J. of the Environmental Sciences
Vol. 12, No. 2, 217~225, 2003

A Study on the Source Apportionment of the Atmospheric Fine Particles in Jeju area

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(Manuscript received 23 December, 2002 ; accepted 14 February, 2003)

Samples of size-fractionated PM10 (airborne particulate matter with aerodynamic diameter less than 10 ㎛) were collected at an urban site in Jeju city from May to September 2002. The mass concentration and chemical composition of the samples were measured. The data sets were then applied to the CMB receptor model to estimate the source contribution of PM10 in Jeju area. The average PM10 mass concentration was 28.80㎍/m³ (24.6~33.49㎍/m³), and the FP (fine particle with aerodynamic diameter less than 2.1㎛) fraction in PM10 was approximately 8% higher than the CP (coarse particle with aerodynamic diameter greater than 2.1㎛ and less than 10㎛) fraction in PM10. The CP composition was obviously different from the FP composition, that is, the most abundant water soluble species was nitrate ion in the FP, but sulfate ion in the CP. Also sulfur was the most dominant element in the FP, however, sodium was that in the CP. From CMB receptor model results, it was found that road dust was the largest contributor to the CP mass concentration (45% of the CP) and ammonium nitrate, domestic boiler, and marine aerosol were major sources to the CP mass. However, the secondary aerosol was the most significant contributor to the FP mass concentration (45% of the FP). In this study, it was suggested that the contributions of soil dust and gasoline vehicle became very low due to collinearity with road dust and diesel vehicle, respectively.

Key words : PM10, Composition, Source apportionment, Chemical Mass balance(CMB), Jeju

1. Introduction

The atmospheric aerosols are concern since they have severe effects on public health, visibility, and weather changes. The aerosols are originated from not only anthropogenic sources such as industry, power plant, transportation, and construction but also natural sources such as marine aerosol, soil dust, and volcanic ashes. Especially the fine airborne particles have long lifetime in ambient air and are able to be transported long distance. Hence, in an clean-air area such as Jeju island, the aerosol levels in ambient air are influenced by the surrounding areas[1,2].

The Jeju island is located in the East China Sea, 100㎞ south of the Korean peninsula, 250 ㎞ west of Kyushu, Japan, 500㎞ east-northeast of Shanghai, China, and 1000㎞ north-northeast of Taipei. This island is considered as one of the most clean-air area in Korea with no large industrial sources and low population density, so aerosol levels in Jeju island are considered as background levels which may be affected by long-range transport of aerosols from outside of Jeju island. Thus, Jeju island is an ideal place for investigating the long-range transport of air pollutants in this region[3].

A few studies have been carried out in relation to the aerosol in the Jeju island since 1991. These studies have mainly focused on the aerosol background levels, the characterization and the composition of aerosol, and the seasonal variations of aerosol mass concentration. In addition, most of these have been performed at Kosan site located about 50㎞ west-south of Jeju city, but only a few studies

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have been carried out in Jeju city[4~13]. No the source apportionment studies of the aerosol in Jeju area have been performed, although it is very important to apportion aerosol source in order to not only well manage regional air quality but also understand long-range transport of the aerosols in Northeastern Asia.

So many studies in relation to aerosol source apportionment have been carried out in our country as well as other countries. And a lot of the source apportionment methods have been developed such as receptor models including chemical mass balance(CMB)models and multivariate models, and dispersion models. Most of the domestic studies have been performed in connection with not methods but application. However, no aerosol source apportionment studies have been reported yet for Jeju area[14~23].

In this study, the measurements of the mass concentration and the chemical composition for PM10(airborne particulate matter with aerodynamic diameter less than 10㎛), collected at an site in Jeju city, are carried out in order to get the data sets for estimating aerosol source contribution. And then the source apportionment of PM10 is performed using the EPA CMB8 receptor model. In this paper, the PM10 mass concentration and composition, and the results of PM10 source apportionment are presented and their characteristics are discussed.

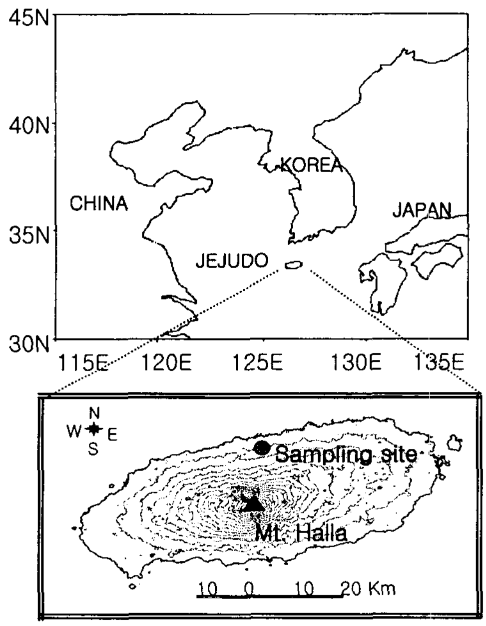

Fig. 1. The location of sampling site and surrounding area.

2. Methods

2.1 Aerosol sampling

Aerosol samples were collected at an site in Jeju city, Jeju island. The locations of the sampling site and its surrounding area are shown in Fig.1. The sampling site is in an urban area and very close to major traffic roads, but there is no major industry nearby. And the site is located approximately 2㎞ away from seashore.

The sampling was carried out from May to September 2002, and the sampling time of each sample was shown in Table 1. Aerosol samples were collected on membrane filters (size ϕ 80mm, pore size 0.1㎛), using 8 stage non-viable cascade impactor, operated at a flow rate of 28.3ℓ/min. This sampler consists of eight stages, and the particles in ambient air is collected separately with aerodynamic diameter on each stage ; 9.0~10㎛ on top stage, 5.8~9.0 ㎛ on 2nd stage, 3.3~4.7㎛ on 3rd stage, 2.1~3.3㎛ on 4th stage, 1.1~2.1㎛ on 5th stage, 0.65~1.1㎛ on 6th stage, 0.43~0.65㎛ on 7th stage, under 0.43㎛ on bottom stage.

2.2 Chemical analysis

After sampling, the membrane filters were stabilized 24hours prior to weighing, corrected by the average weight of the controls, and then weighed to determine the mass concentration of particles in ambient air. And then the filters were cut in half for the chemical

Table 1. Sampling time of each sample

Sampling site	Sample No.	Sampling Period	Sampling Time
	C1	05/18/02~06/01/02	13 days
	C2	06/03/02~06/12/02	9 days
	C3	06/15/02~07/03/02	18 days
Jeju city	C4	07/25/02~08/08/02	14 days
	C5	08/09/02~08/26/02	17 days
	C6	09/01/02~09/16/02	15 days
	C7	09/17/02~10/02/02	15 days

analysis of water soluble ionic species and inorganic elements.

Half of the membrane filters were immersed in 30mℓ distilled water and extracted for 30min using an ultrasonic cleaner. The extracts from the filters were analyzed using an Ion Chromatography method to determine the concentration of the water soluble ions. The residue was wet digested with 12mℓ mixture of nitric acid and hydrochloric acid (HNO_3 : HCl = 3:1) at 175℃, 350psi for 15min. This acid digested solution was filtered through a membrane filter (pore size 0.45㎛), and then the filtrate was analyzed using an Inductively Coupled Plasma method to determine the elements concentration in the sample. Filter and reagent blanks were also taken through the same procedure simultaneously. The analytical items and methods were presented in Table 2.

2.3 Chemical mass balance receptor model

Receptor models use chemical and physical characteristics of particles measured at source and receptor to both define the emission profiles of source and also evaluate their contribution to a specific receptor. Receptor models can be classified into several categories : chemical mass balance(CMB), multivariate analysis, microscopic methods, and source-receptor hybrids. The most commonly applied method is the CMB analysis[24].

The theoretical basis of the CMB model is the assumption of mass conservation between receptor and source if there is no selective elemental removal of particle formation. Therefore, the total particulate mass, M, is the sum of the contributions from each of the individual sources, f_k for each of the p sources:

$$M=\sum_{k=1}^{p} f_k$$

Table 2. Analytical methods used in this study

Species	Analytical method	Instrument Model
Na, Mg, S, Ca, Ti, V, Mn, Fe, Cu, Zn, Cr, Ba, Pb, Al, K, Ni, Cd, Sr	Inductively Coupled Plasma (ICP)	IRIS Advantage
Ca^{2+}, Na^{+}, K^{+}, Mg^{2+}, NH^{+}, Cl^{-}, NO_3^{-}, SO_4^{2-}	Ion Chromatography (IC)	Metrohm module

For the elemental concentrations, the expression is written as

$$x_i=\sum_{k=1}^{p} a_{ik} f_k\ , \text{ for } i=1\sim n$$

where a_{ik} is the concentration of the ith element in particles from the kth source, n is the number of measured elements and x_i is the measured ambient concentration the ith element. In vector form

$$X=Af$$

where X is a n-vector of ambient concentrations and f is a p-vector of source concentrations, respectively, while A is the $n \times p$ matrix of source profiles.

The EPA CMB8 receptor model was applied to PM10 measurements in Jeju city following the EPA application and validation protocols. The model results were evaluated by using several fit indices such as R square, CHI square, and percent of mass accounted for. The runs were performed to avoid collinearity, which means that some individual sources have similar source profiles that yield to unreliable values if included in the same chemical mass balance[23].

3. Results and Discussion

3.1 Particle mass concentration

The measured PM10 (particulate matter with aerodynamic diameter less than 10㎛) mass concentrations in the ambient air in Jeju city were given in Table 3. The FP (fine particle fraction in PM10, with aerodynamic diameter less than 2.1㎛) and the CP (coarse particle fraction in PM10, with aerodynamic diameter ranges from 2.1 to 10㎛) mass concentrations and F/C (FP/CP) ratios were also given in the Table 3.

Average PM10 mass concentration was 28.80 ㎍/㎥ (range from 24.06 to 33.49㎍/㎥) in Jeju city from May to September 2002. This value is significantly lower than the Korean ambient air quality standard of PM10(70㎍/㎥), and the PM10 concentrations measured in Seoul(89.2㎍/㎥), Suwon(65.8㎍/㎥), and Daegu area(75㎍/㎥)[25~27]. And the mean concentration of the FP was 16.26 ㎍/㎥(13.95~19.04㎍/㎥). This is similar to the PM2.5 mass concentration reported in Kosan known as ideal background monitoring site in Jeju island, 16.6$\mu g/m^3$. However, the value is much lower than the PM2.5 mass concentration measured in Kangwha, Korea, 29.1$\mu g/m^3$[28]. It is easily noted that Jeju city is very clean area in comparison with other area. The FP mass concentrations were higher than those of the CP in overall aerosol samples, that is, the average F/C was 1.3, ranged from 1.15 to 1.58.

3.2 Chemical composition of particle

Table 4 shows the average water soluble ionic concentrations in the coarse(CP) and the fine aerosol fraction(FP) in Jeju city. The compositions of the CP was apparently different from those of the FP. Nitrate ion was a most abundant species in the CP, on the other hand sulfate ion was a most abundant species in the FP. The relative differences between the maximum and the minimum concentration of sodium and chloride ion were remarkably greater than those of the other ions. This is suspected that the sodium and chloride ion, which mostly originated from sea salt, is significantly affected by the meteorological conditions in aerosol sampling duration, especially wind speed and direction.

Table 3. Mass concentrations and F/C ratios of PM10 in Jeju city (unit : $\mu g/m^3$)

Sample No.	C1	C2	C3	C4	C5	C6	C7	Average
FP*	17.29	14.72	16.59	13.95	14.10	19.04	18.14	16.26
CP**	12.83	9.34	13.44	11.38	12.30	13.11	15.35	12.54
PM10	31.12	24.06	30.03	25.33	26.40	32.15	33.49	28.80
F/C*** Ratio	1.35	1.58	1.23	1.23	1.15	1.45	1.18	1.30

* FP : Fine suspended particle ($d_p < 2.1\mu m$)
** CP : Coarse suspended particle ($2.1 < d_p < 10\mu m$)
*** F/C Ratio : Mass concentration ratio of the FP to the CP

Table 4. Concentration of water soluble ions in PM10 in Jeju city (unit : $\mu g/m^3$)

		Water soluble cations					Water soluble anions		
		Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	SO_4^{2-}
CP	Average	0.473	0.165	0.035	0.204	0.047	0.610	0.998	0.512
	SD	0.371	0.055	0.016	0.068	0.024	0.474	0.166	0.133
	Minimum	0.169	0.091	0.013	0.144	0.020	0.190	0.810	0.352
	Maximum	1.163	0.236	0.055	0.302	0.092	1.522	1.293	0.732
FP	Average	0.279	1.333	0.158	0.088	0.035	0.232	1.123	3.761
	SD	0.155	0.374	0.078	0.031	0.011	0.140	0.165	0.887
	Minimum	0.154	0.882	0.027	0.045	0.018	0.066	0.903	2.610
	Maximum	0.609	1.722	0.229	0.124	0.048	0.458	1.306	4.773

The component fractions of the water soluble cations in the CP were 51% Na^+, 22% Ca^{2+}, 18% NH_4^+, 5% Mg^{2+}, and 4% K^+, however, those in the FP were 70% NH_4^+, 15% Na^+, 8% K^+, 5% Ca^{2+}, and 2% Mg^{2+}. Also the composition of the water soluble anions in the CP was certainly different from those in the FP. That is, the mass fractions of each anion in the CP were 46% NO_3^-, 30% Cl^-, and 24% SO_4^{2-}, respectively, but those in the FP were 73% SO_4^{2-}, 22% NO_3^-, and 5% Cl^-, respectively. From these results, it suggests that NH_4^+ in the FP could be mainly as forms of $(NH_4)_2SO_4$ and $(NH_4)_3H(SO_4)_2$ but not of NH_4NO_3[28].

The elemental concentrations in the fine and the coarse particulate matter collected from May to September 2002 in Jeju city were presented in Fig. 2. They can be classified into the major elements group whose average concentrations are approximately higher than 100$\mu g/m^3$, and the minor elements group whose average concentrations are lower than 100$\mu g/m^3$. S, Na, Ca, Fe,

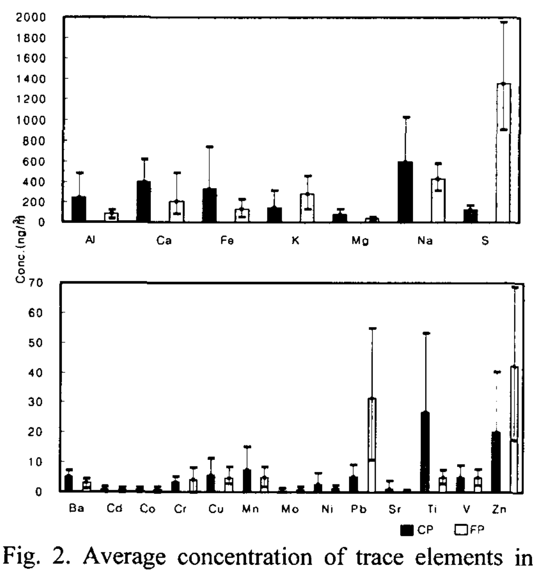

Fig. 2. Average concentration of trace elements in PM10.

Al, K, and Mg belong to the major elements group, on the other hand Zn, Ti, Pb, V, Sr, Ni, Mo, Mn, Cu, Cr, Co, Cd, and Ba are classified into the minor elements group in this study. This result has similar trend to the elemental concentration of TSP collected at Sungsan in Jeju island[7].

Sulfur is the most dominant element in the FP, however, the most dominant element in the CP is sodium. The elements originated mainly from natural sources, such as Al, Ca, Fe, Na, are richer in the CP, on the contrary the elements from anthropogenic sources, such as S, Zn, and Pb are richer in the FP. And it is found that the elements concentration from natural sources is evidently higher than those from anthropogenic sources except sulfur[29].

3.3 Source apportionment using CMB Model

The EPA CMB8 receptor model was applied to the mass concentration and the chemical composition data sets obtained from the aerosol samples collected from May to September 2002 in Jeju city, in order to assess quantitatively the contributions from various sources. The following sources were suspected to contribute to the Jeju area aerosol, and were therefore included in the CMB : domestic boiler used kerosene(DOBOK), external combustion used light oil(EXCL), municipal incinerator(INCIN), cement dust(CDUST), diesel vehicles(DISEL), gasoline vehicles(GASL), road dust(ROAD), soil dust(SOILD), marine aerosol(MARIN), ammonium sulfate(AMSUL), and ammonium nitrate (AMNIT). As no source profile measurements were carried out in the Jeju area, the required source profiles were obtained from the VOC/PM speciate data system of U.S. EPA.

The CMB analysis were carried out on both the 7 FP data sets and the 7 CP data sets, separately. And then, the CMB results were evaluated by using several fit indices such as R-square, CHI square, and percent of mass accounted for. The results of the CMB evaluation are summarized in Table 5. As shown in Table 5, the fit indices value of both the CP in sample C1, C2, and C3 and the FP in sample C1, C4 and C7 are significantly exceeded the criteria of the EPA application and validation protocols. Hence, the CMB results from those data sets were excluded to estimate source contribution.

The source contributions estimated using the CMB model are presented in Fig. 3 and Fig. 4 for the CP and the FP, respectively. The source contributions to the CP concentration data sets are not similar to each other, but it is easily noted that the largest contributor to the CP is road dust. It is also found that marine aerosol, ammonium nitrate, and domestic boiler are major contributor to the CP. Soil dust is generally known as major contributor to the coarse aerosol, although, its contribution to the CP mass is insignificant in this study. It is guessed that the collinearity between road dust and soil dust due to similar composition causes this results. For the FP, the secondary aerosols (ammonium sulfate and ammonium nitrate) contribution is remarkably higher than those for the CP. And road dust is the largest contributor to the FP, however, its contribution fraction is much lower than that for the CP. Also it is presented that road dust, ammonium sulfate, ammonium nitrate, diesel vehicle, and marine aerosol mainly contribute to the FP mass concentration.

Table 5. The values of R square, CHI square, and % mass in the CMB results

Sample		C1		C2		C3		C4		C5		C6		C7	
fit index	criteria	C*	F**	C	F	C	F	C	F	C	F	C	F	C	F
R^2	0.8～1.0	0.97	0.94	0.97	0.96	0.96	0.96	0.95	0.95	0.95	0.99	0.99	0.97	0.94	0.95
x^2	0.0～4.0	1.31	4.81	3.95	3.59	2.63	3.45	3.99	6.72	4.74	3.88	3.89	2.00	5.59	5.95
%Mass	80～120	528	198	465	153	671	147	137	175	89	138	106	137	132	217

C* : Coarse suspended particles (CP, 2.1㎛ < dp < 10㎛)
F**: Fine suspended particles (FP, dp < 2.1㎛)

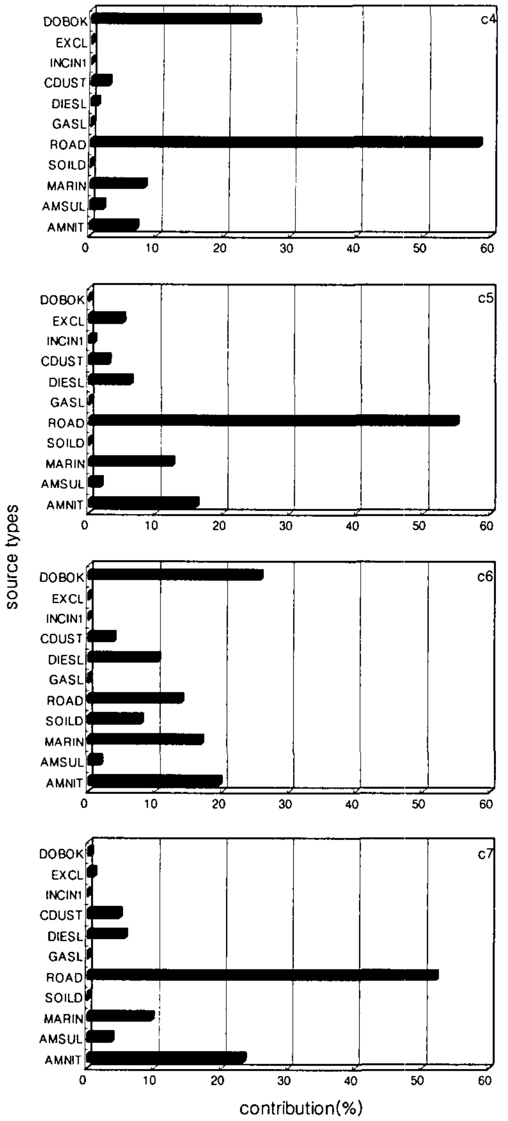

Fig. 3. Estimated source contributions to the CP (2.1<dp<10㎛) mass concentration in Jeju city.

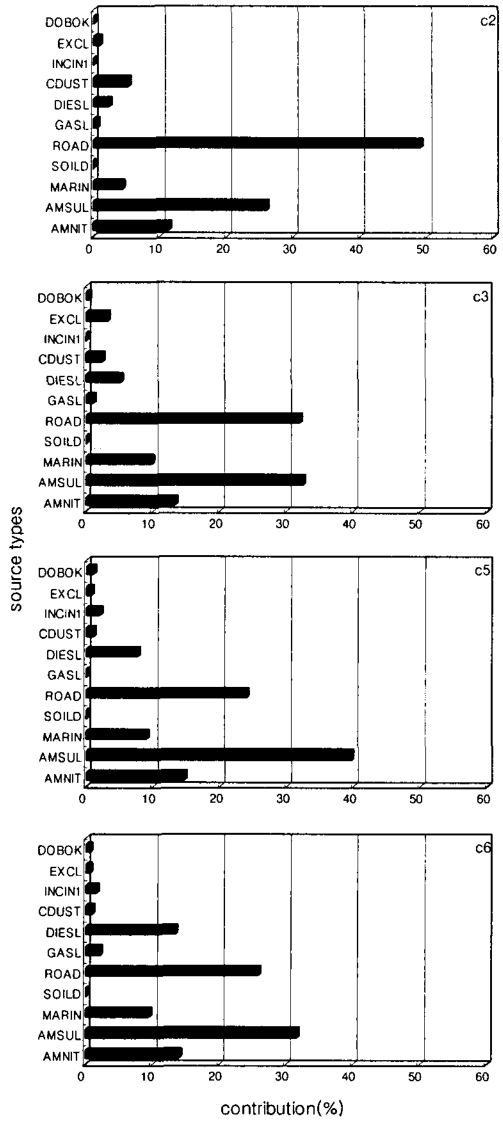

Fig. 4. Estimated source contributions to the FP (dp<2.1㎛) mass concentration in Jeju city.

Fig. 5 shows estimated average source contributions to PM10 in Jeju city. The average contributions were obtained to average arithmatically the contribution data for each sample, given in Fig. 3 and Fig. 4. For the CP, 45% is contributed by road dust, 16% is contributed from ammonium nitrate known as secondary aerosol, 13% from domestic boiler, and 12% is contributed by marine aerosols. For the FP, the secondary aerosol (ammonium sulfate and ammonium nitrate) is the most significant contributor with 45% contribution, and the next most significant contributor is a road dust, with 34% contribution. And diesel vehicle contribute with an average of 7% to the FP mass concentration. However, the contributions of the other sources (gasoline vehicle, domestic boiler, municipal incinerator, and cement dust, etc.) to the FP mass are not considerable.

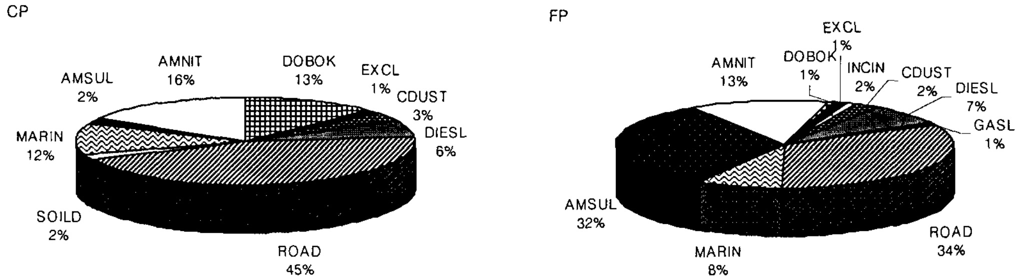

Fig. 5. Estimated average source contributions to PM10 mass concentration in Jeju city. (CP : 2.1<dp<10㎛, FP : dp<2.1㎛)

4. Conclusions

The mass concentrations and the chemical compositions of the aerosol samples, collected in Jeju city from May to September 2002, were determined, and then the source apportionment of those were performed using CMB receptor model.

The average PM10 mass concentration in ambient air was 28.80㎍/㎥ (24.06㎍/㎥~33.49 ㎍/㎥), and the fine particle (FP, $d_p < 2.1$㎛) fraction in PM10 was about 8% higher than the coarse particle (CP, 2.1㎛ $< d_p <$ 10㎛) fraction in PM10. The composition of the coarse particles was apparently different from the fine particle composition in PM10. The most abundant water soluble species is nitrate ion in the FP, on the other hand that is sulfate ion in the CP. And sulfur is the most dominant element in the FP, however, sodium is that in the CP. Also the elements originated from natural sources are richer in the CP, on the contary those from anthropogenic sources are richer in the FP.

As the results of CMB performance, it was found that road dust was the largest contributor to the CP mass concentration (45% of the CP), and also ammonium nitrate, domestic boiler and marine aerosol were major sources to the CP mass. However, the secondary aerosol was the most significant contributor to the FP mass concentration (45% of the FP). External combustion, municipal incinerator, gasoline vehicle, and soil dust were estimated the minor sources to both the CP and the FP mass concentration. But, it is suspected that the soil dust and the gasoline vehicle are estimated minor sources because of the collinearity with road dust and diesel vehicle, respectively.

Acknowledgements

This work was supported by a grant from the Academic Research Fund of the Cheju National University Development Foundation.

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