Source apportionment for indoor PM2.5 and elemental concentrations using by a positive matrix factorization and an instrumental neutron activation analysis

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1. Introduction

Airborne particulate matters, especially the PM2.5 (aerodynamic equivalent diameter, AED, less than 2.5 μm) fraction has been important. This is because of their potential for deposition on to the human respiratory system being accompanied by many harmful trace metals (such as As, Cd, Cr, Cu, Mn, Pb, Se, and Zn) [1]. The indoor air quality has become a great concern since late 1980s, because the population spends a majority of their time in various indoor environments [2]. The indoor particulate matter may be influenced from outdoor environment and indoor sources such as environmental tobacco smoke (ETS), combustion devices, cooking, etc. In this study, we undertake the measurements of about 26 elements using instrumental neutron activation analysis (INAA). Based on our measurement data, we characterize concentration status and mutual relationship between indoor and adjacent outdoor air quality. Next, sources at indoor/outdoor environment were identified and the contributions of each source were quantified by positive matrix factorization (PMF).

2. Experimental

2.1 Sampling site and sampling

A total 120 of fine airborne particulate matter, PM2.5 (aerodynamic diameter less than 2.5 μm), was collected during 2008 with low volume air sampler at indoor environment and their adjacent outdoor environments in Daejeon, Korea. Although the sampling site surrounded in huge residential and commercial area, the study area could be characterized to a roadside site due to having most heavy traffic volumes in Daejeon city. Thus it can represent one of the strongest traffic-related pollution sources in the city. For the collection of indoor and outdoor PM2.5, a mini-volume air sampler (version 4.2, Airmetrics, USA) and annular denuder air sampler (URG, 3000C model) with polycarbonate filter (47mm, 0.4 μm pore size, Nuclepore) were used, respectively.

2.2 Elemental analysis

For the INAA determination of Al, As, Ba, Br, Ce, Co, Cl, Cr, Cu, Cs, Fe, I, In, K, La, Mg, Mn, Na, Sb, Sc, Sm, Se, Th, Ti, V, and Zn, the PM2.5 samples were irradiated using thermal neutrons using the Pneumatic Transfer System (PTS, Φ_{th} = 2.95 x 10^{13} $cm^{-2}s^{-1}$, R_{cd}= 250) at the HANARO research reactor at the Korea Atomic Energy Research Institute. The measurements were carried out using a high-purity Ge detector with a relative efficiency of 25%. This measuring system has a resolution of 1.9 keV (FWHM) at 1332.5 keV of ^{60}Co with a peak-to-Compton ratio of 45:1.

2.3 Positive matrix factorization [3]

PMF is a new model that addresses such negative value problems by restricting the common factor and factor loading to have only a positive value, considering the standard deviation of measured data. Basic equation of the PMF model is shown equation (1). The method is to obtain the unknown matrix, G and F by the solution of a least square method iteratively:

$$X = GF + E \qquad (1)$$

$$Q(E) = \sum_{i=1}^{m} \sum_{j=1}^{n} (e_{ij} / s_{ij})^2 \qquad (2)$$

Where, X(m×n) is the data matrix consisting of the m chemical components analyzed in n samples, G(n×p) is the source contribution to the each sample. F(p×m) is the matrix of source profile. E presents the residual matrix of calculation, and the main process of the PMF is minimizing the Q-value, which is defined in the equation (2) below as the sum of square of the residuals (eij) weighted inversely with error estimates (sij) of the data point.

3. Results and discussions

3.1 PM2.5 and elemental concentration

The average concentration of PM2.5 was enriched by 1.5 times at an indoor environment (47.6 ± 16.5 μg/m^3) than outdoor environment (29.0 ± 13.3 μg/m^3). From the results of the quantitative analysis for indoor/outdoor PM2.5 samples by INAA, it was found that the concentration of elements associated with crustal sources (such as Al, Fe, and K) were much higher than that of any other toxic elements. The distribution patterns of different elements were clearly distinguished with their concentrations ranging across four orders of magnitude.

Based on a simple comparison of the elemental concentrations by their magnitude, the data sets could

be grouped into five different categories: 1)<10^{-1} ng/m^3: In, Sc, and Th; 2)<10^0 ng/m^3: Cs, Co, Sm, and La; 3) 10^1 ng/m^3: As, Br, Cr, Cu, I, Mn, Sb, Se, and V; 4)<10^2 ng/m^3: Ba, Cl, Mg, Ti, Zn; and 4)<10^3 ng/m^3: Al, Fe, K, and Na.

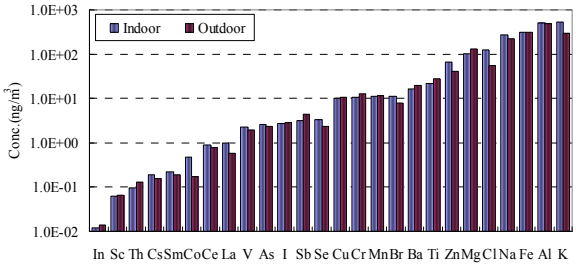

Fig. 1. Logarithmic distribution of metal concentration in indoor and outdoor environment.

The concentration ratios of Fe, Al, Sm, Na, V, Cs, Br, As, La, Se, K, Zn, Ce, Cl, and Co between indoor and outdoor environment were over than 1.3; especially, Cl(3.8) and Co(4.0). Those were distributed as higher portion in the ETS showed higher ratios, which suggests that ETS is a distinctive increasing factor of PM2.5 and elemental concentration at indoor environment. A regression analysis was also conducted for the data sets determined from indoor/outdoor environments. The results of analysis allow us to predict their mutual influences for the elements originated from specific sources. The higher determination coefficients imply that origins of specific species are influenced each other. The determination coefficients of soil-derived elements such as Al, Cr, Mg, Na, and Sm between the indoor and outdoor environments were over than 0.7.

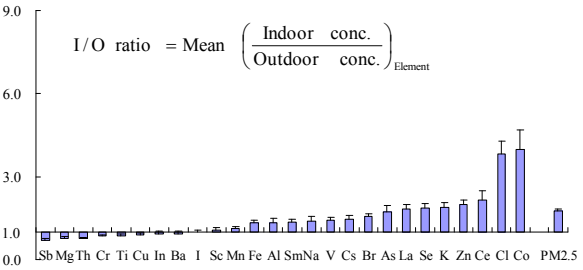

Fig. 2. Concentration ratio of PM2.5 and elements between indoor and outdoor environment.

3.2 Source apportionment

The sources were classified into five and four categories by using PMF based on PM2.5 and elemental data collected in indoor and outdoor environments, respectively. The results of the correlation analysis using the observed versus predicted PM2.5 mass concentrations indicated that the resolved factors effectively accounted for the total mass. Correlation coefficients between the reconstructed and measured portions of three data sets were about 0.80 for both data sets. On average, the extracted factors from the data accounted for 99% and 95% with respect to the corresponding measured PM2.5 concentrations, respectively.

The sources associated with the transportation were divided into road dust and vehicle exhaust. The former was defined as contaminated soil dust on the pave road by vehicle exhaust, particles created through tire, brake, and vehicle wear process as marker of Ba, Cr, Sb, and Ti [4]. On the other hand the latter ones were vehicle exhaust directly emitted from tailpipes as marker of Br, Cl, Sb, and Zn. The elements originating from ETS (like Br, Cl, Co, K, La, Se, and Zn) were used for the identification of ETS source for indoor PM2.5.

The five dominant sources in the indoor environment were estimated and the contribution of each source was quantified by PMF receptor model; road dust (12.0±8.7 μg/m^3, 26%), oil fuel combustion + indoor activities (11.7±9.3 μg/m^3, 26%), vehicle exhaust (10.3±10.3 μg/m^3, 23%), environmental tobacco smoke (7.7±8.2 μg/m^3, 17%), and soil dust (3.6±3.6 μg/m^3, 8%). In the outdoor environment, however, the four sources were apportioned; vehicle exhaust (12.0±5.3 μg/m^3, 46%), road dust (6.1±5.9 μg/m^3, 24%), oil fuel combustion (4.8±4.9 μg/m^3, 19%), and soil dust (2.9±2.8 μg/m^3, 11%).

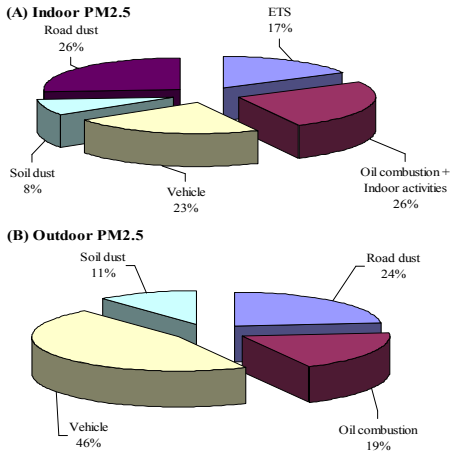

Fig. 3. Source apportionment results by PMF for (A) indoor and (B) outdoor PM2.5.

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