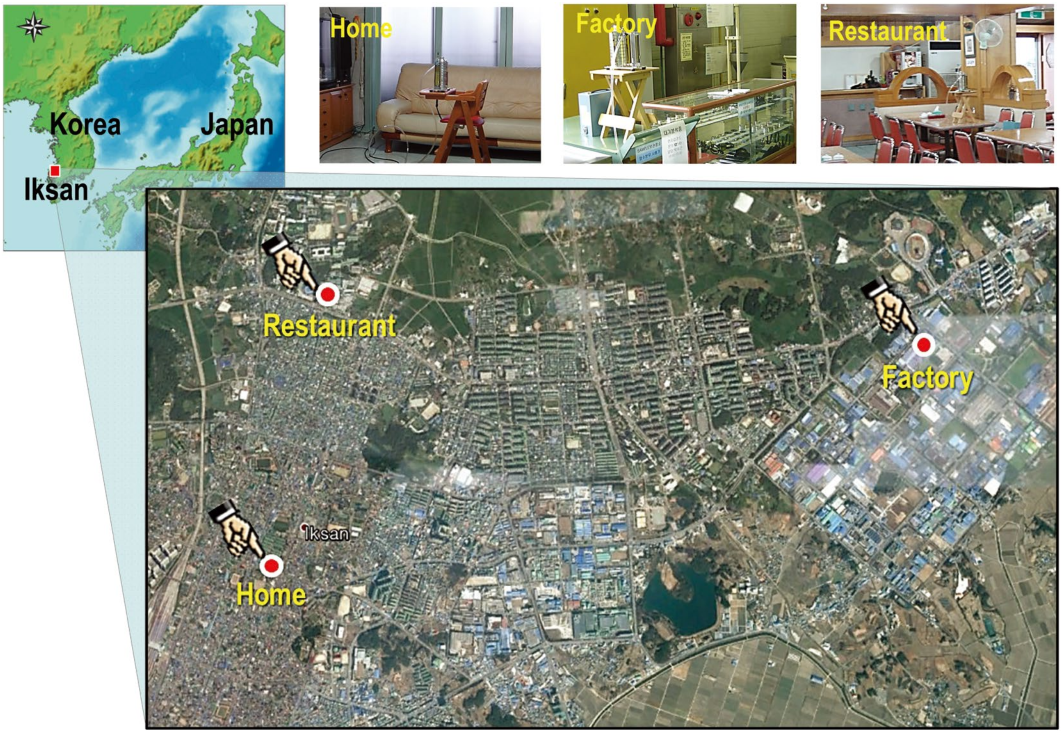

Fig. 1. Locations of the selected three behavior spaces of a factory worker in Iksan, Korea.

of particulate matter (hereafter called "PM"), the subjects of study have nearly always been placed on its mass and number concentrations. As a matter of fact, the elemental components of indoor PM, especially in terms of individual particles, have seldom been evaluated with respect to both their PM size-dependent health hazards and and the characteristics of personal living space.

In light of this situation, the present study was designed to clarify the properties of individual PM collected at each behavior space of a factory worker.

2. MATERIALS AND METHODS

2.1 Site Description

Samplings of indoor PM were conducted at three behavior spaces of a factory worker working in Mando Corp., i.e., the living room of his house (35.946N; 126.955E), the target room in factory (35.960N; 127.008E), and a restaurant (35.964N; 126.958E) using mainly on weekdays (see Fig. 1).

His all daily life was made within Iksan city. Iksan city is located in the southwestern part of South Korea, and it is a modern industrial city and headquarters to numerous factories producing textiles, jewelry, metal, electrical products, and machinery. Mando Corp. completed an auto parts (e.g., steering/suspension system) manufacturing plant in Iksan city in september of 1995. His living room with tobacco smoke-free, fireplace-free, and moderately maintained floor, wall/ceiling coverings, and furnitures was selected as the place of home measurement. The popular restaurant he was using mainly on weekdays was equipped with an wood interior, an open kitchen with wood cabinet and modern appliances, wood tables, plastic cover chairs, and tiled floor.

2.2 Particle Collection

An intensive field survey of PM was conducted at each site described above. For the sampling of PM as a function of its size, an Andersen air sampler (Tokyo Dylec Co., AN-200) was operated at each site (see Fig. 1). PM was collected onto a 80 mm diameter Nuclepore® filter at the 3rd and the 5th stages. Sampling duration time was 24 h and the airflow was maintained at approximately 28.3 L min^{-1}. The 50% cut-off sizes of the 3rd and the 5th stages are 3.3 and 1.1 μm, respectively.

After sampling, the filters were placed in clean sterilized petridishes. And then each petridish was sealed with Teflon tape and wrapped with aluminum foil and was placed in a cold storage bag during air transportation. Blank filters were handled in the same manner as the samples.

2.3 Chemical Specification of Trace Elements and Their Distribution in Individual PM

Elemental specification (i.e., relative elemental content and distribution in and/or on individual particles was performed by the micro-PIXE system equipped at the facilities of the TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) in Japan atomic energy research institute.

Micro-PIXE relatively recently debuted by the combination of PIXE analysis and the microbeam technique is one of the most powerful micro-analytical techniques having an excellent sensitivity. Minimum Detection Limits (MDL) (for a typical 3- to 4-minute analysis of an accumulated charge of 2.5 microcoulombs) are in the range of sub-ppm-10 ppm, with precisions of 1-10% for a surface layered sample. Quantity of an element in a sample can be measured by counting the number of characteristic X-rays. The X-ray yield for the characteristic X ray of element A (Y_A) can be calculated by following equation:

$$Y_A = N_A \times Q \times d\Omega \times E \times R_a \times \sigma^x \times 4\pi S^{-1} \quad (1)$$

where N_A is the number of element A in sample, Q is the total number of incident particle, $d\Omega$ is a solid angle, E is detecting rate of detector, R_a is the absorption rate of characteristic X-ray, σ^x is a section area of characteristic X-ray generation, S is beam spot, respectively.

The overview of the analysis facility and analytical principle/conditions of TIARA's micro-PIXE can be referred to other papers (Ma, 2010; Sakai *et al.*, 2005).

By means of micro-PIXE analytical technique, a total of 350 particles randomly sectected from each coarse and fine PM collected at three different sampling sites were analyzed.

3. RESULTS AND DISCUSSION

3.1 Individual Coarse PM Collected at Home

Since the particles often overlap and form clusters on the impaction media of Andersen sampler, the edge portion of a particle conglomeration was the target of micro-PIXE analysis.

An example of micro-PIXE X-ray spectrum (top) and elemental maps (bottom) for several individual particles in $PM_{2.1-1.1}$ is shown in Fig. 2. It was possible to draw obviously separated elemental spectra from the micro-beam scanning of a $10 \times 10\ \mu m^2$ area. Corresponding to micro-PIXE imaging with scanning transmission ion microscopy, it also enables us to obtain the distributions of trace elements of individual particles. Ca, Cl, P, and Fe were principally detected in and/or on several individual particles and their localizations were varied in each particle. From the visualized elemental distribution for individual particles, it is possible to know the particle-to-particle variation in composition.

The number of X-ray counts for each element of individual particles and filter background at the region of interest (ROI) (i.e., energy range of the target element) could be calculated by normalizing with the dose (nC) of 3.0 MeV H^+ microbeam. Fig. 3 shows the box plots (left) and scatter grams for several selected elements (right) of micro-PIXE elemental net-counts of individual particles in $PM_{4.7-3.3}$ collected at a home.

On the basis of box plots of elemental net-counts, every element was classified into three groups, i.e., a group of high net-counts (Na, Al, and Si), a group of intermediate net-counts (Mg, S, Cl, K, and Ca), and a group of minor trace elements (P, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, and Pb). Among a group of high net-counts, Si shows a preponderantly high net-count compared with other two elements. In view of the particle size, it is suggested that the mineral elements such as Mg, Al, Si, K, and Ca were originated from soil.

Household PM may be accumulated from both internal and external sources over long periods of time. Soil particles can be easily drawn into the indoor from outside by one's shoes, especially in the case of no partition between the front door and the living room. Na and Cl showing high interrelationship with 0.85 R^2 level were considered to be of marine origin because the target home is geographically close to the sea, A good correlation ($R^2 = 0.68$) shown between Co and Fe was attributable to the fact that cobalt and iron were chemically alike in many respects and occur together in many geologic materials (Kubota, 1958).

One of high-hazard characteristic sources in home is a coat of paint on the walls. If people live in an older home, many of paint chips get pulverized to microscopic particles that become part of the interior dust people breathe (Salo *et al.*, 2008). In the present study, to classify the paint origin into non-paint sources for the elements in individual coarse particles collected at a home, the enrichment factors (EF) was applied. As a simple means to assess the sources of elements in PM, the EF has long been used (Reimann and Caritat, 2000).

Although the non-lead paint is widely used recently, Ti and Ca are known as its major components. Fe and S are also the trace element found in lead-free paint (Aichi electrical engineer, 2008). EFs of several elements including Ti, Ca, Fe, and S were calculated by the following equation.

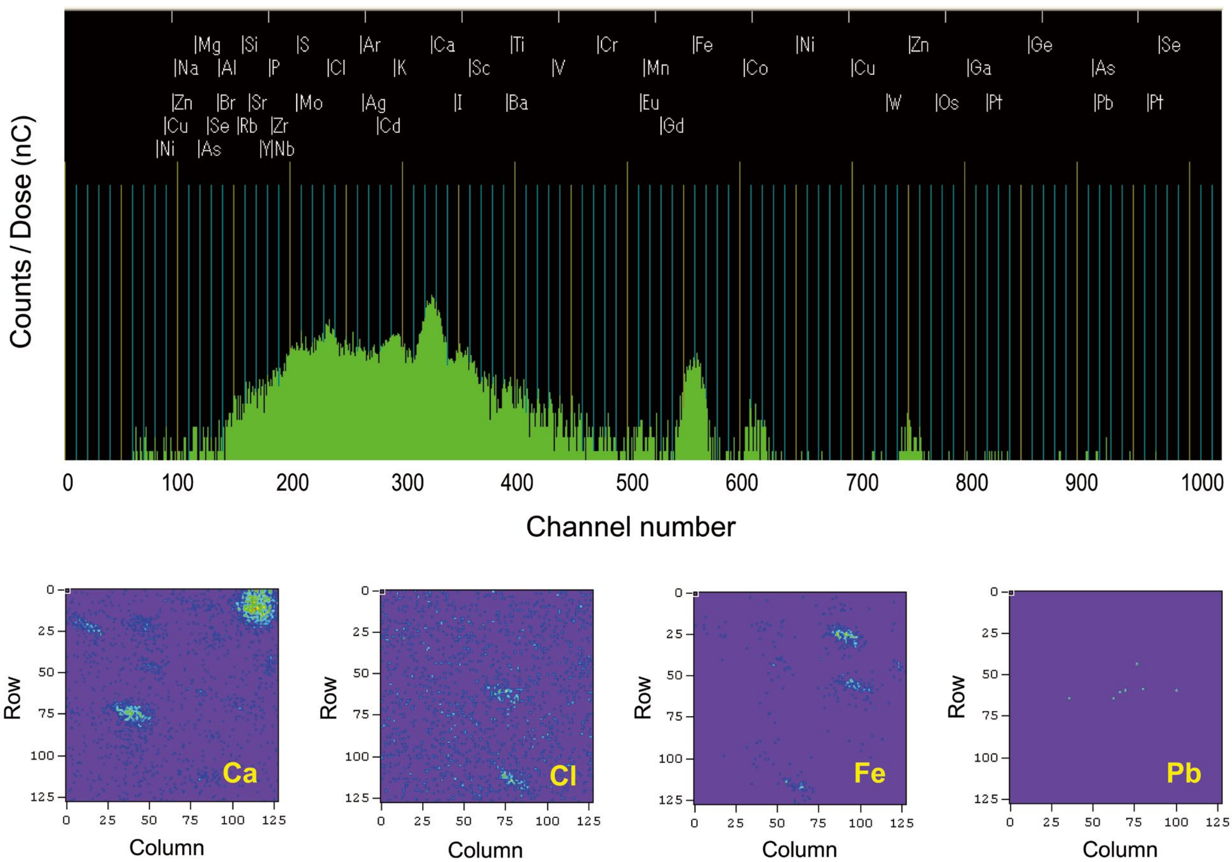

Fig. 2. A micro-PIXE X-ray spectrum (top) and elemental maps (bottom) drawn from several particles ($PM_{2.1\text{-}1.1}$) collected at a home.

$$EF_x = \frac{n - C_{x,p}/n - C_{ref,p}}{C_{x,paint}/C_{ref,paint}} \quad (2)$$

where, $n - C_{x,p}$ and $n - C_{ref,p}$ are the net-counts of x element and the referantial element in indoor particles, respectively. $C_{x,paint}$ and $C_{ref,paint}$ are the concentrations of x element and the referantial element in paint, respectively.

As the suitable referential marker for paint source, Ti was selected in this study. The relative concentrations of elements in lead free paint were based on the data reported by Aichi electrical engineer (2008). Fig. 4 shows the EFs of individual coarse particles collected at a home. The highest EF was shown at Si and relatively high EFs were found at P, S, and Ca, while Fe and Zn showed very low EFs. The fact that Fe is one of trace elements in lead-free paint is probably why its EF was very low. As mentioned above, although Ca and S are also detected lead-free paint, the results of EF indicate that they were not originated lead-free paint.

Rasmussen *et al.* (2001) investigated the element profiles of indoor dust versus exterior soils from 50 residences located in Ottawa, Canada and reported that the level of heavy-metal contamination in household dust was higher than that in garden soil. The results of EF therefore suggest that several heavy metals like Fe and Zn can be generated from the sources within the house itself including paint.

3.2 Identification of Coarse PM Properties of Work Place

In general, it is expected that larger particles are more easily generated than small ones in industrial work places like auto parts manufacturing plant and electronic waste recycling facility. Kim *et al.* (2015) reported that the mass concentration of coarse particle ($PM_{10\text{-}2.5}$) was 3-4 times more abundant than those of fine/ultrafine PM (<2.5 μm) in electronic waste recycling facility.

Fig. 5 shows the box plots and scatter grams of micro-PIXE elemental net-counts of individual coarse particles ($PM_{4.7\text{-}3.3}$) collected at the indoor of Mando Corp.

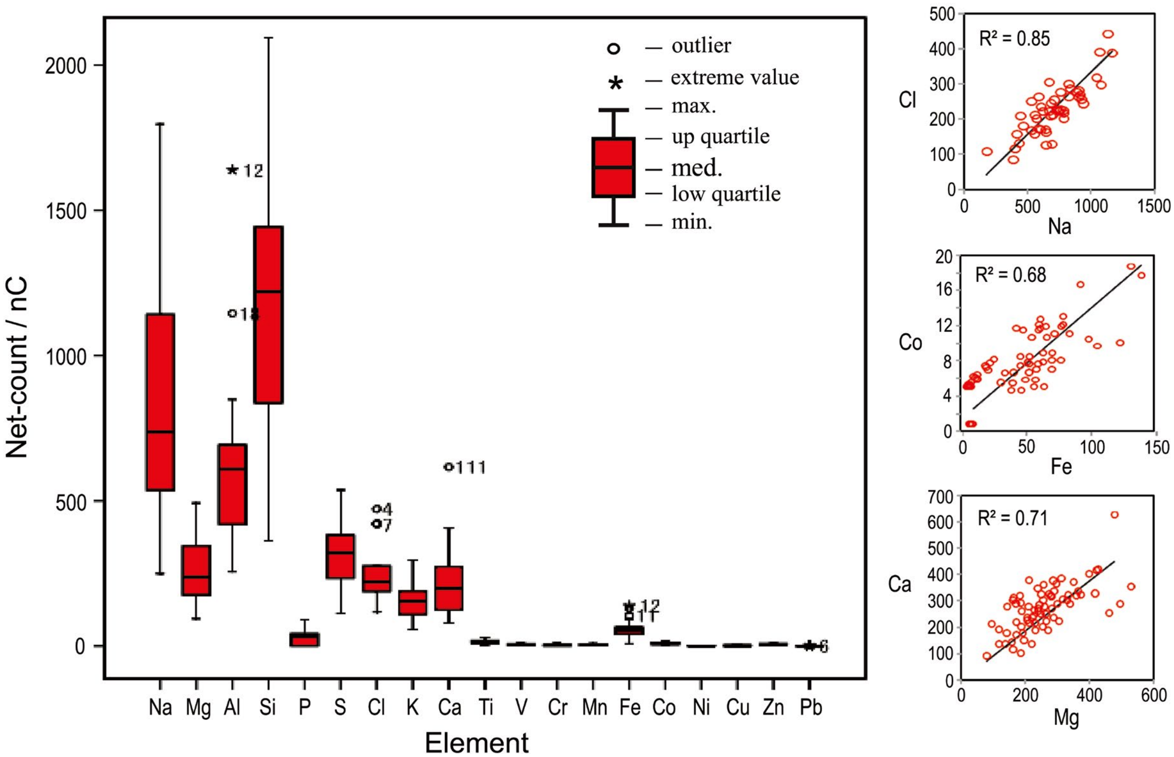

Fig. 3. Box plots (left) and scatter grams (right) of micro-PIXE elemental net-counts of individual particles ($PM_{4.7\text{-}3.3}$) collected at a home.

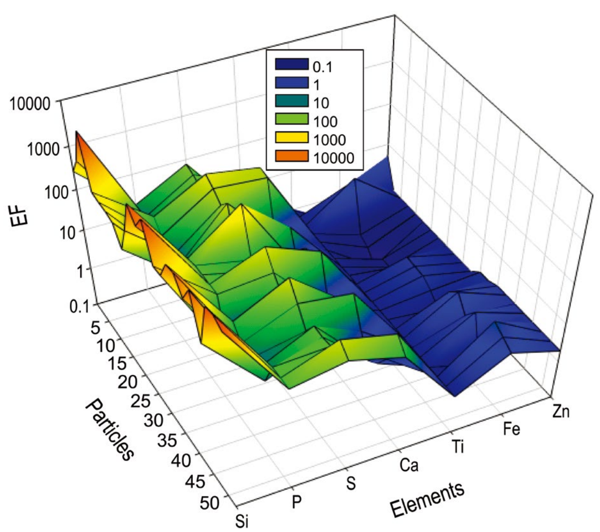

Fig. 4. Enrichment factors of individual coarse particles ($PM_{4.7\text{-}3.3}$) collected at a home.

Compared to the same size range PM collected at home, one of unusual points is that the net-counts of mineral components were none too high. The strict measure not carrying exterior soil dust into indoor workplace might have led to this result. The correlation plot between Na and Cl and those net-counts indicate that sea-salts, on the other hand, might not be completely blocked. High elemental net-counts of Na and Cl also suggest that their other sources might exist in indoor workplace. Another exceptional feature shown in the individual PM in workplace is that Cr, Mn, and Co clearly formed their ne-counts. Among them, a significant correlation was found between Cr and Mn ($R^2 = 0.99$). Co net-counts were found to correlate more highly ($R^2 = 0.97$) with the Fe in PM of workplace than that in PM of home ($R^2 = 0.68$). Ibanez *et al.* (2010) suggested that although, the bioaccessibility of heavy metals depends on chemical speciation and particle size, those of Cr, Mn, and Co are 10-50%, 40-60%, and 15-50%, respectively. The health effects resulting from breathing air containing chromium (chromium(III) and chromium(VI)) particles are well known. Long-term deposition of high levels (100 to 1,000 times higher than those found in the natural environment) in lung has been associated with lung cancer in steel workers (Toxicological profile for chromium, 1992).

In order to classify individual particles collected at the indoor of factory, the micro-PIXE elemental net-

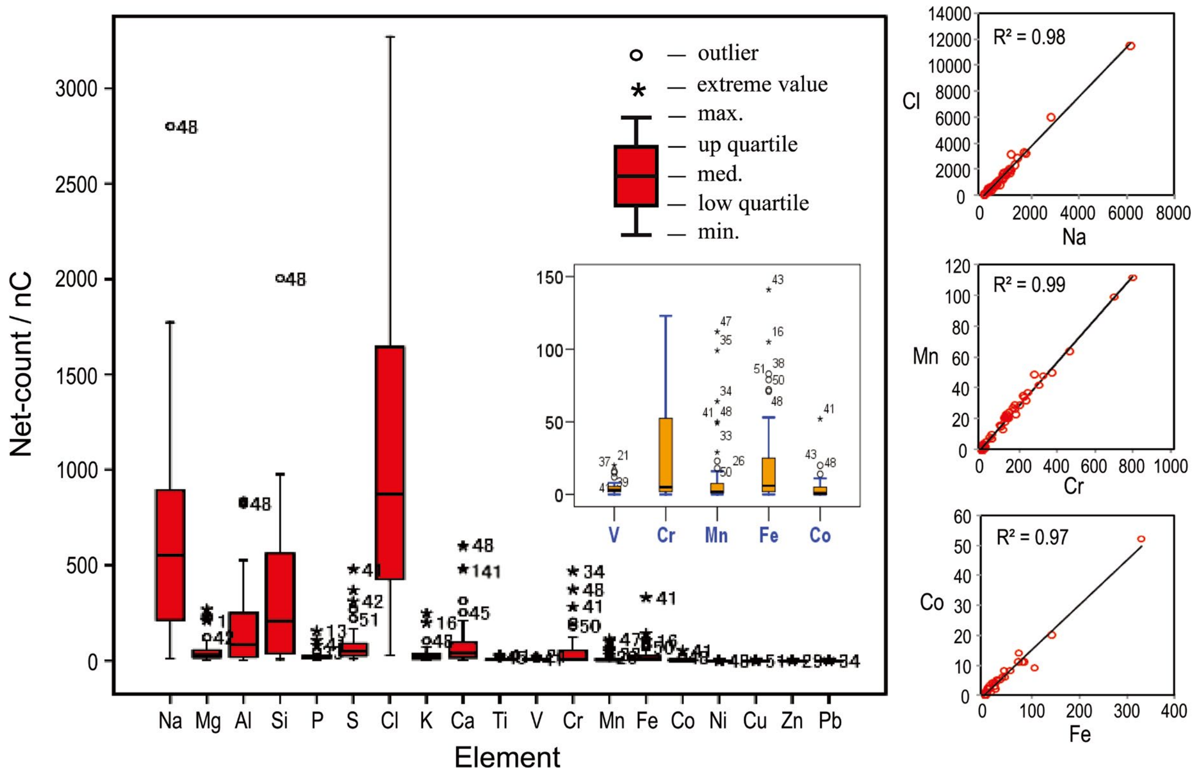

Fig. 5. Box plots (left) and scatter grams (right) of micro-PIXE elemental net-counts of individual particles ($PM_{4.7\text{-}3.3}$) collected at the indoor of factory.

Table 1. Categorized individual particles ($PM_{4.7\text{-}3.3}$) at the indoor of factory by cluster analysis.

	Characteristic components in cluster	% of the total particle number
Cluster 1	sea-salt/mineral ≫ sulfur ⋙ trace*	73.5
Cluster 2	sea-salt ≫ mineral/sulfur > trace	18.4
Cluster 3	sea-salt/mineral + trace	8.1

*Co, Cr, and Mn

counts were subjected to cluster analysis. The agglomeration scheduled hierarchical cluster analysis can identify relatively homogeneous groups of variables based on selected characteristics. In this study, the individual particles that all components (from Na to Pb) were detected were the subject of the cluster analysis. Table 1 summarizes the result of cluster analysis for the individual coarse particles ($PM_{4.7\text{-}3.3}$) at the indoor of factory. It shows that individual particles were categorized into three clusters. Cluster 1 with a large portion of total particle population (73.5%) was found to be the particles with sea-salt/mineral ≫ sulfur ⋙ trace (Co, Cr, and Mn). Cluster 2 with a small account rate of total particle population (18.4%) was showing sea-salt ≫ mineral/sulfur > trace (Co, Cr, and Mn). Cluster 3 with a poor particle number (8.1%) had high mass for sea-salt and mineral with coexisting of trace elements (Co, Cr, and Mn). Clustering of individual particles suggests that the individual coarse particles ($PM_{4.7\text{-}3.3}$) collected at the indoor of factory were chemically heterogeneous and most particles could be clustered with highly dissimilar dendrogram which expresses the modification of individual particles with sea-salt, mineral, and artificially derived elements.

3.3 Individual Coarse PM Collected at Restaurant

Compared to labor and sleep, even though mealtime is short, it is meaningful to investigate PM properties collected at restaurant used by a factory worker in a regular every day. Since the popular restaurant was equipped with an open kitchen with the most thorough

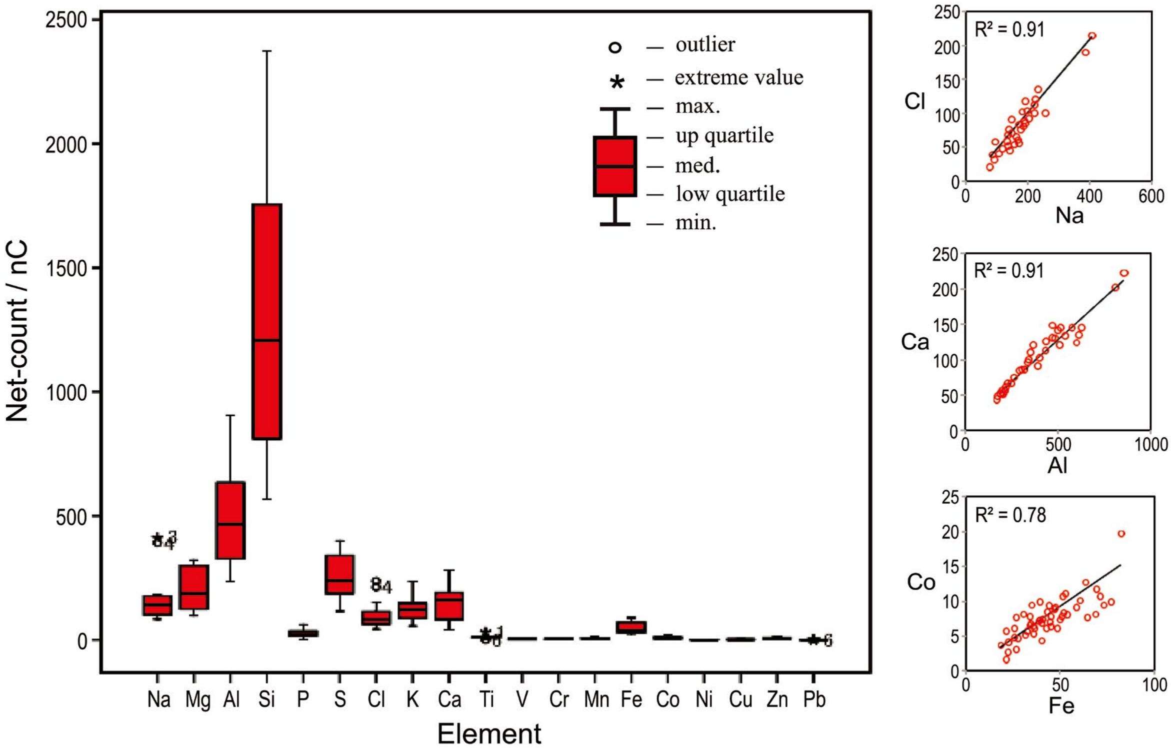

Fig. 6. Box plots (left) and scatter grams (right) of micro-PIXE elemental net-counts of individual particles ($PM_{4.7\text{-}3.3}$) collected at the indoor of restaurant.

ventilation, it can be thought that PM were mainly the externally inflowed coarse PM due to customer access.

Fig. 6 shows box plots and scatter grams of micro-PIXE elemental net-counts of individual coarse particles ($PM_{4.7\text{-}3.3}$) collected at the indoor of restaurant. A notable feature is the enrichment of mineral components like Si, Al, and Ca, Na and Cl, as the next major components, were presumably derived from sea-salt. Their interrelationship was very high with 0.91 R^2 level. The scattering plot between Co and Fe shows a significant positive correlation ($R^2=0.78$). As a result, the principal components in individual PM collected at restaurant were sea-salt and mineral without mixing with harmful trace elements like Cr and Mn.

3. 4 Individual Fine PM Collected at Three Indoor Sites

Due to the limitations of current micro-PIXE technologies for elemental quantification, the sufficient assessment of the absolute elemental amount (e.g., $\mu g\ m^{-3}$ or $\mu g\ m^{-2}$) according to the PM sampling place could not be performed in the present study. However, the valued elemental net-count of individual PM at high sensitivity will be very useful indicator for the evaluation of the PM characteristics of each indoor place.

Fig. 7 shows the comparison of micro-PIXE elemental net-counts of individual fine particles ($PM_{2.1\text{-}1.1}$) collected at three indoor locations. The elements related to marine and soil still tend to have a overwhelmingly higher net count than others at all three sites. Among them, the net-counts of Cl and Na appeared unusually high in the PM collected at the indoor of factory. Owing to geographical features of sampling locations, it is easily taken into account that sea-slats will have a strong impact on the elemental composition in the indoor PM. Add to this, sodium carbonate used as machine dishwashing products and surface cleaners might have contributed to the unusually high net-counts of Cl and Na because its impurities may include NaCl, Na_2SO_4, $CaCO_3$, $NaHCO_3$, and Fe. Among various heavy metals, the net-count of Cr in the PM collected at the indoor of factory was noticeably higher than those of two other indoor sites. Cr ($ZnCr_2O_7$ and Cr_5O_{12}) is used as a corrosion-resistant coatings on numerous vehicle components. A thorough measure is needed in terms of workers' health hazards because the elemental Cr and Cr(VI) can have adverse human health effects (Jacobs and Testa, 2004). In addition, several trace elements, including Pb and Co, also showed high net-counts in the PM collected at factory. Pb as an alloy in Al widely used in wheel rims, engine parts, and win-

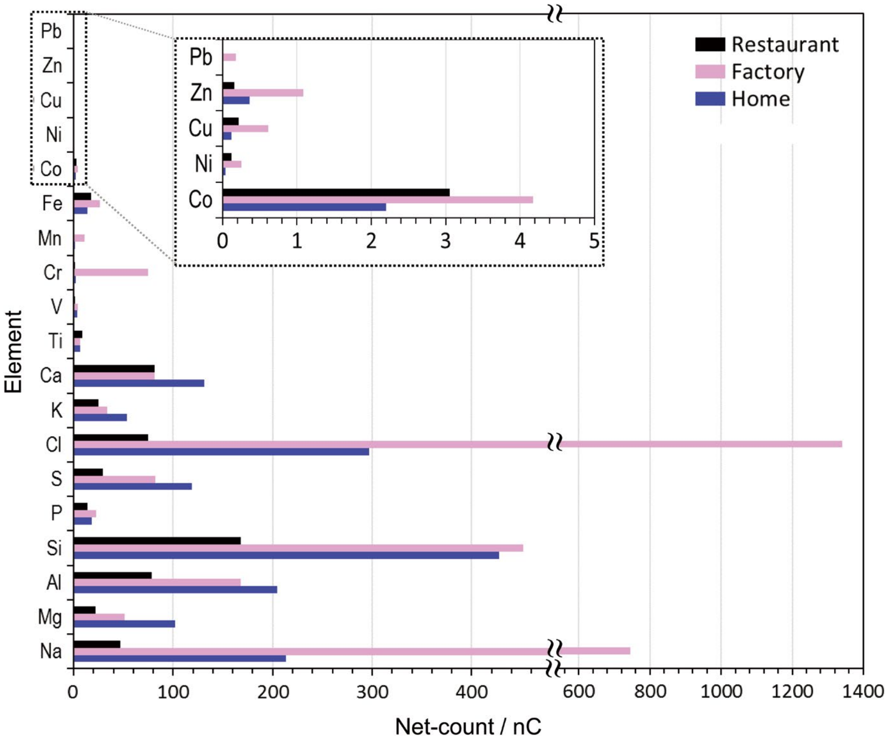

Fig. 7. Comparison of micro-PIXE elemental net-counts of individual fine particles ($PM_{2.1\text{-}1.1}$) collected at three indoor locations.

dow levers. Co is a metal used in corrosion- and wear-resistant alloys and it is also used to make airbags in automobiles (Cobalt statistics and information, 2017).

Comparing restaurant and home indicates that relatively light elements (from Na to Ca) were more enriched in the PM of home. Meanwhile, in the case of some toxic heavy metals (e.g., Co, Ni, and Cu), the elemental net-counts were much higher in the PM of restaurant than in those of home. If a factory worker breathe the same amount of particles from home and factory in his daily life, the PM of factory containing much more harmful heavy metals have far greater influence on his health.

4. CONCLUSION

As previously stated, the indoor air quality at each behavior space of worker's daily life has become an important occupational health and safety concern. The present study was designed to clarify the chemical properties of indoor individual particles collected at the daily behavior spaces of a factory worker. A comparison of the elemental net-count for the fine particles captured at three indoor sites can be summarized as follows:

Many harmful heavy metals including Pb, Co, and Cr showed a relatively high net-count in the factory. Among them, the net-count of Cr was noticeably higher than those of two other indoor sites. In comparison with home, the elemental net-counts of several toxic heavy metals (e.g., Co, Ni, and Cu) were much higher in the PM of restaurant. Although, we cannot obviously conclude whether a possible health risk exists at three different behavior spaces of a factory worker, it can be said with some confidence that some kinds of particles generated from sources within the indoor itself can lead to exposures to hazardous substances.

ACKNOWLEDGEMENT

This paper was supported by Wonkwang Health Science University in 2017.

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(Received 6 April 2017, revised 2 June 2017, accepted 6 June 2017)