IDENTIFICATION OF POTENTIAL SOURCES AND SOURCE REGIONS OF AMBIENT AEROSOL IN NORTHEAST ASIA USING ADVANCED HYBRID RECEPTOR MODEL JOINTED WITH POSITIVE MATRIX FACTORIZATION

J. S. Han\textsuperscript{a}, K. J. Moon\textsuperscript{a} and Y. J. Kim\textsuperscript{b}

\textsuperscript{a}Department of Air Quality Research, National Institute of Environmental Research, Environmental Research Complex, Kyeongseo-dong, Seo-gu, Incheon, 404-170, Republic of Korea, first-author: iamiyan@me.go.kr
\textsuperscript{b}ADvanced Environmental Monitoring Research Center at Gwangju Institute of Science and Technology, Oryong-dong, Buk-gu, Gwangju, 500-712, Republic of Korea

ABSTRACT

The size- and time-resolved measurement of particulate trace elements was made using an eight-stage DRUM sampler and S-XRF system from 29 March to 29 May in 2002 at Gosan, Korea, which is one of the representative background sites in East Asia. As a result, continuous 3-hr average concentrations were obtained for 19 elements including S, Si, Al, Fe, Ca, Cl, Cu, Zn, Ti, K, Mn, Pb, Ni, V, Se, As, Rb, Cr, Br. The size-segregated data sets were applied to newly developed hybrid receptor model, advanced concentration weighted trajectory (ACWT) jointed with positive matrix factorization (PMF), in order to identify the possible sources areas and estimate their contributions to particulate matter mass in separate size ranges.

Key Words : Source apportionment, Drum sampler, Receptor model, Positive matrix factorization, Advanced concentration weighted trajectory

1. INTRODUCTION

To develop effective control strategies for anthropogenic pollutants in ambient air, it is necessary to identify the pollution sources and exactly estimate their influence on ambient concentration. For that reason, various receptor models, such as potential source contribution function (PSCF) (Hopke et al., 1993), concentration weighted trajectory (CWT) (Seibert et al., 1994), and residence time weighted concentration (RTWC) (Stohl, 1996), have been developed by using the backward trajectories of air parcels in combination with air quality measurements. However, the commonly used hybrid receptor models have several drawbacks. These statistic receptor models often showed unrealistic source areas for the flux of anthropogenic pollutants influenced by a trailing effect of trajectories in previous works (Han et al., 2004; Lupu and Maenhaut, 2002; Lin et al., 2001).

In this study, advanced concentration weighted trajectory (ACWT) was newly developed in order to complement the weak points in the previous hybrid receptor modeling. This model combines the estimated motion of air backward in time not only with the measured chemical composition of pollutants, but also with the source inventories in the domain. Moreover, the pathways of trajectories that have low intensity of pollutants on arrival are removed from the potential source regions with a
point filter. This feature makes the estimation of hot spot possible and the confidence level of the result increase.

In this study, the result of ACWT hybrid receptor model was especially combined with positive matrix factorization (PMF) analysis while the most previous studies directly applied the composition of collected material to the input data of a receptor model. As a result, it is possible to identify the regional distribution of various aerosol sources impacting the chemical composition of ambient aerosol in Northeast Asia. At this time, PMF analysis has been performed on the Gosan aerosol data collected by a DRUM sampler which has merit to collect the fine particles in six stages below 2.5 μm. The size-segregated composition data with high time resolution are expected to improve the efficiency of PMF analysis and the accuracy of anthropogenic source apportionment.

2. SAMPLING AND ANALYSIS

Ambient aerosol collection using an eight-stage Davis Rotating Unit for Monitoring (DRUM) sampling system was made at the western tip of Gosan, Jeju Island, Korea (33°17' N, 126°10' E, 70m asl), which is a representative background site in East Asia, from 29 March to 29 May 2001. During the measurement period, two Asian dust (AD) outbreaks were observed on 8-10 April and 17 April. In this study, aerosol data pertaining to those AD periods were not subject to PMF and ACWT models in order to focus on the estimation of anthropogenic aerosol sources.

The DRUM sampler collects size-resolved aerosol samples on Apiezon™ coated Mylar™ strips in eight stages, having the equivalent aerodynamic cut-off diameters 0.07, 0.26, 0.34, 0.56, 0.75, 1.15, 2.5, 5.0, and 12 μm (Cahill et al. 1985). The DRUM sampler was operated continuously during the 61-day sampling period. The collected aerosol samples were then analyzed for inorganics (19 elements between aluminum and lead) using synchrotron X-ray fluorescence (S-XRF) at the Lawrence Berkeley National Laboratory Advanced Light Source (Perry et al., 2004). A detailed description of the sampling and analysis methods is provided by Cahill et al. (1993).

3. DESCRIPTION OF MODEL SIMULATION

3.1. Data Analysis By PMF

Two-way Positive Matrix Factorization (PMF2) method was developed by Paatero (Paatero, 1997) to provide flexible modeling approach that effectively uses the information in the data. A robust mode of PMF2 has been selected for handling outlier data, in order to degrade the disproportional affect of excessively large data points (Paatero, 1996). In this study, the value α=4 was chosen for the outlier threshold distance in the present study as done by Lee et al. (1999) and Hien et al. (2004). In addition, The other important parameter of PMF2, F_peak was used to control the rotational freedom until an appropriate distribution of the edges is achieved.

3.2. Trajectory Calculation And Model Details

3-day backward trajectories were computed by a three-dimensional trajectory model, HYPLIT4 (Version 4.7) (Draxler, 1998). In this study, 3-day back-trajectories are used in order to focus on the estimation of the primary aerosol source areas in Northeast Asia. The arrival elevation of 750m is within the average mixed height
during the measurement period (1571m), which means that there could be effective coupling between transport and surface measurement. NCEP/NCAR reanalysis data with 6hr resolution on a $2.5^\circ \times 2.5^\circ$ global grid have been used as meteorological data input to the trajectory model. Backward trajectories were computed by isobaric method eight times a day at 01, 04, 07, 10, 13, 16, 19, 22 hr UTC, corresponding to the measured data.

The position errors for 3-day back-trajectories are of the order of 100km (Stohl, 1998), but the use of the analyzed wind field can make the trajectory uncertainty reduce significantly (Lin et al., 2001). Moreover, the grid size $1^\circ \times 1^\circ$, which is comparable to the uncertainty at the end of a 3-day trajectory, was used in this study, in order to remove the effect of trajectory uncertainty from the results of receptor model. Therefore, the effect of trajectory uncertainty can be efficiently eliminated from the results of receptor model. Figure 1 shows the grid system and the method deriving initial and boundary conditions. The model domains include the most of China, Korea peninsular, Japan, and Taiwan. At this time, China is divided into four regions, including Central China (C China), South China (S China), North West China (NW China), and North East China (NE China), based on the boundary lines of provinces. Most of the simulation results are based on this regional-scale domain.

![Figure 1. Model domain and grid structure.](image)

### 3.3. Advanced Concentration Weighted Trajectory (ACWT)

In the present work, a method of weighting trajectories with associated concentrations (CWT) (Seibert et al., 1994) was improved using source inventory and point filter. It is assumed that the pollutants emitted within grid cell is swept into the air parcel and transported to the receptor site without loss through diffusion, chemical transformation, or atmospheric scavenging (Cheng et al., 1993). This method starts with the first guess concentration field based on Eq. (1). The redistributing concentrations along trajectory $l$, $C_{kl}$ is computed with the equation

$$C_{kl} = C_I \frac{S_{kl} \times N_l}{\sum_{k=1}^{N_c} S_{kl}} = C_I \frac{S_{kl}}{S_l}$$

(1)
where \( l \) is the index of the trajectory, \( C_l \) is the concentration observed on arrival of trajectory \( l \), \( S_k \) is the amount of source emission in segment \( k \) of trajectory \( l \), and \( S_l \) is total source emission produced on the pathway of trajectory \( l \). After the redistribution is finished for all trajectories, the mean weighted concentration calculated for each grid cell \((i, j)\) with the equation

\[
C_y = \frac{1}{\sum_{l=1}^{M} \sum_{i=1}^{N_l} \sum_{k=1}^{N_i} C_{kl}} \times \sum_{l=1}^{M} \sum_{i=1}^{N_l} \sum_{k=1}^{N_i} C_{ijkl} \times \tau_{ijkl}
\]

where \( M \) is the total number of trajectories, \( N_l \) is total number of segments of trajectory \( l \), \( \tau_{ijkl} \) is the residence time of segment \( k \) of trajectory \( l \) in grid cell \((i, j)\). Eq. (2) is similar to the equation of the average weighted concentration in CWT, but the redistributed concentrations \( C_{kl} \) are used instead of the measured concentration \( C_l \). In addition, this method is different from other models such as RWTC in some respects. Especially, it does not use the logarithmic concentrations and the information of source emission was applied to the redistribution of concentration along trajectory \( l \). This feature eliminates the errors by a trailing effect observed in other receptor models and makes the solution physically realistic.

ACWT modeling procedure contains a point filter, which eliminates areas passed by trajectories with low concentration of pollutants. The point elimination was applied to the grid cells that have trajectory endpoints associated with low measured concentration on arrival and lower height level than a constant vertical limit. In the present study, the height of 500m lower than the start points of trajectories was applied to the filtering function

\[
F(i,j) = \begin{cases} 
1.0 & C_i \geq 0.05 \\
0.5 & C_i < 0.05 \land h_{ijkl} \geq 500 \\
0.0 & C_i < 0.05 \land h_{ijkl} < 500 
\end{cases}
\]

where \( h_{ijkl} \) is the height of segment \( k \) of trajectory \( l \) in grid cell \((i, j)\). This restriction is also expected to reduce the trailing effect observed in other receptor models.

In previous works, hybrid receptor models directly applied the measured concentration of each pollutant to calculating the weighted concentration in each segment of trajectories (Han et al., 2004; Lupu and Maenhaut, 2002; Hsu et al., 2003). As a result, the estimated source regions revealed mixed spatial distribution because each pollutant could be emitted by various sources. In this study, ACWT model was associated with PMF results in order to identify the source locations classified by emission sources. The weighting scheme in ACWT method was conducted using not the measured sample concentration, but the source intensity resolved from PMF analysis; \( C_l \) in Eq. (1) is replaced to the source intensity observed on arrival of trajectory \( l \), and \( C_{kl} \) in Eq. (1) and (2) means source intensity of segment \( k \) observed on arrival of trajectory \( l \). The transformed logical algorithm of ACWT model is summarized in Figure 2.
4. EMISSION INVENTORY USED IN THE MODEL

In the present work, several emission databases were used to estimate the aerosol source regions. The emission database for NOx and SO2 are derived from the China Map project, and the emissions from natural sources such as soil dust and Chinese loess are obtained from Earth Science Data Interface of Global Land Cover Facility (GLCF). In addition, industrial SO2 and dust emission data are derived from the database on the emission source inventory in Korea and China’s environment yearbooks. Obtained emission data were then assigned to the model domain with grid size 1° × 1° shown in Figure 1 to redistribute the source intensity along trajectory.

Generally, ambient particulate materials can be produced by a wide range of emission sources including natural and anthropogenic sources. In order to identify the origin and transport pattern for each aerosol source, different source inventories should be used for the individual sources. In the case of the emission sources related to the land cover, such as Chinese aerosol and biomass burning, global land cover data sets (GLCF, 2001) was used to redistribute the source intensities, and the strength of volcano emission was weighted by the SO2 emission inventory by volcano (Carmichael, 2000a). NOx and SO2 emission data in East Asia (Carmichael, 2000b) were used for motor vehicle and coal combustion sources, respectively. Residual oil fired boiler and municipal incineration sources used population distribution to redistribute the source intensity. Finally, the intensities of industrial sources, such as ferrous metal source, copper smelter, oil heating furnace, and nonferrous metal source were rearranged by using province-level pollution data for China (NIPR, 2005; Dasgupta, 1997) and Korea (NIER, 2005).
5. MODEL SIMULATION RESULTS
5.1. Potential Source Intensities Estimated By PMF

In previous work (Han et al., 2005), used parameters and verified result of PMF were described in detail for all size ranges. Overall, twelve distinct primary sources were resolved in fine size ranges (0.07~1.15 μm) for the ambient aerosols collected at the Gosan site in spring of 2001 excluding AD periods. The resolved sources include not only natural sources such as Chinese aerosol, and volcano emission, but also anthropogenic sources including biomass burning, municipal incineration, coal combustion, oil heating furnace, residual oil fired boiler, gasoline vehicle, diesel vehicle, nonferrous metal source, ferrous metal sources, and copper smelter. Figure 3 represents the corresponding temporal variations of these potential sources in fine size range (0.07~1.15 μm). The mass concentration of each source was calculated from the sum of scaled intensity values in the resolved size ranges. Overall, apparent differences in temporal variations of these anthropogenic sources confirm the independence of the estimated source contributions.

![Figure 3: Temporal variations of fine PM mass (0.07~1.15 μm) by each of the resolved sources during the non-AD periods.](image1)

The average source contributions in fine size range (0.07~1.15 μm) are shown in Figure 4. The contribution of natural sources including Chinese aerosol and volcano emission, was much lower than that of anthropogenic sources. Especially, the contributions of diesel vehicle, biomass burning, and coal combustion were larger than other anthropogenic sources, accounting for 29.5%, 25.3%, and 23.3%, respectively. Vehicle emission sources including gasoline and diesel vehicle

![Figure 4: Temporal variations of fine PM mass (0.07~1.15 μm) by each of the resolved sources during the non-AD periods.](image2)
occupied about 32%, the largest portion of the fine aerosol mass. In addition, the sources related to fossil fuel combustion contributed to more than 30%.

Figure 5. Average source contributions to the mass concentration of fine particle (0.07~1.15 μm).

5.2. ACWT Results
ACWT model was applied to the twelve aerosol sources resolved from PMF in fine size range (0.07~1.15 μm). Overall, apparent differences in spatial distributions of these potential sources confirm the independence of the estimated source contributions. Moreover, the strong spatial gradients of flux value effectively separate the large source areas in the field.

The potential areas of natural sources were more obviously distinguished from other anthropogenic sources as shown in Figure 4. The ACWT results for Chinese aerosol showed that the sources areas were mainly located in the bare ground and sand desert in Northeast China. This region partially includes Gobi sand desert and loess plateau in Central China. Because the AD periods were eliminated from the PMF analysis, this result indicates that the long-range transport of small mineral particles from China could be occurred during NAD periods in dry spring season.

Although the source intensity was smaller than other sources, the influence of volcano emission produced in Japan apparently observed. The concentration gradients of volcano emission show that the area below the island of Kyushu was a hot spot source. In the region, there are several active volcanoes, which intermittently produce a large amount of ash and gas plume.

Fuel combustion sources contributing more than 1% to the fine PM mass such as diesel vehicle, coal combustion, and biomass burning were also distinguished by the flux distribution. The emission flux of diesel vehicle was concentrated around the populated and industrial centers, which includes Taiwan, major industrial areas in China including Shanghai, Shenyang, Anshan, and Dalian, and central metropolitan region in Korea. The marine areas located between Taiwan and Japan were also suggested to be moderate source areas of diesel vehicle influenced by the movements of shipping. Compared with the results for diesel vehicle, the possible source areas for coal combustion were more widely distributed to the inland provinces. The flux of coal combustion source was not only concentrated around the coastal provinces
from Guangdong north to Liaoning, but also centered on a west-to-east belt across
the inland of China, from Hubei Province to Hebei Province.
In the case of biomass burning, the spatial distribution pattern was different with
other industrial anthropogenic sources that closely tied to industrial centers. ACWT
model resolved the cultivated regions in South and Central China as the main
biomass burning sources impacting the ambient aerosol at Gosan, Korea. The region
contains the rich Yangzi River Basin producing about 40% of total grain in China.
Therefore, it is inferred that field combustion of agricultural residues considerably
emitted fine particles in this area. In addition, the source regions were widely spread
over the grassland and forest located in Northeast China implying the impact of
biofuel combustion.
The potential source areas of gasoline vehicle and oil fired boiler were also different.
The source areas of gasoline vehicle were mainly distributed in Central and South
China centering around Taiwan, Guangzou, and Shanghai while the flux of oil fired
boiler source was concentrated around Northeast China including Liaoning and Jilin
provinces.
Major industrial areas such as Anshan, Beijing, Zheongzhou, Hangzhou, and
Guangzhou were resolved as the potential source areas of ferrous metal source. the
major source locations of the oil-heating furnace were also similar to those for
ferrous metal source implying these industrial sources can be mutually related. The
source locations of copper smelter were a little different with these two sources.
They were only observed in Shanghai and Beijing industrial areas, and central Korea.
Figure 4. Potential source areas of twelve possible aerosol sources impacting the chemical composition of ambient aerosol at Gosan, Korea over the study period.

REFERENCES

Carmichael, G., 2001b, Datasets (Year 2000) – NOx (1°×1°, Area), URL: http://www.cger.uiowa.edu/people/carmichael/ACESS/nox_area_1deg.txt (assessed in February, 2005).