Study on the long-range transport of particulate pollutants by using DRUM sampler during ACE-Asia

K. J. Moon, J. S. Han, Y. D. Hong, Y. J. Kim, S. S. Cliff[1], K. D. Perry[1], T. A. Cahill[1]

Air Quality Division, National Institute of Environmental Research (NIER), Environmental Research Complex, Kyeongseodong, Seogu, Incheon, 404-170, Republic of Korea, nierhan@me.go.kr, iamiyan@me.go.kr, Tel: +82-32-560-7103, Fax: +82-32-568-2035

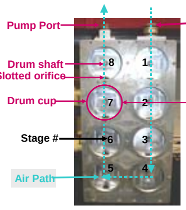
INTRODUCTION

- It is necessary to identify aerosol sources and estimate their influence on ambient PM concentration in order to formulate effective control strategies for ambient particulate matter.
- Recently, the improvement of sampling technique makes it possible to extract much information on the pollution source.
- This study presents the analysis results of size- and time-resolved samples simultaneously collected using an eight-stage Davis rotating unit for monitoring (DRUM) sampler from 24 March to 18 April in 2001 at Gosan, Korea and at Beijing, China during the Asia-Pacific Regional Aerosol Characterization Experiment (ACE-Asia).
- The size-resolved data sets were then analyzed using the positive matrix factorization (PMF) technique in order to identify possible sources and estimate their contribution to particulate matter mass.

SAMPLING AND ANALYSIS

- Ambient aerosol collection was continuously made at Gosan super site (33.29°N, 126.16°E, 78m ASL), Beijing (39.92°N, 116.42°E, 55m ASL), Hefei (31.90°N, 117.16°E), and Tango (35.7°N, 135.17°E, 600m ASL) from 24 March to 18 April 2001.
- One Asian dust outbreak (4/10~14) was observed during ACE-Asia.

➔ Preferentially, the collected data at Gosan and Beijing were analyzed in detail in order to estimate the possibility of long-range transport of particulate pollutants.



- The DRUM sampler continuously collects the size-resolved aerosol samples on ApiezonTM coated MylarTM strips in eight stages. The equivalent aerodynamic cut-off diameter of each stage is 0.07, 0.24, 0.34, 0.56, 0.75, 1.15, 2.5, 5.0㎛.

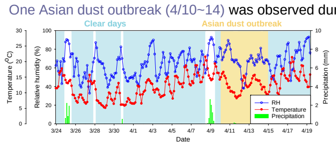
➔ The collected aerosol samples were then analyzed for 19 elements (S, Si, Al, Fe, Ca, Cl, Cu, Zn, Ti, K, Mn, Pb, Ni, V, Se, As, Rb, Cr, Br) using synchrotron X-ray fluorescence by DELTA Group in UC Davis.

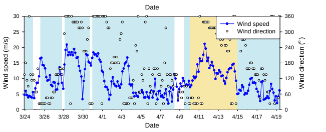
XRF analysis conditions using beamline 10.3.1

Photon Energy Range (keV)	Monochromator	Photon Flux (Photons/s)	Spectral Resolution (E/△E)	Spatial Resolution (mm)	Detectors	Sensitivity of Detection
6-15 (with multiplayer mirrors)	White light, multiplayer mirrors in Kirkpatrick-Baez configuration	3×10^{10} (at 12.5 keV)	25 (at 12.5 keV)	1.0×1.0	Si(Li) x-ray detector	~0.1ng/㎥

ATMOSPHERIC CONDITION

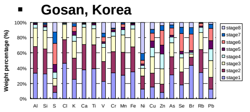
- One Asian dust outbreak (4/10~14) was observed during ACE-Asia.





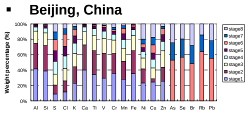
SIZE DISTRIBUTION OF TRACE ELEMENTS

- **Gosan, Korea**



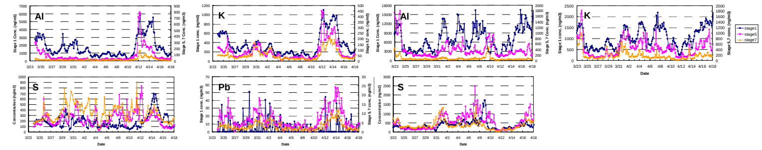
stage 1: 5.0㎛~Inlet,
stage 2: 2.5~5.0㎛,
stage 3: 1.15~2.5㎛,
stage 4: 0.75~1.15㎛,
stage 5: 0.56~0.75㎛,
stage 6: 0.34~0.56㎛,
stage 7: 0.26~0.34㎛,
stage 8: 0.07~0.24㎛

- **Beijing, China**



stage 1: 5.0㎛~Inlet,
stage 2: 2.5~5.0㎛,
stage 3: 1.15~2.5㎛,
stage 4: 0.75~1.15㎛,
stage 5: 0.56~0.75㎛,
stage 6: 0.34~0.56㎛,
stage 7: 0.26~0.34㎛,
stage 8: 0.07~0.24㎛

As, Se, Br, Rb, Pb have 1~5 Stage data



DATA ANALYSIS USING PMF

- **PMF (Two-way Positive Matrix Factorization, Paatero, 1997) analysis** is applied to the size-segregated measured data.

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

(g_{ik} : source contributions, f_{kj} : source compositions, e_{ij} : residuals)

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left(\frac{e_{ij}}{s_{ij}}\right)^2 \quad (2)$$

($g_{ik} \geq 0$ and $f_{kj} \geq 0$, s_{ij} : error estimate for x_{ij})

$$e_{ij} = x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj} \quad (3)$$

(p= the number of factor extracted)

Robust mode (α=4), Fpeak parameters are used.

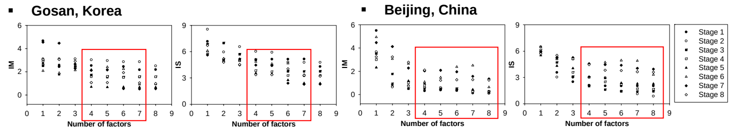
- The information from the scaled residual matrix (R , $r_{ij}=e_{ij}/s_{ij}$) in PMF is used to reduce the ambiguity due to manual judgment on the number of factors (Lee et al., 1999).

⇒ IM (the maximum individual column mean) : $IM = \max_{j=1...m}\left(\frac{1}{n}\sum_{i=1}^{n} r_{ij}\right)$ (4)

IS (the maximum individual column standard deviation) : $IS = \max_{j=1...m}\left(\sqrt{\frac{1}{n-1}\sum_{i=1}^{n}(r_{ij}-\bar{r}_j)^2}\right)$ (5)

- F_{peak} value is determined in a range within which Q value (2) remains relatively constant to reduce the ambiguity due to the rotational freedom.
- At this time, the largest element in *Rotmat* (rotational matrix) have to be as low as possible.

- **Gosan, Korea** · **Beijing, China**



PMF RESULTS

1. Source identification

- Overall, ten distinct primary sources were resolved for the ambient aerosols collected at Gosan and Beijing during ACE-Asia, respectively.

Source	Marker element	Stage 1	Stage 2	Stage 3	Stage 4	Stage 5	Stage 6	Stage 7	Stage 8
1 Continental soil	Si, Al, Fe, Ca, K, S, Pb	+	+	+	+	+	+	+	+
2 Local soil	Si, Al, Fe, K, Ti, Ca	+	+	+	+	-	-	-	-
3 Sea salt	Cl, S, K, Br	+	+	+	-	-	-	-	-
4 Biomass burning	S, K, Cl, Si, Al, BC	-	-	-	+	+	+	+	+
5 Municipal incineration	Cl, Fe, S, Al, Ca, Zn, Pb, BC	+	+	-	-	-	-	-	-
6 Coal combustion	S, Si, K, Zn, Fe, As, Se, BC	-	-	+	+	+	+	+	+
7 Oil fired boiler	S, V, Si, Ni, BC	-	-	-	+	+	+	+	+
8 Gasoline vehicle	S, Si, Ca Fe, Zn, Cl, K, BC	-	-	-	-	+	+	+	+
9 Diesel vehicle	Si, S, Al, K, BC	-	-	-	-	-	-	+	+
10 Ferrous metal source	Fe, Si, Al, K, Zn, Mn	-	+	+	+	+	-	-	-
Number of factor		4	4	4	5	5	7	7	7
Chi square		10547	10565	9008	11054	7009	8132	4132	7022
Fpeak		0.0	0.0	-0.2	0.0	0.0	0.0	-0.2	0.0
Robust mode (α)		2	2	2	2	2	2	2	2

Source	Marker element	Stage 1	Stage 2	Stage 3	Stage 4	Stage 5	Stage 6	Stage 7	Stage 8
1 Continental soil	Si, Al, Fe, Ca, K, S, Pb	+	+	+	+				
2 Local soil	Si, Al, Fe, K, Ti, Ca	+	+	+	+				
3 Biomass burning	S, K, Cl, Si, Al, BC					+	+	+	+
4 Municipal incineration	Cl, Fe, S, Al, Ca, Zn, Pb, BC	+	+	+					
5 Coal combustion	S, Si, K, Zn, Fe, As, Se, BC						+	+	+
6 Oil fired boiler	S, V, Si, Ni, BC					+	+	+	+
7 Gasoline vehicle	S, Si, Ca Fe, Zn, Cl, K, BC				+	+	+	+	+
8 Diesel vehicle	Si, S, Al, K, BC						+	+	+
9 Ferrous metal source	Fe, Si, Al, K, Zn, Mn	+	+	+	+	+	+	+	+
10 Nonferrous metal source	Cu, S, Zn, Fe, Cr, Pb				+	+	+	+	+
Number of factor		4	4	4	5	6	7	6	7
Chi square		3835	2407	4578	6350	7006	13715	13415	13074
Fpeak		-0.1	-0.2	-0.2	0.0	0.0	0.0	0.0	-0.2
Robust mode (α)		2	2	2	2	2	2	2	2

stage 1: 5.0㎛~Inlet, stage 2: 2.5~5.0㎛, stage 3: 1.15~2.5㎛, stage 4: 0.75~1.15㎛, stage 5: 0.56~0.75㎛, stage 6: 0.34~0.56㎛, stage 7: 0.26~0.34㎛, stage 8: 0.07~0.24㎛

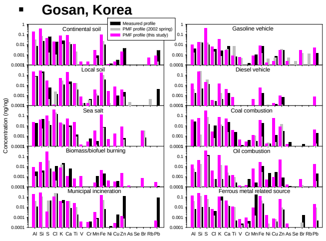
- Corresponding sources of the resolved sources were estimated from the index species of various sources and the known profiles obtained from previous works.

➔ Among the resolved sources at Gosan, Korea, eight sources were overlapped with the ones extracted at Beijing, China:

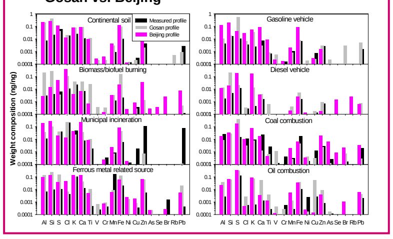
- **continental soil**
- **biomass/biofuel burning**
- **municipal incineration**
- **ferrous metal related source**
- **gasoline vehicle**
- **diesel vehicle**
- **coal combustion**
- **oil combustion**

2. Average profiles of the resolved sources

- **Gosan, Korea**



- **Gosan vs. Beijing**



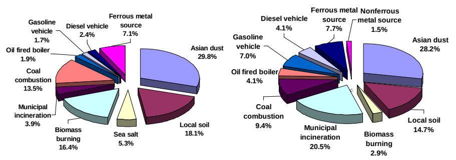
Source	Reference	Source	Reference
Chinese aerosol	He et al., 2001	Oil fired boiler	EPA profile 13505
Soil dust	EPA profile 41340	Gasoline vehicle	Watson et al, 1994
Marine aerosol	Watson, 1979	Diesel vehicle	Watson et al, 1994
Field burning	EPA profile 42320	Ferrous metal source	Watson, 1979
Municipal incineration	EPA profile 17106		
Coal combustion	Mamuro et al, 1979		

➔ The resolved source profiles were similar with the other ones obtained from the data measured in spring in 2002.

➔ In addition, common sources at two sites showed considerably similar average profiles.

3. Total source contributions

- **Gosan, Korea** · **Beijing, China**

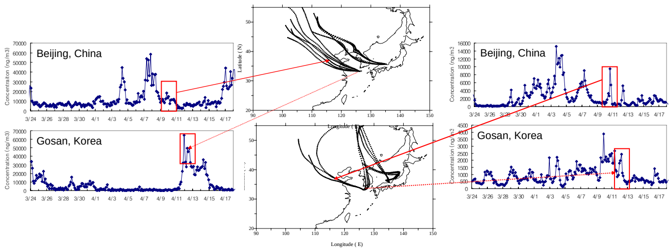


➔ The total amount of particulate matter at Beijing was more than 3 times higher than that at Gosan.

➔ The contribution of natural source containing sea salt, continental and local soil was more than 50% at Gosan while the anthropogenic sources largely contributed at Beijing such as municipal incineration.

4. Temporal variation of the resolved sources

- In order to investigate the long-range transport of particulate pollutant, the time series of source contribution at two sampling sites were compared with the air mass trajectories.
- Comparison of temporal variation of source contribution and air mass trajectories suggested that several common sources could be transported from the continent to background region during subdivided periods.



SUMMARY AND CONCLUSION

- The long-range transport of particulate pollutants was investigated by using the size- and time-resolved DRUM data and PMF receptor model.
- As a result, ten sources were resolved in eight size ranges (0.07~12 um) at Gosan and Beijing, and eight sources were commonly observed at two sites.
- Common eight sources included continental soil, biomass/biofuel burning, municipal incineration, ferrous metal related source, gasoline vehicle, diesel vehicle, coal combustion, and oil combustion.
- The average profiles of them in different size ranges were considerably similar at two sites implying that these sources could be originated from same emission source.
- In addition, the time series of several source intensities at two sites suggested the possibility of long-range transport during sectional periods.