



## Investigation of Indoor and Outdoor PM<sub>2.5</sub> contents through NMDS and PLS regression, supported by I/O ratios

Mohammed O.A. Mohammed <sup>a,b,\*</sup>, Wei-wei Song <sup>b</sup>, Yi-Fan Li <sup>b</sup>, Ahmed A. Elzaki <sup>a</sup>,  
Mohammed A.E.M. Ibrahim <sup>a</sup>, Alshebli A Ahmed <sup>a</sup>, Eithar E.E. Sabil <sup>a</sup>

<sup>a</sup> Faculty of Public and Environmental Health, University of Khartoum, Khartoum 205, Sudan

<sup>b</sup> International Joint Research Center for Persistent Toxic Substances (IJRC-PTS), State Key Laboratory of Urban Water Resource and Environment, School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China

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[mohammedosman@uofk.edu](mailto:mohammedosman@uofk.edu)  
Phone: +249917515992;  
Fax: +21329824945

### Abstract

Investigation of indoor/outdoor (I/O) relations of PM<sub>2.5</sub>, is considered essential since people spend approximately 90% of their time indoors, with great segment of indoor pollution being originated outdoors. Using PM<sub>2.5</sub> I/O ratios, and infiltration factors, the PM<sub>2.5</sub> I/O relations are firstly estimated. Thereafter, analysis with Non-metric Multi Dimensional Scaling (NMDS) and Partial Least Square(PLS) regression was performed to pinpoint the consistency of results obtained by the different methods. PM<sub>2.5</sub> mass levels showed spatial homogeneity across the sampling sites, which is attributed to absence of cooking, indoor smoking, and natural ventilation. For wintertime, while NMDS showed distinct clusters of source tracers, an advanced PLS regression revealed overlapping of coal combustion related species with traffic markers between indoor and outdoor. For summertime, NMDS showed that, coal related tracers were split up into sub clusters suggesting contribution from factories besides winter-heating, and that, the distances between the same markers for outdoors and indoors were closer than that for wintertime. It is also suggested that, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> were not influenced by the same factors/sources.

## 1. Introduction

Atmospheric particulate pollution requires particular attention in areas with extreme temperatures and that are subject to annual fluctuations in weather conditions, for several reasons. For instance, atmospheric stagnation (lower dispersion) and slow chemical transformations during colder months increase mass concentrations of particles and levels of Volatile Organic Compounds (VOCs) [1, 2]. High relative humidity (RH) increases aerosol mass levels through absorption of water vapor by inorganic species/precursors and their subsequent oxidation to form secondary aerosols [3-5]. Large-scale burning of agricultural wastes and combustion of large amounts of coal for winter heating release a wide spectrum of semi volatile and nonvolatile components, such as Polycyclic Aromatic Hydrocarbons (PAHs), elemental carbon (EC), and organic carbon (OC), to air [6]. Rapid temperature inversions during spring and winter trap pollutants in the lower layer of the atmosphere, leading to high pollution levels [4, 7]. Specific weather conditions, such as high solar radiation in summer, favor formation of secondary aerosols from chemical reactions such as oxidation of NO<sub>2</sub>, NO<sub>x</sub>, and VOCs (precursors of secondary organic aerosols (SOAs)) to semi volatile and nonvolatile components, leading to an increase in particle mass concentrations [3, 8, 9].

Exposure to particulate matter, especially to fine particles (PM<sub>2.5</sub>), has been associated with several potentially-deleterious health effects, such as bronchitis and pneumonia in children [10], increase in respiratory mortality [11], cardiovascular effects [12-14], and pulmonary inflammation [15, 16], and DNA damage or cytotoxicity [17, 18].

Broadly speaking, the fine particles found indoors are results of outdoor-infiltrated and indoor-generated particles as well as the secondary formation/chemical transformations occurring indoors [19, 20]. The relationships between the indoor and outdoor PM<sub>2.5</sub> mass concentrations and its associated contents are rather complex [21], which are investigated through different approaches. One of the widely used and easily understandable methods of addressing this relationship is the estimation of indoor/outdoor (I/O) ratios [22-24]. Values of I/O well below one, indicates the insignificant influence/absence of indoor sources, while values greater than 1, may suggest existence of indoor sources [25]. The second commonly applied method is the calculations of infiltration factors ( $F_{INF}$ ), through estimating the  $F_{INF}$  from a regression analysis [26-29], or using the I/O ratio of ( $SO_4^{2-}$ ) under the assumption that there are no indoor sources of sulfate in the studied sampling sites [30, 31].

PLS regression is superior to PCA [32] and multiple linear regression (MLR) [33], and may provide useful information about the apportioning and I/O relationships of PM<sub>2.5</sub>. Briefly, take  $I \times K$  as a matrix denoted Y, where I observations and K dependent variables, while  $I \times J$  matrix denoted X, where I observations and J independent variables. The aim of PLS regression is predicting Y from X and describing their common structure. PLS regression detects components from X, which are also relevant for Y. It searches for a set of components (known as latent vectors) that performs a simultaneous/concurrent decomposition of Y and X with a constraint that these components explain as much as possible of the covariance between Y and X. Computing these latent vectors has another advantage of solving the problem of Multicollinearity [34].

On the other hand, NMDS analysis is presumed to give better description of indoor-outdoor connections between inorganic contents of PM<sub>2.5</sub> since it relies on certain data matrices such as Pearson's correlation matrix [35, 36]. NMDS perform reduction in the dimensionality of data through reconstructing a low-dimensional coordinate set. That is, NMDS tries to represent pairwise dissimilarity between objects in a low dimensional space. The distance between objects and their locations on the x axes or y axes is very meaningful. In addition, the NMDS plots visually illustrate sub clusters, therefore support results obtained by other multivariate techniques [37, 38]. Last but not least, there are other potentially valuable methods for studying the influence of outdoor pollution on indoor air quality including Generic programming [39], and multivariate prediction through principal component regression (PCR) [40].

To our knowledge, few studies have investigated and apportioned sources of PM<sub>2.5</sub> in complex situations and severe cold zones [41]. Harbin city is one such case; the city is surrounded by agricultural land where large-scale biomass burning takes place during late summer and at the beginning of winter. The area is also characterized by extremely low temperatures. Such low temperatures, coupled with continuous high vehicle emissions, are expected to result in high particulate air pollution in the lower layer of the atmosphere. There are also several additional factories (such as pharmaceutical plants).

## 2. Material and Methods

### 2.1. Study area and sampling

Harbin city, the capital of Heilongjiang province, is located in the northeast part of mainland China at 125°42'–130°10' E and 44°04'–46°40' N [42]. Weather conditions are characterized by extremely cold winter temperatures (average winter temp. ~ [-14]) that last for six months (mid-October to mid-April) [43]. Spring usually occurs between April and May and is a transitional period between winter and summer. Annual temperatures range from -30 °C to +30 °C [42, 44] and intermittent rainy days occur almost every week during summer, with annual mean rainfall of 520 mm [42]. Winter heating is usually used from October 20 to April 20.

Samples were collected using Laoying, 2030 intelligent flow sampler (qingdao laoshan, Applied Technology Research Institute, peoples' republic of China). Samples were collected on 12-hour basis, firstly, to estimate daytime and nighttime variations then to calculate the 24-h concentrations of PM<sub>2.5</sub>. Four sampling sites were selected (three residential houses and one office). For the outdoor measurements, samplers were placed on top roofs, while for indoor measurements, samples placed in living rooms and an office. All the selected sampling sites include no smokers and non-cooking activities during the period of the study. A total of 176 sample collected on quartz filters on basis of 22 sample from each site, each season (88 sample during winter and 88 sample in summer). Samples were first thermally treated in oven for 6-8 hours at 450 °C degree. Before conducting gravimetric analysis, samples were subjected to neutralization in desiccators for 24-48h under controlled laboratory conditions (35 ± 5 relative humidity and (25 ± 5 °C). Eighteen blank samples were treated the same as the real samples for the purposes of quality control.

### 2.2 Chemical analysis

Carbonaceous content (OC, EC) of PM<sub>2.5</sub> was analyzed using a Thermal/Optical Carbon Analyzer (Model 2001, Desert Research Institute, Atmoslytic Inc., Calabasas, CA, U.S.A) using IMPROVE\_A protocol (Interagency

Monitoring of Protected Visual Environments) [45, 46]; this provided concentrations of OC and EC based on the thermal optical reflectance (TOR) method. A 0.53 cm<sup>2</sup> circular punch from each sample was analyzed [47, 48]. The temperature program of the IMPROVE\_A protocol used for analysis is explained in detail in the literature [47-49]. Trace metals were first digested by closed vessel microwave-assisted acid digestion using nitric acid (HNO<sub>3</sub>), aided by aqueous hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>.30% (w/w)[50], before being analyzed with a quadrupole inductively coupled plasma-mass spectrometer [ICP-MS, X series 2, ThermoFisher, Inc.US].

For water-soluble ion analysis, one quarter of each sample was ultrasonically extracted. Each piece of filter was extracted twice. Each time, 20 ml of ultrapure water were added to the sample in a glass tube, with this then transported to an ultrasonic water bath (at 30 °C for one h). The water for extraction was purified using *NANOpure Diamond*, Model D111911, Barnstead International, USA. All extracts were filtered through 0.45 μ filters. Samples were then analyzed using an ion chromatography system (ICS-90; from Dionex, Thermo scientific, Inc.U.S), following an approach described in the literature [51].

### 2.3 Non-metric Multi-Dimensional scaling

As a prerequisite to non-metric multi-dimensional scaling (NMDS) analysis, a data matrix with the same number of rows and columns (squared matrix) is required. There are three common types of matrices for NMDS analysis including, a dissimilarity matrix using for example Bray-Curtis distances[52], similarity matrix and Pearson's correlation matrix where the latter type is used for NMDS analysis in this study[35, 36]. The starting configuration was Standard Guttman-Lingoes, with minimum and maximum iteration of 6 and 100, respectively. The task of Kruskal's NMDS which was set to one, is to attempt minimizing what is known as "stress". As stated by De Blast, et al.2010, the parameter "stress" indicates the extent of the distortion of distances' monotony in the MDS plot. In this study, the NMDS ordinations were performed for concentrations of inorganic chemical species of all sampling sites and times using STATISTICA software version10 (StatSoft Inc., Dell Software, Tulsa, OK, 74104 USA).

### 2.4 Partial least square (PLS) regression

Partial least square (PLS) regression was done based on NIPALS algorithm (nonlinear iterative partial least squares) using STATISTICA software. The validation was done with leave-one-out approach of cross-validation to estimate prediction errors. This is done by fitting a PLS model into n – 1 samples and making a prediction of the y(dependent variable)value for the omitted sample (y<sup>^</sup>) (i.e. one sample is removed at a time while model is formed on remaining samples). When this is performed for every sample in the dataset, a predicted residual error sum of squares (PRESS) is then estimated for that model as in Eq. (2-3)[53]. PLS algorithm searches for a model with lowest PRESS value to be chosen. That is, if the obtained results showed low value of PRESS this indicates accuracy and validity of the results. It is therefore presumed that cross-validation, optimize the number of PLS components [54].

$$\text{PRESS} = \sum_{i=1}^n (y_i - \hat{y}_i)^2 \quad \text{Eq. (2-3)}$$

The underlying principles of PLS are also discussed elsewhere in the literature [55-57].

### 2.5 Calculations of I/O ratios and F<sub>INF</sub>

In this study, prior to application of NMDS and PLS, the indoor-outdoor relations of PM<sub>2.5</sub> were first investigated through estimation of PM<sub>2.5</sub> I/O ratios and calculating the infiltration factors(F<sub>INF</sub>), which in turn estimated via two approaches: through calculating the F<sub>INF</sub> from a regression analysis using Eq.(4-1) below, with more details given elsewhere [26-29]. Secondly, using sulfate (SO<sub>4</sub><sup>2-</sup>) as a reference marker under the assumption that there are no indoor sources of sulfate in the studied sampling sites. More details on using sulfate for F<sub>INF</sub> estimation ( Eq. (4-2)) which is simply the I/O ratio of (SO<sub>4</sub><sup>2-</sup>) is provided elsewhere [30, 31].

$$C_i = F_{INF}C_a + C_{ig} = C_{og} + C_{ig} \quad \text{Eq. (4-1)}$$

$$F_{INF}^{Sulf} = \frac{C_i^{Sulf}}{C_a^{Sulf}} \quad \text{Eq. (4-2)}$$

Where: C<sub>i</sub> indoor PM<sub>2.5</sub> concentration; C<sub>a</sub> ambient PM<sub>2.5</sub> concentration C<sub>ig</sub> indoor-generated PM<sub>2.5</sub>; C<sub>og</sub> outdoor-generated PM<sub>2.5</sub>. C<sub>i</sub><sup>sulf</sup> is indoor sulfate concentration; C<sub>a</sub><sup>sulf</sup> is ambient sulfate concentration.

### 3. Results and discussion

#### 3.1 Estimation of indoor/outdoor ratios of PM<sub>2.5</sub> and F<sub>INF</sub>

In this study, indoor and outdoor PM<sub>2.5</sub> contents showed fair resemblance in the distributions (Fig.3.1), which are likely driven by infiltration of outdoor air and absence of indoor chemical transformations that may alter the compositional profile of PM<sub>2.5</sub>. This claim is also supported by a fact that, photolysis (reactions via the action of light) is expected to be low indoors[20]. In reality, I/O ratios of PM<sub>2.5</sub> are affected by several factors such as meteorological parameter (ambient temperature, RH, wind etc.) and building structure [58-60]. The I/O ratios calculated in the present study were almost similar to those estimated in different parts of the world (see Table 3.1). our findings agreed also with results of a relatively recent large-scale review, where I/O ratios were reported to be around the unity (1) for European cities and ranged between 1.2-2.5 for US cities, with the higher I/O ratios in the latter case are attributed to indoor sources mainly to indoor smoking[61]. The I/O relations and calculations are well discussed elsewhere[62, 63]. I/O ratios of individual chemical species showed that, Cr, Co, Mn, Ba, and Pb are the species with higher I/O ratios, in partial agreement with findings of previous studies [64-67].

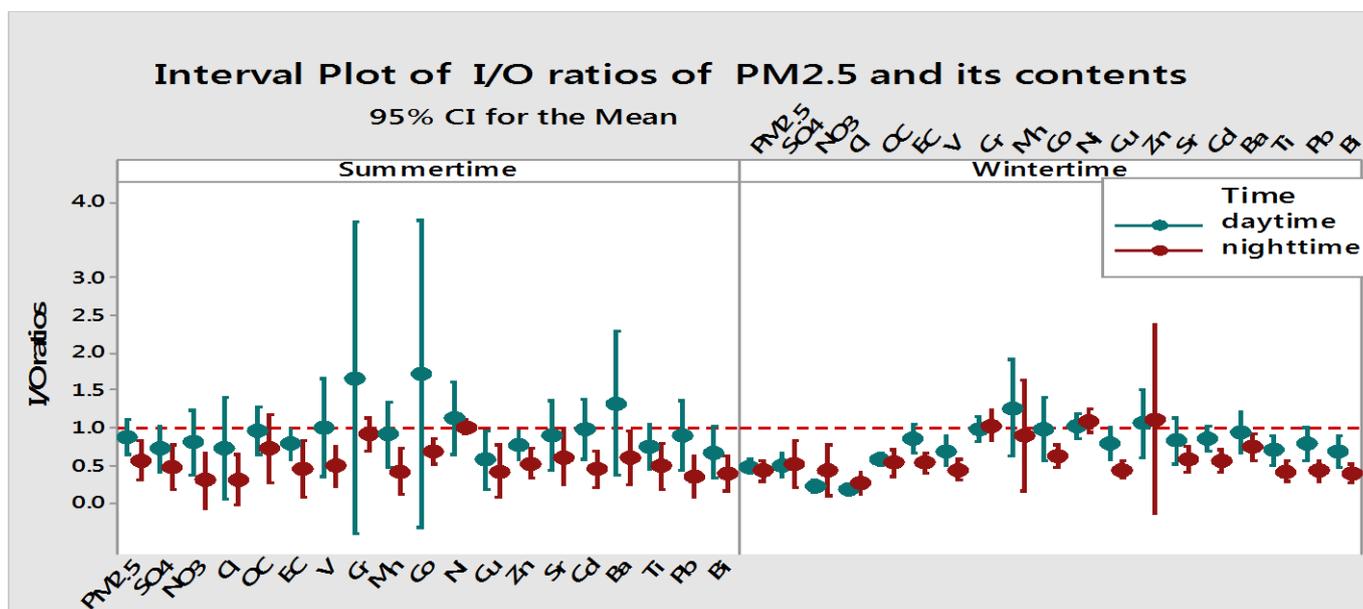


Figure. 3.1 I/O ratios of PM<sub>2.5</sub> mass and associated contents

According to regression analysis (Fig 3.2), slightly higher mean F<sub>INF</sub> (0.35) in summer than winter (0.27) was reported and the average indoor-generated PM<sub>2.5</sub> equal to 27.7 and 17.1 for summer and winter respectively. It is also indicated that, the correlation between indoor and outdoor PM<sub>2.5</sub> was moderate during summer (R<sup>2</sup>=46%) compared to low (R<sup>2</sup>=27%) in winter. The regression analysis has therefore, confirmed the slightly higher infiltration during summer compared to winter, that estimated by indoor/outdoor ratios. On the contrary, the F<sub>INF</sub> estimated by the I/O ratio of SO<sub>4</sub><sup>2-</sup> was suggested to be invalid since its values were inconsistently high and disagree with the results of regression analysis (mean I/O SO<sub>4</sub><sup>2-</sup> = 0.6 for summer and 0.5 for winter).

Based on the F<sub>INF</sub> estimation also, PM<sub>2.5</sub> mass levels spatial homogeneity has been reported across the four sampling sites, in an agreement with evidence from the literature [21, 68]. This homogeneity in patterns of PM<sub>2.5</sub> distributions is principally due to absence of cooking and indoor smoking as stated earlier. It is also partially explained by a fact that, all the selected sampling sites are naturally ventilated and no any sort of artificial air conditioning is used that may affect the particles' deposition or resuspension in the indoors, with more details on this matter is mentioned elsewhere [69-71]. However, a precaution has to be taken that; considerable spatial variability could be expected in urban areas where some parts of the cities are significantly impacted upon by PM<sub>2.5</sub> point sources or uneven blown of wind from multiple directions [72, 73].

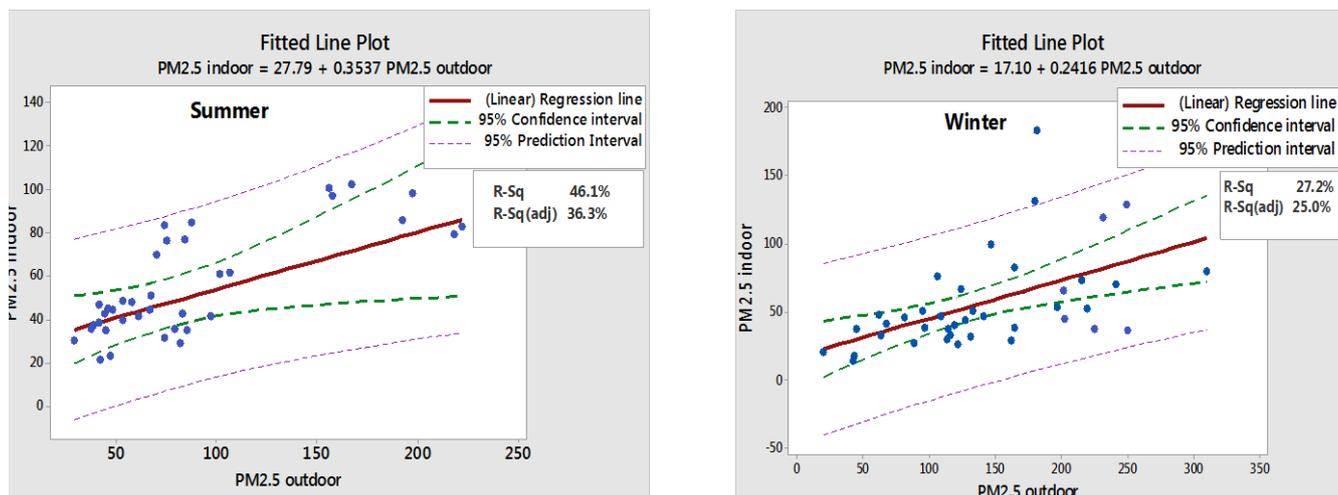


Figure. 3.2 Regression of indoor vs. outdoor PM<sub>2.5</sub>

Table 3.1 comparison of I/O ratios of different studies

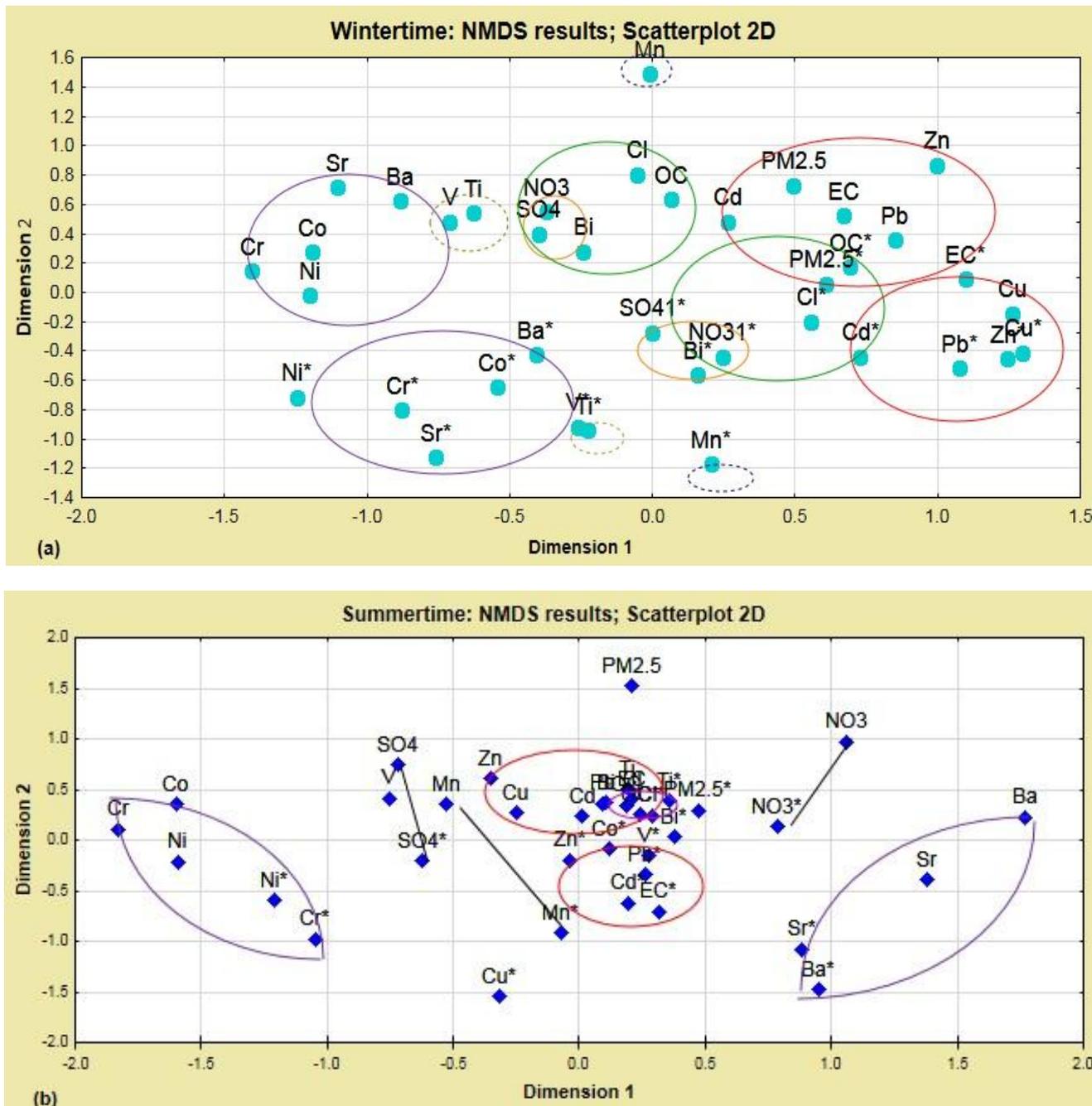
| PM <sub>2.5</sub> I/O ratios | Types of microenvironments                       | Places                  | reference  |
|------------------------------|--|-------------------------|------------|
| 0.7 (average)                | Urban residential houses                         | Harbin, China           | This study |
| Ranged 0.76 - 1.13           | Urban residential houses                         | Agra, India             | [70]       |
| 0.94 (average)               | Urban residential houses                         | Guangzhou, China        | [71]       |
| Ranged 0.2-4.7               | Urban residential houses                         | Greater Cincinnati, USA | [72]       |
| 0.74 (average)               | Urban residential houses                         | Bologna, Italy          | [73]       |
| 1.01 (average)               | Urban residential houses                         | Agra, India             | [74]       |
| 1.02 (average)               | Urban residential houses and schools             | Stockholm, Sweden       | [75]       |
| 1.2 (average)                | Suburban residential houses                      | Melbourne, Australia    | [76]       |
| Ranged 0.9-1.32              | Classrooms                                       | Chennai, India          | [77]       |
| 0.81 (average)               | Office buildings and classrooms                  | Beijing, China          | [78]       |
| 0.69 (average)               | Office buildings                                 | Milan, Italy            | [79]       |
| Ranged 0.36-1.08             | Public facilities (restaurants, libraries, etc.) | Beijing, China          | [22]       |
| Ranged 0.8-2                 | Confined school building                         | Wroclaw, Poland         | [80]       |

### 3.2 Evaluation of infiltration and similarity between indoor and outdoor PM<sub>2.5</sub> using NMDS

Non-metric multidimensional scaling (NMDS) analysis based on correlation matrix was performed, first, to confirm the estimated I/O ratio results. Secondly, to test whether patterns of PM<sub>2.5</sub> and associated inorganic contents are changed due to factors such as secondary formation, weather conditions, and possible intermittent emission sources during winter and summer. Finally, to visualize the similarity between indoors and outdoors in terms of distributions of individual inorganic species. The parameters for the NMDS 2-dimensional solution, were similar to those mentioned in the literature[37]. Briefly, maximum number of iterations = 20, the number of runs with real data = 100 (there were no runs with randomized data), stability criterion set to 0.00001 (over the last 15 iterations), and initial step length was 0.20.

NMDS plots generated for inorganic contents of PM<sub>2.5</sub> for the indoors and outdoors are showed in Fig.3.3. During winter, the first observable trend is that, the total indoor PM<sub>2.5</sub> mass was associated with traffic related markers (Cd, Zn, EC, and Pb) while the total outdoor PM<sub>2.5</sub> mass linked to these traffic markers and to OC, Cl, where the latter two species together indicates biomass burning, with the Cl is considered an excellent indicator of biomass. It was obvious that, during winter, the outdoor distribution of the inorganic contents of the fine particles was separated from that of the indoor, with clear clustering of source markers. Six clusters were identified

for both indoors and outdoors with no overlapping. Traffic source with the above mentioned tracers, coal combustion indicated by Cr, Co, Sr, Ba, and Ni, was placed towards the negative corner of axis 1 for the outdoor PM<sub>2.5</sub> while laid on the positive site of axis 2 for the indoor PM<sub>2.5</sub>. Biomass burning identified by Cl, OC, along with SO<sub>4</sub><sup>2-</sup>, and NO<sub>3</sub><sup>-</sup>, while the latter two species were laid near each other indicating secondary formation as well. The distance between indoor Mn and outdoor Mn was the longest among all species, with this marker showed no clear connection with the other markers, implying that it has been affected by unique factors that stated earlier in chapter three. Unexpectedly, the typical soil tracer, Ti neighbored V for the data of indoor and outdoor PM<sub>2.5</sub>.



Note: The sign “\*” indicates outdoor measured species.

Figure 3.3 NMDS 2-dimensional solutions for winter and summer

On the contrary, during summer, the compositional profiles showed heterogeneous patterns and were substantially different from that of winter. With the exception of traffic-related markers which showed two clear clusters, all

other tracers revealed different patterns. Overall, the distances between the same species of outdoors and indoors were closer, for instance, indoor  $\text{SO}_4^{2-}$  placed not far from outdoor  $\text{SO}_4^{2-}$ , and the indoor  $\text{NO}_3^-$  laid on a side close to outdoor  $\text{NO}_3^-$ , suggesting that,  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were not influenced by the same factors. The distance between indoor Mn and outdoor Mn become shorter, while Cl, OC of outdoor and indoor were overlapped/closer compared to winter time. The coal combustion tracers were split up into two subclusters. One cluster showed Sr neighboring Ba and the second sub cluster included Cr, Co and Ni. Last but not least, indoor Cu loaded on the negative end of axis 1, relatively far from outdoor Cu on the positive end of axis 2. These results suggest existence of more industrial activities in summer affecting trends of Cr, Co, Ni and Cu where these species are also simultaneously emitted from industrial plants[85]. It is possible that, the obvious association between indoor and outdoor fine particles, was partly due to a fact that, the studied buildings are naturally ventilated that results in more exchange of air, unlike in mechanically ventilated buildings as indicated by Gupta and Cheong, (2007) [58]and Riain, et al. (2003)[60], in agreement with finding of another related study[86].

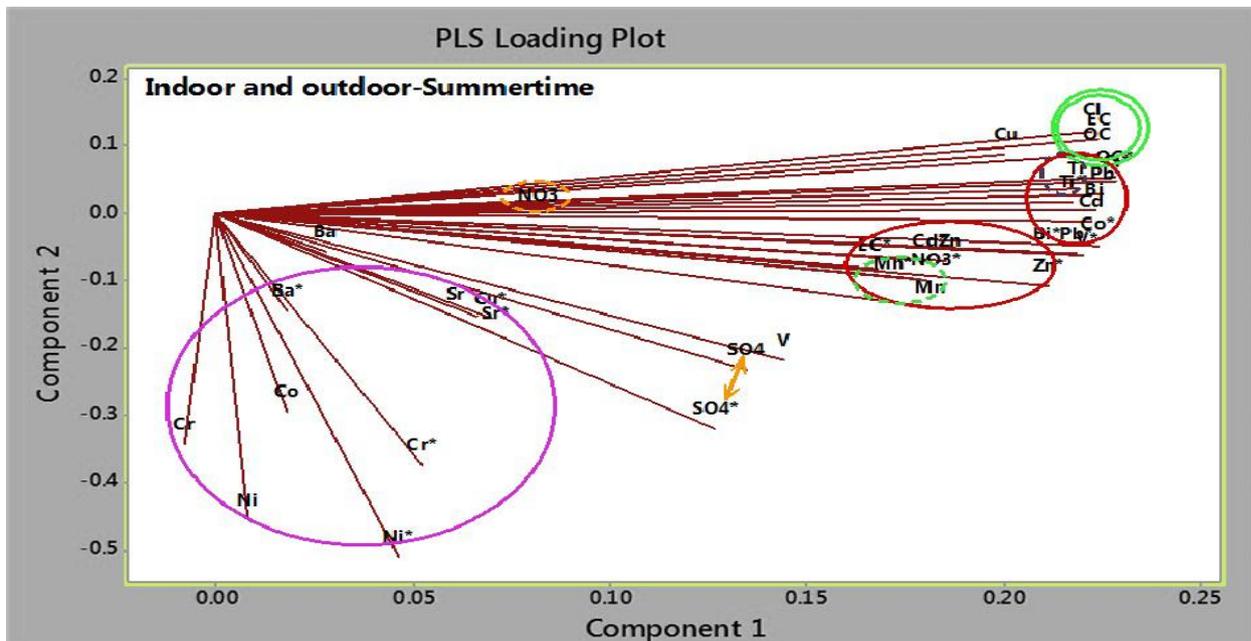
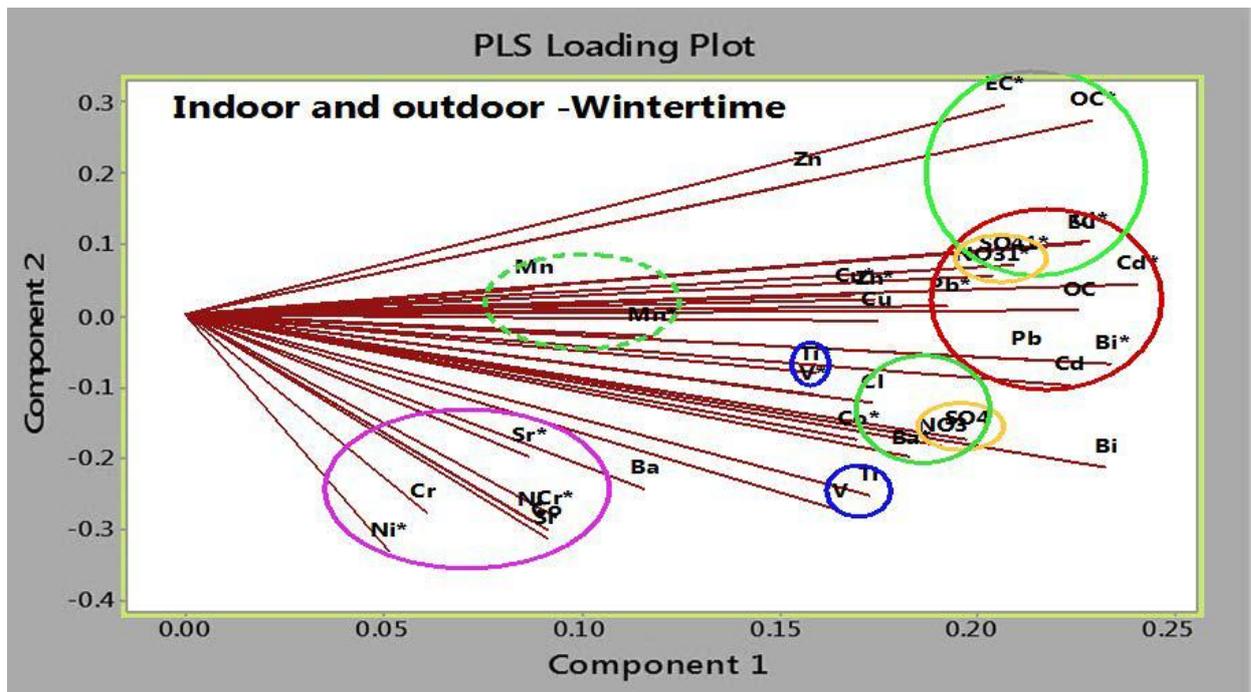
### 3.3 Confirmatory analysis and source apportionment with Partial Least Square regression

PLS regression was conducted to support NMDS results, to apportion the emission sources and most importantly, to detect the “variables of importance” (VIPs). PLS was done based on NIPALS algorithm (nonlinear iterative partial least squares) and *leave-one-out* method of cross-validation, at 95% confidence level. The underlying principles of PLS are discussed elsewhere in the literature [55-57]. Fig.3.4 illustrates the PLS findings, including the loading plots and VIPs.

To begin with, for winter, the PLS showed interesting results by confirming existence of traffic, biomass burning, and coal combustion as the major pollution sources, explaining approximately 30%, 22%, and 13% of the variance in outdoor  $\text{PM}_{2.5}$  and 25%, 18%, and 11% for indoor  $\text{PM}_{2.5}$ , respectively. Major ions ( $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ ) formed two clusters that in turn, attached to another two clusters classified as biomass burning sources (see Fig.3.4), implying that these two ions resulted from both secondary aerosol formation and biomass burning as well. It has to be noted also that, PLS revealed more overlapping of the  $\text{PM}_{2.5}$  outdoor sources with the sources of indoor  $\text{PM}_{2.5}$ , unlike NMDS that showed distinct clusters of sources. For instance, coal combustion source of  $\text{PM}_{2.5}$  measured outdoor, overlapped with coal combustion of indoor measured  $\text{PM}_{2.5}$ , with a similar trend for traffic source. In addition, outdoor Mn source (metallurgical sources) neighbored indoor Mn. Therefore, the overall results of NMDS and PLS indicated fair link between the indoor and outdoor  $\text{PM}_{2.5}$ . In fact, this link is explained by absence of cooking, and indoor smoking, the most powerful determinants of indoor particles' levels [21, 87-90], although there are other factors such as cleaning activities [91] and the occupancy (living in or using a building), that accelerate resuspension of particles [92].

For summer, with the exception of traffic-related markers that gathered to form two distinct clusters for the indoor  $\text{PM}_{2.5}$  and outdoor  $\text{PM}_{2.5}$ , all other emission sources revealed overlapping. The biomass burning source for outdoor  $\text{PM}_{2.5}$  is suggested to be the same biomass burning source of indoor fine particles, as indicated in Fig3.4. Interestingly, the major ions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ) assumed to be influenced by different factors since  $\text{NO}_3^-$  placed far from  $\text{SO}_4^{2-}$  in the PLS loading plot. This may imply some sort of chemical transformation during summer having more impact on  $\text{PM}_{2.5}$  compared to winter. Last yet importantly, coal related species were dispersed over a wider range and positively linked to axis 1 and negatively to axis 2, confirming also that coal-related tracers (Co, Cr, Sr, Ba and Ni) are emitted from similar sources.

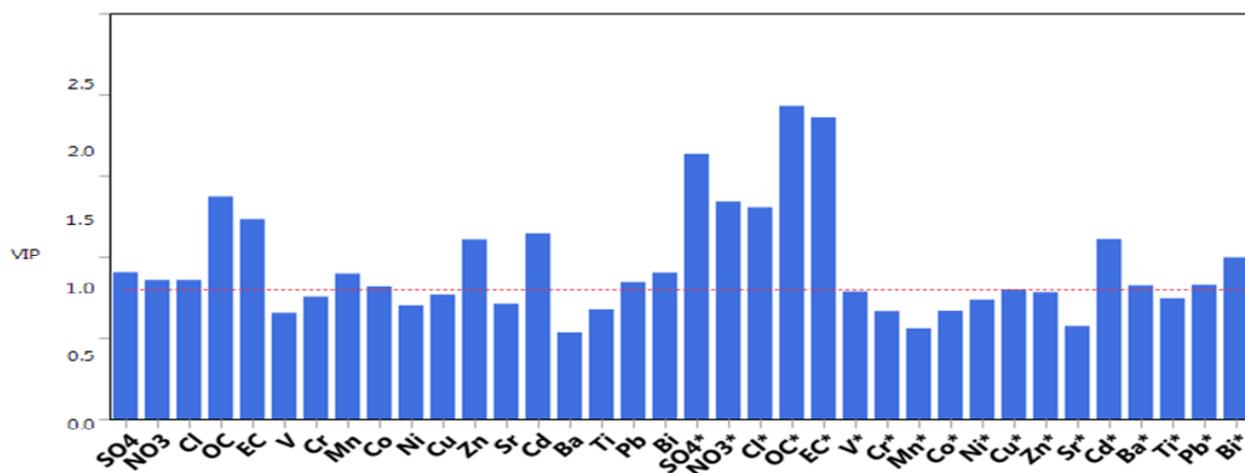
As part of PLS algorithm, *variable importance plots* are generated which show the variables significantly influenced trends of fine particles. Variables with VIP values greater than the unity are classified significant. These variables are not necessarily the species of high influence on the particles' total mass; rather these are the most important determinants of the spatiotemporal variations in the fine particles. For winter, these variables included OC, EC,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , Zn, Cd, Pb, Mn, and Bi, while for summer the same variables in addition to Ni, Cu, Cl, and Sr were detected (see Fig.3.5). This finding suggest four things; first, traffic as indicated by Cd, Zn, Pb, and EC is a serious pollution source throughout the entire sampling period. Second, since coal burning-related tracers are showed low VIPs in winter, hence, heating systems, the major sector contributing to coal-related emissions, are assumed to emit almost the same levels of emissions throughout the different months during winter, i.e. less influence on  $\text{PM}_{2.5}$  variability in winter. Third, since Cl, a typical marker of biomass burning, has high VIP value of 1.4 for summer compared to winter, it suggests intermittent emissions from biomass during the former period. Fourth, nitrate and sulfate are both less abundant indoors, in agreement with an evidence from the literature which related this to the absence of reactive species such as ozone and free radical reactions indoors [93].



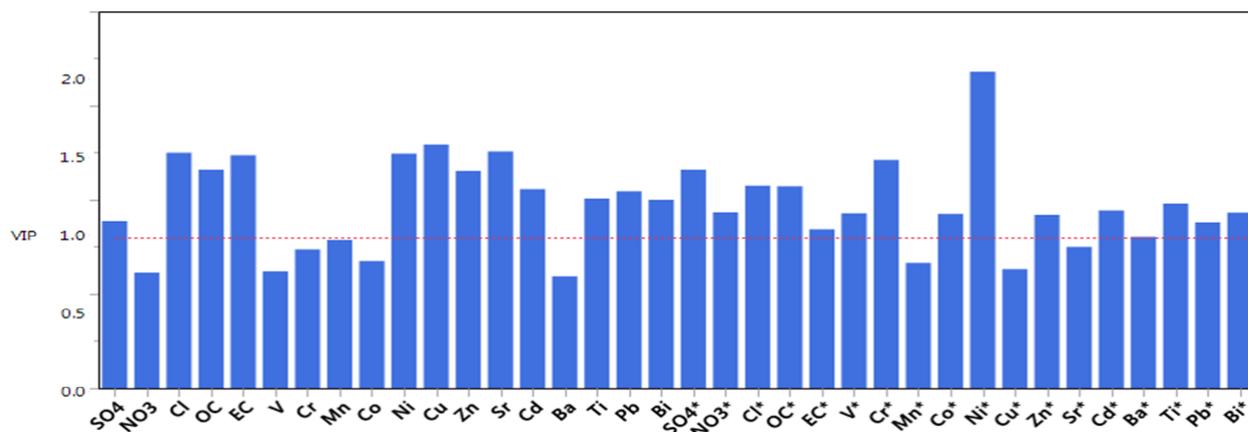
Note: The sign “\*” indicates outdoor measured species.

**Figure. 3.4** PLS loading plots for (a) winter and (b) summer

The consistently high contribution from traffic that detected by both PLS and NMDS, is therefore, important for some reasons. Firstly, the combustion related emissions particularly from vehicle exhaust are more potent in causing adverse health effects compared to those from non-combustion process, because they contain host of inorganic and organic materials [20, 90]. Secondly, traffic is generally considered the most significant outdoor source of aerosols in countries with higher industrial activities, accounting for more than 50% of the total source contributions[20]. Traffic-related atmospheric pollution remained a crucial target issue for the public health officials[91], because it affects not only the physical and chemical aspects of the particles, but also their toxicological characteristics [92].



(a) wintertime



(b) Summertime

Note: The sign “\*” indicates outdoor measured species.

**Figure. 3.5** Values of variable of importance (VIPs) for (a) winter and (b) summer

## Conclusion

This study was intended to determine the chemical characteristics of PM<sub>2.5</sub> and to investigate its spatial and temporal trends and to identify the possible emission sources of PM<sub>2.5</sub> in Harbin city, China. Different conventional ( $F_{INF}$  and I/O ratios) and advanced analytical methods (NMDS and PLS regression) were applied. The relationships between indoor and outdoor PM<sub>2.5</sub> and its contents are investigated through NMDS, and PLS regression simultaneously. These two methods together, provided better explanation of the negative and positive relationships between indoor and outdoor PM<sub>2.5</sub> and possible effects of factors such as type of sources, existence of significant indoor emissions, and effects of house characteristics.

During winter, distinct clusters of inorganic tracers are identified, however, coal combustion related species and traffic markers were overlapped between indoor and outdoor. This implies that, in winter outdoor sources were different from indoor ones with the exception of traffic and coal combustion sources. During summer, chemical transformation in the air could be a reason behind the heterogeneity of distribution of inorganic contents and that,  $SO_4^{2-}$  and  $NO_3^-$  were not influenced by the same factors.

Traffic, coal combustion, biomass burning, and industrial emissions are major emission sources of inorganic contents of PM<sub>2.5</sub> in general, with the secondary aerosols, metallurgical processes and Ca-enriched source (from building construction) being principal sources during summer. It is suggested that, secondary aerosol formation and resuspension of soil dust have high impact on PM<sub>2.5</sub> mass in summer while coal combustion and biomass burning cause more pollution during early winter.

Concurrently, three methods for investigating infiltration of fine particles and its inorganic contents into indoors are performed to give a valid estimation of the amount of indoor-generated PM<sub>2.5</sub>. These methods are the I/O ratios, F<sub>INF</sub> calculation using SO<sub>4</sub><sup>2-</sup> and F<sub>INF</sub> estimated through regression analysis. The compositional profiles of PM<sub>2.5</sub> of indoors resembled that of outdoors, with the infiltration of outdoor PM<sub>2.5</sub> being slightly high (F<sub>INF</sub> = 0.35) in summer than winter (F<sub>INF</sub> = 0.27). In addition, homogeneity in patterns of PM<sub>2.5</sub> distributions was suggested and attributed to absence of cooking and indoor smoking.

Generally, concentrations of levels of PM<sub>2.5</sub> are exceeded both the national and international air quality standards which need a permanent solution. It is presumed also that, air pollution is affected by local sources rather than by long-range transport of air masses.

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**Conflicts of Interest** - There is no conflict of interest to declare

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