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Source Identification and Quantification of Coarse and Fine Particles by TTFA and PMF

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Abstract

Receptor modeling is one of statistical methods to achieve reasonable air pollution strategies. In order to maintain and manage ambient air quality, it is necessary to identify sources and to apportion its sources for ambient particulate matters. The main purpose of the study was to survey seasonal trends of inorganic elements in the coarse and fine particles. Second, this study has attempted emission sources qualitatively by a receptor method, the PMF model. After that, both PMF (positive matrix factorization) model and TTFA (target transformation factor analysis) model were applied to compare and to estimate mass contribution of coarse and fine particle sources at the receptor.

A total of 138 sets of samples was collected from 1989 to 1996 by a low volume cascade impactor with 9 size fraction stages at Kyung Hee University in Korea. Sixteen chemical species (Si, Ca, Fe, K, Pb, Na, Zn, Mg, Ba, Ni, V, Mn, Cr, Br, Cu, Co) were characterized by XRF. The study result showed that the weighted arithmetic mean of coarse and fine particles were 51.3 and 54.4 $\mu g/m^3$, respectively. Contribution of both particle fractions were estimated using TTFA and PMF models. The number of estimated sources was seven according to TTFA model and 8 according to PMF model. Comparison of TTFA and PMF revealed that both methodologies exhibited similar trends in their contribution pattern. However, large differences between contributions were observed in some sources. The results of this study may help to suggest control strategies in local countries where known source profiles do not exist.

Key words : Aerosol, Cascade impactor, Receptor modeling, TTFA, PMF

1. INTRODUCTION

Diverse pollutants from a variety of sources have resulted in atmospheric pollution, which in turn have resulted in grave problems such as visibility degradation and acidic deposition on the local and regional scale, and such as ozone depletion and the global climate changes on the global scale. Since the conditions also pose a direful threat to human health and welfare, individual countries are developing and executing all sorts of environmental policies to control atmospheric

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pollutants with making every effort to set up new environmental criterion and evaluating the sources of the atmospheric pollutants. One of these increasing efforts has been made to survey the physicochemical properties of atmospheric particulate matters and thereby quantitatively understand to what extent of specific pollution sources affects the adjacent regional atmosphere. For regulations to be effective it must be preceded by qualitative and quantitative analyses of air pollution sources for the sake of preserving the ambient air quality. These control steps, using mainly receptor methods, must be the basis for establishing sound environmental policies.

Receptor modeling, a field of chemometrics, is based on manifold applied statistics and is a statistical methodology that analyzes the physicochemical properties of gaseous and particulate pollutants on various atmospheric receptors, identifies the sources of air pollutants, and quantifies the apportionment of the sources to the receptors. Numerous researchers have applied these methods to actively study and assess the contribution of sources to enable reasonable atmospheric pollution control. However, in Korea, only a few researches have been performed concerning the source apportionment study (Hwang and Kim, 1998).

Receptor methods consist of physicochemical analyses and applied statistical analyses that can mostly start from interpreting the measurement data. Enrichment factor method and/or time series method had been intensively used during the early history of receptor methods, but multivariate statistical analyses such as chemical mass balance (CMB) and factor analysis are now widely used. The CMB method originated from Miller *et al.* (1972) and Friedlander (1973) is universally used to estimate the quantitative contribution of source to the ambient. However, it requires local source profiles, which are not yet available in Korea due to the insufficient inventory studies, so that a caution should be exercised when it is employed without local source information. Thus, multivariate statistical analyses such as factor analysis, multiple regression analysis, and cluster analysis have been more generally used in Korea. Factor analysis is one of candidates, which can be used to interpret the complex and vast environmental data and it is thus most popularly employed to estimate source of pollution without source profile, despite the fact that it can only provide the quantitative information of the source. In addition, multivariate statistical analyses such as principal component analysis (PCA), Absolute PCA, SAFER/UNMIX, positive matrix factorization (PMF), and target transformation factor analysis (TTFA) are also widely used in the receptor modeling area (Kim *et al.*, 2001; Hien *et al.*, 1999; Huang *et al.*, 1999; Simcik *et al.*, 1999; Hwang and Kim, 1998; Poissant *et al.*, 1996).

The objective of this study is to estimate the concentration trends of the aerosol particles in the samples collected by a cascade impactor according to the particle size as well as the concentration trends of inorganic elements. Quantitative contribution of fine and coarse particles to the ambient air is also estimated using the PMF model, whose algorism is more advanced than that of factor analysis. Finally, both estimated contributions by TTFA method and PMF method are compared.

2. EXPERIMENTAL METHODS

2. 1 Sampling methods

The aerosol samples were collected from the top (5th) floor of the Natural Science Building, Kyung Hee University, Suwon-Campus, Kyunggi Province in Korea from November 1989 to March 1996 (Fig. 1). A total of 138 sets (each set consists of 9 filters) was collected during the sampling periods. The building is located about 10 km east of Suwon-City and 4 km south of the Singal interchange on the Kyungboo Highway. In the vicinity is Lake Singal, having storage area of 231 ha, and many small and medium industries are scattered around its tributaries. Traffic is heavy due to the well-developed network of roads. Vast housing land development was in progress near sampling site during the sampling period. Presently, with housing land dev-

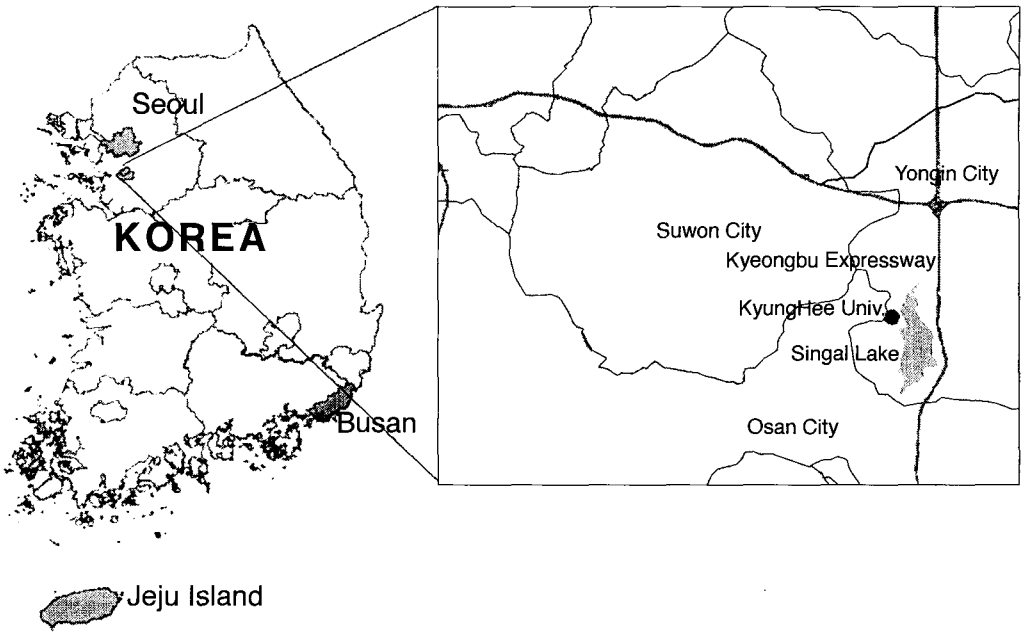

Fig. 1. The location of the sampling site.

eloping completed, there is a larger floating population and heavier traffic than before.

Aerosol samples were collected using a 9-stage cascade impactor (KA-200, Dylec Co., Japan) with an average flow rate of 28.3 L/min. Filters were replaced between 7-30 days to obtain sufficient sample quantity until when the flow rate was sharply dropped below 25.0 L/min. Diameter and pore size of the membrane filter (GN-6, Gelman Science Co., USA) used in this study was 80 mm and 0.43 μm, respectively. The filters had been kept in desiccators for three days, before and after sampling, with maintaining constant temperature and moisture to keep its weight unchanged, and then reweighed using a micro-balance (A&D Co., Model HM-202) having sensitivity of 0.01 mg. Aerosol mass concentrations were calculated from these data.

2. 2 Analytical methods

X-ray fluorescence (XRF) spectrometry that does not requires pre-treatment was used to analyze 16 inorganic elements on the filter samples. Out of 138 sets of aerosol samples, only 63 sets were analyzed by XRF. The membrane filter with collected aerosol was cut using a puncher with 38 mm diameter, and then placed onto the sample holder and covered with a Mylar film before analysis. Analytical conditions of XRF are shown in Table 1.

Table 1. Analytical conditions of x-ray fluorescence spectrometer.

Primary x-ray		
	X-ray tube target	:Rh
	Voltage	:50 kV
	Current	:50 mA
	Power	:2.5 kW
	Beam path	:Vacuum
	Fluorescent x-rays	
	Analytical crystal	:LiF(200), EDDT, RX-4 TAP, GE, ADP
Detection		
	Detector	:Scintillation counter (SC) Proportional counter (PC)

The SRM 1832 (NO. 195) and SRM 1833 (NO. 1207) from the NIST were used as standard reference materials. Both standard films contained various elements

with known concentration and possess uniform thickness of 0.55 μm. Sixteen inorganic elements such as Si, Ca, Fe, K, Pb, Na, Zn, Mg, , Ba, Ni, V, Mn, Cr, Br, Cu, and Co were analyzed. Detailed description of the analytical method can be referred to elsewhere (Lee and Kim; 1997).

2. 3 Target transformation factor analysis

The following formula is generally used to estimate the quantitative contribution of aerosol sources to the receptor:

$$x_{ij} = \sum a_{ik} f_{kj} \quad (1)$$

where, x_{ij} is the ith species concentration measured in the in the jth sample, a_{ik} is the mass concentration of the ith species from the kth source (i.e., source profile), and f_{kj} is the concentration of aerosol from the kth source collected in the jth sample (i.e., source contribution). The unknown value f_{kj} can be calculated from the two known values of x_{ij} and a_{ij} by using one of least square methods.

Generally, matrix f_{kj} cannot be calculated mathematically without matrix a_{ij}. However, in case there are sufficient number of samples, the number and the type of sources imbedded in the matrix x_{ij} can be statistically determined. This is the very purpose of TTFA development (Hopke *et al.*, 1982). Through applying TTFA, similar factors were found and clustered using the correlation among variables to determine the number of sources. In this study, Q-mode was used to compose correlation matrix. This matrix was then diagonalized to yield eigenvalues and eigenvectors, which were used to determine the number of factors, i.e., sources. Once the number of factors was determined, the factor axis was rotated to obtain easier physical interpretation. Finally further target transformation processes were executed by using the weighted least square method until a proper source profile was obtained. Detailed description of the TTFA method can be referred to in various literatures (Lee and Kim; 1997; Hopke, 1988).

In TTFA, the user specifies likely target shapes for the composition factors. The algorithm attempts to rotate the computed solution so that the target shapes are reproduced as well as possible. Although TTFA has been successful in many practical problems, it suffers from the fact that rotations are performed a posteriori, after choosing the subspace with an eigenanalysis.

2. 4 Positive matrix factorization

Factor analysis has been widely used in the discipline of atmospheric science to identify chemical properties of aerosol sources. However, factor analysis depends solely on the covariance matrix and implies various defects, such as insufficient information, appearance of negative factor loading, rotational ambiguity (impossibility of perfect physical analysis), and others. A more advanced methodology, positive matrix factorization (PMF), was developed to overcome such defects (Paatero and Tapper, 1994). The PMF always provide positive factor loadings considering the standard deviation of measured data value (Paterson *et al.*, 1999). The primary difference of PMF from factor analysis is that PMF always presents the factor loading as positive value and depends on least-squares minimization algorithm, i.e. error estimate information for each individual datum, not on the correlation matrix information. In addition, corresponding values for the data under the detection limit and missing data can be estimated on the grounds of error estimate.

The PMF approach is usually feasible for two-dimensional and three-dimensional matrix. The following is the mathematical formula for the two dimensional PMF model (Chueinta *et al.*, 2000):

$$X = G F + E \quad (2)$$

where, X is the known $n \times m$ matrix of the m measured chemical species in n samples. G is an $n \times p$ matrix of source contributions to the samples. F is a $p \times m$ matrix of source compositions (p is the number of factors). The values in the rows of matrix G and F are always positive and E is the residual matrix. Details and detailed analytic method are referred to in a variety of literatures (Hwang *et al.*, 2001; Lee *et al.*, 1999; Paatero, 1998; Juntto and Paatero, 1994).

The important advantage of the PMF model is the ability to handle missing and below detection limits data by adjusting the corresponding error estimates of these data points. And the method being introduced utilizes error estimates of elements of the measured data matrix and implements strict non-negativity constraints for the factors, i.e. for the basis and for the coefficients used for approximating the matrix. Thus the PMF model is more quantitative and better suited for physics, chemistry, and environmental sciences than the customary PCA-based models.

3. RESULTS AND DISCUSSIONS

3. 1 Mass concentration

Average concentration of size segregated aerosols by a 9-stage cascade impactor during the sampling period is shown in Table 2. Weighted arithmetic mean was calculated by dividing sample concentration multiplied by sampling duration in days into total sampling duration in days. The range of the weighted arithmetic mean value and arithmetic standard deviation value in each stage was 8.4 ~ 17.4 μg/m^3 and 5.5 ~ 10.6 μg/m^3, respectively. Average concentration of the coarse particles (> 2.1 μm) was 51.3 μg/m^3. The average concentration for the fine particles (< 2.1 μm) was 54.4 μg/m^3, showing higher portion in particles.

Table 2. Average value of aerosol mass concentration (μg/m^3) for each size range in the study area during the sampling periods of 1989 to 1996.

Particle size range (μm)	Number of samples	Weighted arithmetic mean	Arithmetic std. dev.	Min	Max
> 11.0	138	8.4	8.3	0.4	51.0
7.0 ~ 11.0	138	8.6	9.9	0.1	76.4
4.7 ~ 7.0	138	11.5	8.8	0.7	49.5
3.3 ~ 4.7	138	12.6	9.6	0.9	67.5
2.1 ~ 3.3	138	10.2	9.1	1.0	66.1
Coarse	138	51.3	45.7	3.1	310.5
1.1 ~ 2.1	138	13.5	10.6	0.9	57.6
0.65 ~ 1.1	138	13.7	8.8	0.1	60.9
0.43 ~ 0.65	138	9.8	5.5	0.2	31.3
< 0.43	138	17.4	9.7	2.9	87.2
Fine	138	54.4	34.6	4.1	237.0
Total	138	105.7	80.3	7.2	547.5

3. 2 Inorganic element concentration

Weighted arithmetic mean concentrations in aerosol samples collected during the sampling period among sixteen inorganic species, Si, Ca, Fe, K, Pb, Na, Zn, Mg, Ba, Ni, V, Mn, Cr, Br, Cu, and Co were 2,573.8, 1,102.9, 877.3, 711.8, 245.0, 176.8, 153.0, 139.1, 112.6, 89.6, 68.3, 38.8, 31.1, 28.9, 27.8, and 9.4 ng/m^3, respectively.

The species Ba, Ca, Cr, Co, Fe, K, Mg, Mn, Na, Ni, Si, and V showed higher concentration in coarse particles and Br, Cu, Pb, and Zn showed higher concentration in fine particles. In terms of average seasonal concentration of each inorganic element in fine and coarse particles, Si, Fe, Ca, K, Mg, Na, Ba, Cr, Ni, V, Mn, and Co showed higher value in coarse particles, and Pb, Zn, Br, and Cu showed higher value in fine particles throughout all the four seasons. In addition, most of these inorganic species showed high concentration in spring and winter.

3. 3 Results of TTFA model

To identify source of particulate matters collected in study area and to estimate quantitatively the contribution of the pollution sources, TTFA model was executed with FANTASIA program developed by Hopke *et al.* (1982).

The first step was to determine the number of factors through the variables such as eigenvalue, chi-square, RMS (root-mean-square), and Exner value (Eq. 3). Where x^0 is a grand ensemble average value.

$$E^p = \left[\frac{\sum_{i,j} (x_{ij} - \bar{x}_{ij})^2}{\sum_{i,j} (x^o - \bar{x}_{ij})^2} \right]^{1/2} \quad (3)$$

Tentative number of factors was determined at which these values showed distinct decrease in value. This number was then reduced step by step until the average mass value and scaling factor showed no negative val-

Table 3. Average seasonal source contributions for fine and coarse particles when using TTFA model. (Unit : %)

		Oil burning	Soil related	Field burning	Automobile related	Coal burning	Marine related	Glass related
Fine	Spring	3.5	–	36.9	22.9	33.1	–	3.5
	Summer	9.1	–	24.6	23.4	38.3	–	4.7
	Fall	11.7	–	18.6	26.9	40.5	–	2.2
	Winter	3.3	–	33.3	23.1	37.2	–	3.2
Coarse	Spring	23.3	58.2	5.6	3.5	8.1	1.3	–
	Summer	26.7	49.5	7.7	4.8	8.7	2.7	–
	Fall	27.1	45.0	9.8	4.6	11.1	2.4	–
	Winter	29.2	49.7	4.8	5.0	10.1	1.2	–

ue. Five factors in stage 1, five in stage 2, five in stage 3, five in stage 4, four in stage 5, five in stage 6, four in stage 7, and five in stage 8 were estimated through the above mentioned steps. Since some of factors were overlapped in each stage, a total of seven sources were finally identified in this study area.

The second step was to identify and to specify pollution sources from among the seven sources according to concentration and mass fraction in constituents. The first was oil-burning source, the second was soil related source, the third was open burning source, the fourth was automobile related source, the fifth was coal burning source, the sixth was marine related source, and the last was glass-related source.

The last step was to determine the contribution of each source to the ambient using the source profiles developed in the second step (the matrix F in equation (1)) and the measured data at the receptor (the matrix X in equation (1)). In this study, the summation of stages 1, 2, and 3 was presented as the contribution of fine particles and then summation of stages 4, 5, 6, 7 and 8 was presented as the contribution of coarse particles. Seasonal average contributions of fine particles and coarse particles are also shown in percentage under assumption that the sixteen inorganic species stand for the mass of total aerosol particles (Table 3). Anthropogenic pollution sources such as field-burning, automobile-related, and coal-burning source appeared to be the major contributors of the fine particles throughout the whole seasons, and natural sources such as soil related source and oil burning source appeared to be the major contributor of the coarse particles.

3. 4 Results of PMF model

Data collected during sampling period was applied as raw data in executing PMF model. First step in PMF modeling was to determine the factors, i.e., number of the sources. Too many factors will result in seeking an increase of statistical uncertainties and too few factors will result in hiding sources that do exist in reality. Scale residual matrix R was used in PMF modeling to determine the number of factors. Two values obtained from matrix R, IM (maximum individual column mean) and IS (maximum individual column standard deviation), decreased prominently as the number of factors approached the critical value. Optimum number of factors in both fine and coarse particles was determined as 6 based on IM and IS value criteria. However, since 4 sources were overlapped in both fine and coarse particles, a total of 8 sources were finally identified.

The next step was to identify sources qualitatively using EV (explained variation) value. The sources in fine particles were identified as automobile related, coal burning, field burning, waste incineration, glass related, and oil burning source. For in the coarse particles the sources were identified as marine related, field burning, coal burning, soil related, automobile related, and oil burning source.

The final step was to determine the contribution of sources using the matrix F in equation (2) from the PMF modeling over fine and coarse particles, and the data for the collected aerosol particles (the matrix X in

Table 4. Average seasonal source contributions for fine and coarse particles using PMF model. (Unit : %)

		Oil burning	Soil related	Field burning	Automobile related	Coal burning	Marine related	Glass related	Waste incineration
Fine	Spring	4.1	–	33.6	40.9	5.5	–	7.7	8.1
	Summer	8.8	–	25.1	40.3	8.3	–	11.9	5.7
	Fall	6.5	–	25.4	46.4	7.9	–	7.6	6.3
	Winter	5.0	–	25.5	52.2	7.8	–	7.3	2.3
Coarse	Spring	5.3	69.3	7.9	3.4	8.1	6.0	–	–
	Summer	10.8	40.9	21.4	9.6	10.4	9.9	–	–
	Fall	7.7	47.8	21.6	7.3	9.1	6.4	–	–
	Winter	6.2	61.0	14.0	7.3	6.5	5.0	–	–

equation (2)). Seasonal average contributions of fine and coarse particles are presented in percentage under assumption that the sixteen inorganic elements stand for the mass of total aerosol particles (Table 4).

3. 5 Comparison of contribution results between TTFA and PMF models

Contributions of fine and coarse particles to the receptor were estimated using TTFA and PMF models. The number of estimated sources was seven according to TTFA model and eight according to PMF model. Seasonal contributions for fine particles based on each of these two models are shown in Fig. 2.

According to TTFA model, contributions for the fine particles by open burning, automobile related and coal -burning source in the spring were 37%, 23%, and 33 %, respectively. On the other hand, according to PMF model they were 34%, 40%, and 6%, respectively (Fig. 2(a)). In summer, the contribution by these same sources were 25%, 23%, and 38% respectively by TTFA model, and 25%, 40%, and 8% respectively by PMF model (Fig. 2(b)). In fall, the contributions were 19%, 27% and 40% respectively by TTFA model, and 25%, 47%, and 8% respectively by PMF model (Fig. 2(c)). Finally, in winter, contributions were 33%, 23%, and 37% respectively by TTFA model and 25%, 53%, and 8% respectively by PMF model (Fig. 2(d)).

When comparing contribution results for fine particles obtained from both models, big differences were observed in the automobile related source and the coal -burning source. The results of the other sources were similar each other. In case of automobile related source, the contribution calculated from PMF model was almost twice over-predicted than that from TTFA model. It must be noted that only gasoline related source as an automobile related source was separated by TTFA model; however, both diesel and gasoline related sources were separated together by PMF model.

On the other hand, contribution of coal burning source calculated from TTFA model was about 5 times higher than that from PMF model. According to the Kyunggi Province Statistical Year Book (1998), the amount of coal use in this Province has been markedly decreased since 1990. It was felt that the TTFA result was highly exaggerated when considering the amount of fuel consumption and industrial activities in the study area.

For coarse particles, contributions by oil burning, coal burning, and soil related source in the spring were 23%, 8%, and 58%, respectively, according to TTFA model, and open burning, coal burning, and soil related source were 8%, 8%, and 70% respectively according to PMF model (Fig. 3(a)). In summer, contributions by oil burning, coal burning, and soil related source were 26%, 9%, and 49% respectively according to TTFA model, and oil burning, open burning, and soil related source were 11%, 21%, and 41% respectively according to PMF model (Fig. 3(b)). In fall, contributions by oil burning, coal burning, and soil related source were 27%, 11%, and 45% respectively according to TTFA model, and open burning, coal burning, and soil related source were 21%, 10%, and 48% respectively accord-

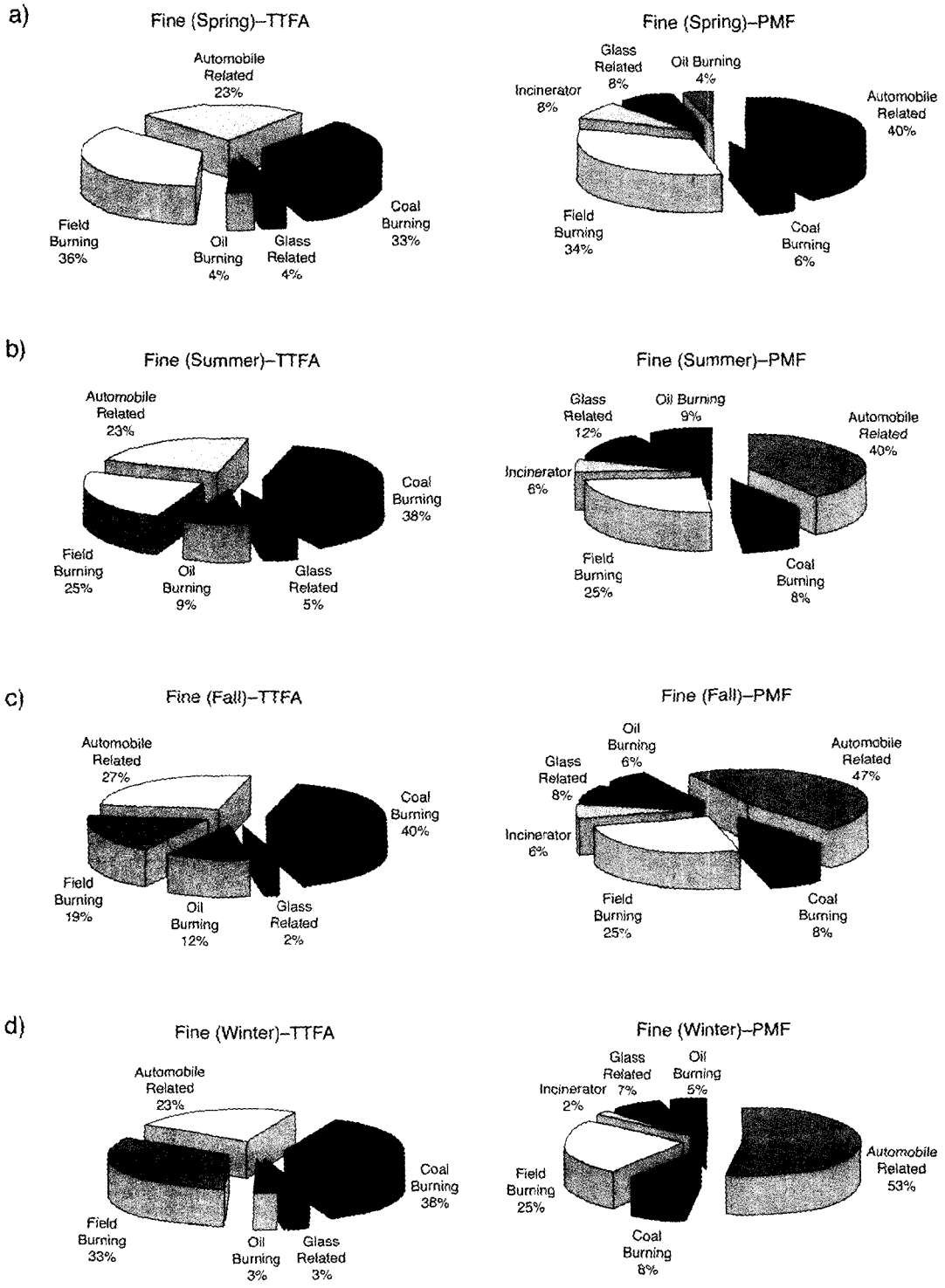

Fig. 2. Comparison of average source contributions (%) calculated by TTFA and PMF model.

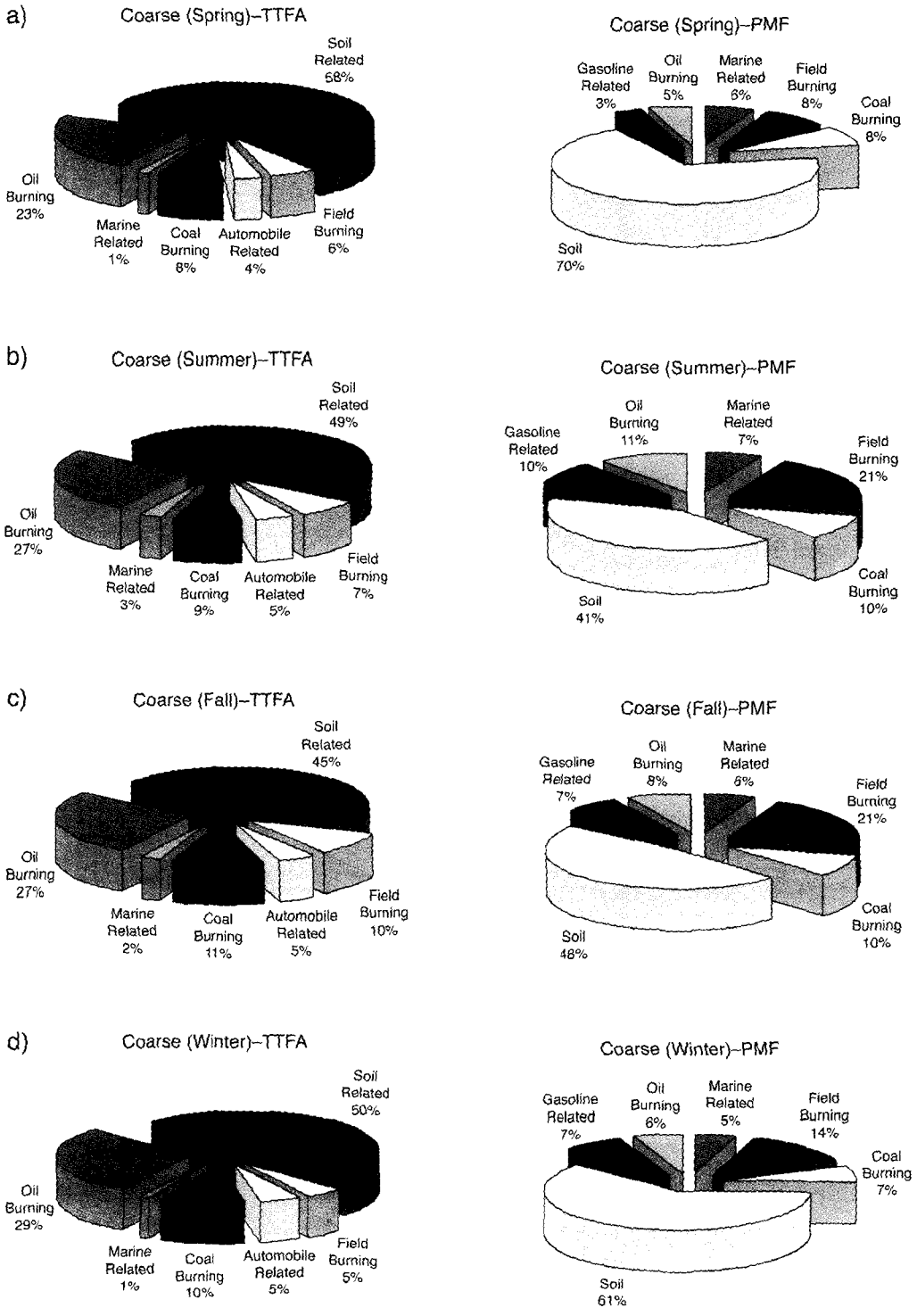

Fig. 3. Comparison of average source contributions (%) calculated by TTFA and PMF model.

ing to PMF model (Fig. 3(c)). Finally in winter, contributions by oil burning, coal burning, and soil related source were 29%, 10%, and 50% respectively according to TTFA model, and open burning, coal burning, and soil related source were 14%, 7%, and 61% respectively according to PMF model (Fig. 3(d)).

When comparing results for coarse particles, contribution of oil burning and field burning sources predicted by TTFA model was in substantial disagreement with that predicted by PMF model. For oil burning source, the contribution calculated from TTFA model was 2 to 5 times over-predicted than that from PMF model. These discrepancies might be occurred in TTFA model that could not well distinguish between oil source and automobile source.

4. CONCLUSIONS

Aerosol samples were collected for seven years by a 9-stage cascade impactor. A total of sixteen inorganic species were analyzed and the concentration trends were examined using these data. The PMF model was used to estimate the quantitative contribution of air pollution sources to the pollution, which was then compared with the contribution results from the TTFA model.

Comparison of the two different methodologies, TTFA and PMF, revealed that both methodologies exhibited similar trends for apportioning aerosol mass. However, large discrepancies between contribution results were observed in some sources. These differences may be reduced, after analysis of more elements and ion compositions, C and S, by improving confidence of models through more precise source estimation, and improvement in the precision of source profile.

In this study, a variety of pollution sources could be identified by the two methodologies of TTFA and PMF. These methodologies, through which the quantitative source apportionment can be calculated without existing source profiles, should be compatible receptor methods with such circumstances in Korea.

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