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Chemical Characteristics of Atmospheric Aerosol Particles on a High Mountain and an Urban Area in Sapporo, Japan

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Abstract

To determine characteristics of the source of air pollution in relation to the pressure patterns as well as the difference in total amount of the atmospheric aerosol particles by wind direction classified four quadrants, samplings of aerosol particles at the top of Mt.Teine (1,024 m a.s.l.) and on the campus of Hokkaido University in Sapporo City, Hokkaido, Japan were carried out. They were performed under two pressure patterns of Bai-u front pressure (June 30 - July 5, 1987) and Pacific Ocean high pressure (August 10 - 17, 1987) in summer period. To accomplish them, the levels of Carbon, SO_4^{2-}, NO_3^-, NH_4^+ and Cl^-, and metal element concentrations of Al, Na, Cu, Ni, K, Zn, Ca, Mn, Mg and Cd were measured. In the urban site, Hokkaido University, the total concentration showed relatively high values under westerly winds, and especially, the concentrations of marine aerosols and of soil particles were higher than those of anthropogenic aerosols.

In addition to these analyses, the aerosol particles collected on millipore filter papers using a continuous aerosol sampler were analyzed by a scanning electron microscope (SEM) and an energy dispersive X-ray microanalyzer (EMAX) during January and August, 1987. Soil particles which were rich in Al, Si etc. showed the highest frequency (%) in the aerosol particles at both sites. The frequency of the element Na was higher under the winter monsoon pressure pattern characterized by northwesterly winds as compared with other pressure patterns in the urban site. Especially, the element Fe rich in studded tire metal particles from vehicles was outstanding at the end of March. The component of S based on a comparison between Si : Na : S ratios was relatively higher in the urban site as compared with the mountain site under a travelling high pressure pattern in which the highest particle concentrations were recorded in the winter period. The anthropogenic aerosol particles observed under a travelling high pressure pattern may have a remarkable influence on the air pollution in Sapporo City, Hokkaido, Japan.

Key words: Chemical characteristics, Atmospheric aerosol particles, Wind direction, Pressure patterns, Anthropogenic aerosol particles.

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1. Introduction

Aerosol particles exist as organic or inorganic compositions, and as liquid or solid physical states in the atmosphere. They are multicomponent particles of small sizes, in general, ranging roughly from 0.01 to 10 μm in diameter (Pilinis et al., 1987) and have some effects on environment and human health as well as the life of plants or animals. These effects will be changed according to the meteorological and environmental situations (Stoker and Spencer, 1972). Especially, the phenomena of air pollution are seen more frequently in urban areas consisting of large population and buildings as compared to rural areas (Warker and Warner, 1976; Khemani et al., 1987). Therefore, it is important to survey the variation of aerosol concentrations with respect to the meteorological parameters for the prediction of air pollution and estimation of air quality level in urban areas.

To understand the air quality more practically in urban areas, it is important to know the variation of aerosol concentrations by the wind systems among meteorological parameters, since the concentrations of aerosol particles may change with the wind direction which is governed by the atmospheric pressure patterns.

There have been a number of studies aimed at identifying both the sources of pollutants and the contribution of chemical compositions in urban areas (Hammerle and Pierson, 1975; Usero and Gracia, 1986; Gregory et al., 1987). Furthermore, the studies on the comparison of aerosol concentrations for each wind direction and pressure patterns in the winter season have been carried out up to date (Lee et al., 1987, 1988a,b). However, no detailed study of the chemical characteristics of aerosol particles during the summer season with special consideration to the atmospheric pressure patterns has been made till the present. The purpose of this study is, therefore, to understand not only the representative sources of aerosol particles but also the characteristics and comparison of their concentrations for each wind direction according to the atmospheric pressure patterns between the ground surface (urban site) and mountain area (mountain site). In addition, analyses using SEM and EMAX of aerosol particles in each pressure pattern were described. Using these results, furthermore, it was found that the wind system has a significant effect on the air quality level in Sapporo City, Hokkaido, Japan.

2. Materials and Methods

2-1. Observation methods

Sapporo City is located on the Ishikari Plain of Hokkaido Prefecture, the northernmost island of Japan. This city has the largest urban area (1,118.01 km^2) in Hokkaido with a population of about 1.6 million. The characteristics of air pollution in this city is a typical urban style characterized by burning of fossil fuel for heating in winter season, the effluents of vehicles and so on. Specially, a serious social problem has been the air pollution by "Sha -fun" (literally 'automobile - dust') arising from studded tires of vehicles.

A vertical aerosol concentration has a great difference in the air quality in Sapporo City, because of the influence on the surface inversion layer and the variation of wind system in the boundary layer at approximately 400 - 500 m a.s.l. (Magono and Endoh, 1979;

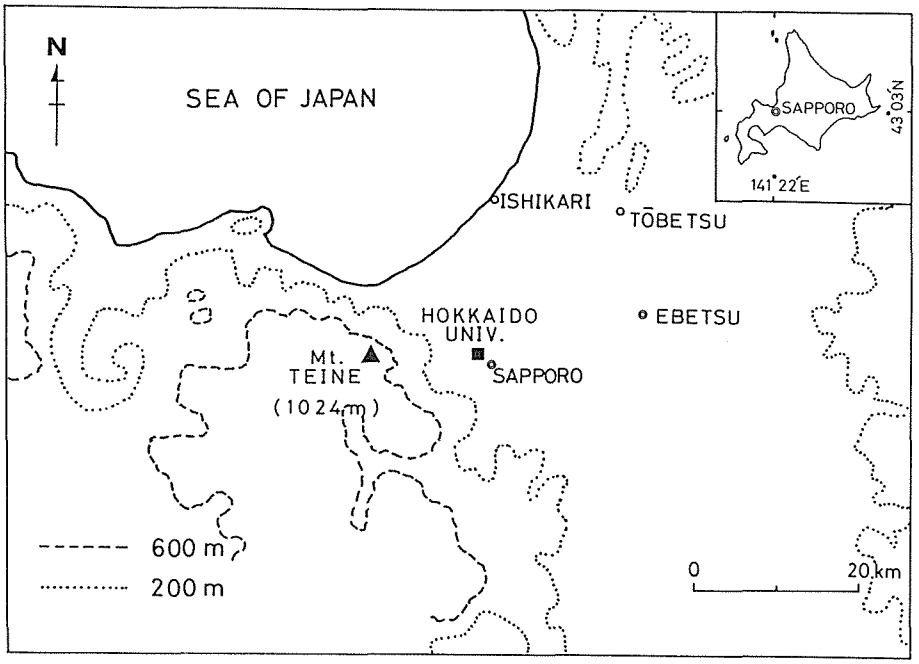


Figure 1. Location of Sapporo and sampling sites.

Endoh et al., 1981). Thus, the observation of aerosol particles was performed at the following two sites, namely the campus of Hokkaido University (15 m a.s.l.) (urban site) in Sapporo City and the Cloud Physics Observatory of Hokkaido University at the top of Mt. Teine (1,024 m a.s.l.) (mountain site) located above the polluted atmospheric layer on Sapporo (Kikuchi, 1984). The locations of Sapporo City and sampling sites are shown by a double circle, solid square and triangle in Figure 1. As shown in this figure, Mt.Teine is 10 km apart from the city center to the west. The Japan Sea lies from the sampling sites to the north.

A short-term sampling of aerosol particles using filter papers was carried out from June 30 to August 17, 1987 to determine the difference of aerosol concentrations for each wind direction in relation to the atmospheric pressure patterns. Furthermore, during January 24 to March 30, collections of aerosol particles were also done to investigate the chemical compositions of aerosol particles at both sites.

The apparatus used in this study were an aerovane and a selection circuit unit assembly for four quadrants of wind directions and calm condition. Detailed content regarding the aerovane and the wind controlled air sampler were described in previous papers (Lee et al., 1987, 1988a,b). Additionally, to collect automatically the atmospheric aerosol particles at intervals of 2 hours, a millipore filter paper of 90 mm in diameter was mounted on a small turn table of a new continuous aerosol sampler developed by Taniguchi and Kikuchi (1989). In this sampling, a total sampling air of approximately 120 liters containing the aerosol particles was collected on the individual patches of the filter papers.

2-2. Sample preparation and analytical method

Table 1 indicates the sample preparation procedures for chemical analyses. After sampling, each teflon filter was divided into two halves precisely, to measure the SO_4^{2-}, NO_3^-, NH_4^+ and Cl^- and 10 metal elements of Al, Na, Cu, Ni, K, Zn, Ca, Mn, Mg and Cd contained in the aerosol particles. Several discs of 1 cm in diameter were cut from a quartz fiber filter for carbon analyses. Ninety mm millipore filters were cut to a size of 11 mm in diameter to analyze the chemical compositions of aerosol particles by SEM and EMAX analyzer, respectively. Analytical methods used for individual chemical components were described in detail in a previous paper (Lee et al., 1987). A more detailed content regarding the sampling mechanism and method in a new continuous aerosol sampler was explained by Taniguchi and Kikuchi (1989).

Table 1. Sample preparation procedures for chemical analyses.

Sample collection	Filters (0.5μm)	Sample preparation
Aerosol particles	Teflon filter (47mm)	(half): Ionic components (NH_4^+, NO_3^-, $C\ell^-$, SO_4^{2-}) (half): Metal elements (Al, Ca, Cd, Cu, K, Mg, Mn, Na, Ni, Zn)
	Quartz fiber filter (47mm)	(d: 10mm): Total carbon (T.C.) (d: 10mm): Elemental carbon (E.C.) (T.C.−E.C.): Organic carbon (O.C.)
	Millipore filter (90mm)	(d: 11mm): Shape, Size and Components

2-3. Meteorological conditions

The total mean values of each meteorological parameter, such as temperature, wind speed and precipitation, measured during the summer season at both sites are shown in Table 2. The average values of wind speed for each wind direction in the summer season are described in Table 3. A Bai-u front (June 30 - July 5) was stationary at the south of Honshu Island and heavy rainy weather continued during the earlier part of July 1987. In Hokkaido, a trough was formed throughout this period. On the other hand, during the middle 8 days of August 1987, the weather situation indicated a Pacific Ocean high pressure pattern (August 10 - 17) over Honshu Island and a low pressure passed over the north of Hokkaido Island.

Table 2. Mean values of each meteorological parameter at both sites during two sampling periods in summer.

Meteor. parameter	Temperature (°C)		Wind speed (m/s)		Precipitation (mm)	
Site / Date	H.U.	M.T.	H.U.	M.T.	H.U.	M.T.
June 30–July 5	19.6	12.6	2.3	4.5	31.0	24.0
August 10–17	21.6	15.3	1.6	3.0	3.0	7.0

H.U.=Hokkaido University, M.T.=Mt. Teine

Table 3. Data of averaged wind speed (m/s) at each wind direction in the summer season.

Date (1987)	N		E		S		W	
	H.U.	M.T.	H.U.	M.T.	H.U.	M.T.	H.U.	M.T.
June 30–July 5	5.0	2.2	3.1	4.3	4.0	5.1	2.8	3.9
August 10–17	4.6	2.4	4.6	3.0	6.5	3.4	3.4	2.2

H.U.=Hokkaido University, M.T.=Mt. Teine

3. Results and Discussion

Figure 2 shows the surface weather map on July 3, 1987 as a representative pattern of the sampling period (June 30 - July 5) which mainly showed the pressure - trough pattern in Hokkaido and stationary phenomenon of Bai-u front with heavy rainfall in Honshu Island. During this period, the cumulative time (hrs) and concentration rate (%) of total aerosol concentration for each wind direction at both sites are shown in Figure 3 and Table 4. As shown in Figure 3, southerly winds prevailed in the urban site, Hokkaido University as compared with westerly winds in the mountain site, Mt. Teine. On the other hand, the concentration rate by wind direction indicated high values in westerly winds and calm (C) at the urban site, and calm and easterly winds at the mountain site. Therefore, specific wind directions, S and W at the urban site, and W and E at the mountain site, which had a long cumulative time and a high concentration rate, respectively, at both sites were selected to investigate the representative characteristics of air quality during this observation period. Figure 4 shows the concentration ($\mu g/m^3$) of each chemical composition in the southerly and westerly winds at the urban site, and westerly and easterly winds at the mountain site. In general, aerosol concentrations at each wind direction in relation to the pressure patterns in summer showed smaller values than those that were determined in winter, 1987 (Lee et al., 1988b). There was high concentration in westerly winds at the urban site. Especially, the concentrations of marine aerosols (Na and Mg) such as sea-salt particles and soil particles (Al and Zn) were high. However, in the mountain site, the absolute values of aerosol concentration were less than those at the urban site. Their vertical difference for each wind direction in summer was less than that in winter, 1987 (Lee et al., 1988b). Marine aerosols (Na, Cl, Mg etc.) in westerly winds at the mountain site showed small values. This proves that large aerosols such as sea - salt particles were not

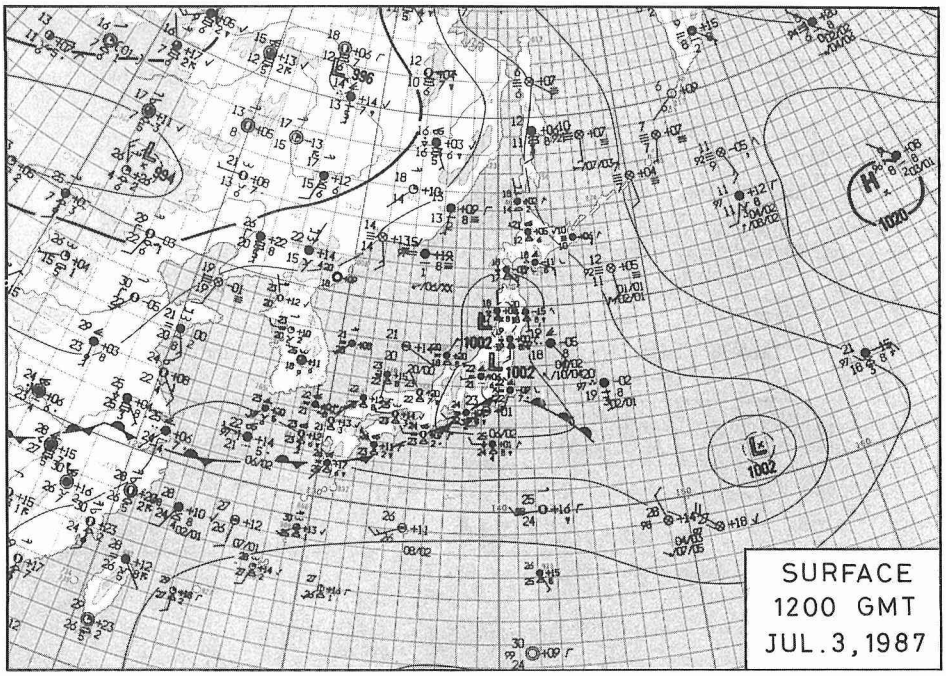


Figure 2. Surface weather map on July 3, 1987 (After Japan Meteorological Agency).

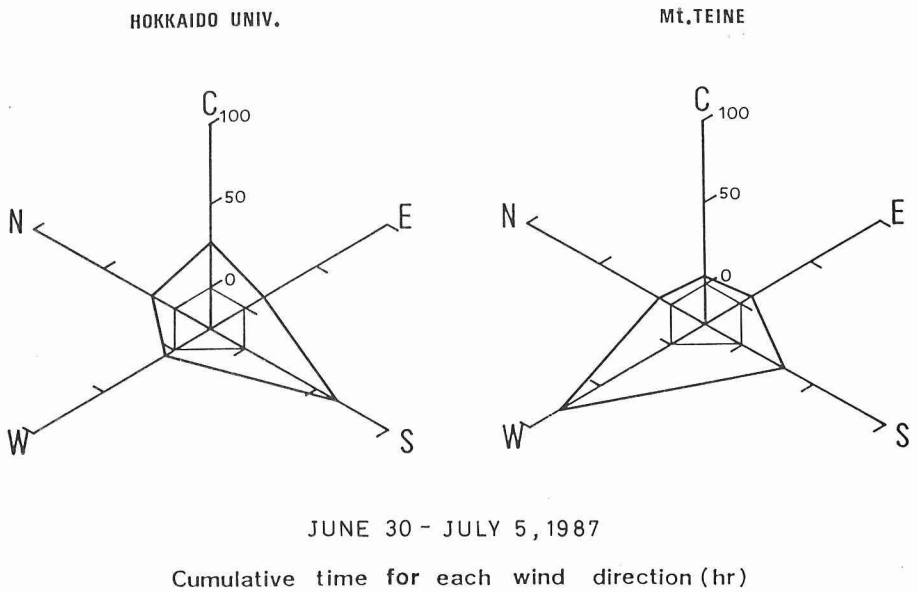


Figure 3. Cumulative time (hrs) for each wind direction at both sites on June 30 - July 5, 1987.

transported easily to the summit of mountain owing to their own weights.

The surface weather map in August 15, 1987 which was a representative day of Pacific Ocean high pressure pattern at the sampling period (August 10 - 17) was shown in Figure 5. Cumulative time (hrs) and concentration rate (%) of total aerosol concentrations for each wind direction at both sites in August 10 - 17, 1987 are shown in Figure 6 and Table 5. Although the southerly winds and calm condition at the urban site and westerly winds at the mountain site prevailed, the total aerosol concentration was high in the westerly winds at the urban site and calm and easterly winds at the mountain site. Hence, the concentrations of each aerosol particle for the southerly and westerly winds at the urban site and the westerly and easterly winds at the mountain site were analyzed. The results of each aerosol particle at these wind directions are shown in Figure 7. In the urban site, the concentration values of carbons (Total carbon, Elemental carbon and Organic carbon) and Al were higher than other elements in southerly winds. However, in westerly winds, Na, Mg and K concentrations generally originated from marine aerosols and Al and Zn concentrations originated from soil particles were high in the urban site, Hokkaido University. At the mountain site, carbons and soil particles for both westerly winds and easterly winds had slightly higher concentrations than other elements. As shown in this figure, the total aerosol concentration obtained during the sampling period in August was higher than that in July 1987. This may be explained by the fact that a lesser scavenging effect was shown in August in contrast to in July, since the precipitation amounts (31 and 24 mm) during June 30 - July 5 at both sites were higher than in August as shown in Table 2 (Engelmann, 1971, 1988; Murakami et al., 1983; Schumann et al., 1988).

In totality, 40 or 50 aerosol particles which were collected with a continuous aerosol sampler were analyzed using a SEM and an EMAX systems at both sites. Table 6 shows the percentage frequencies of the number of the chemical components containing in each aerosol particle at the urban site, Hokkaido University during each observation period, 1987. Numbers (No.) written on the right hand side of the table indicate the number of analyzed aerosol particles. All the numbers of each element shown in the individual spectral peaks were considered in the percentage measurement. As seen in this table, the elements of Si and Al which are typical components of soil particles were predominant in all kinds of aerosol particles analyzed. In January, the percentage frequencies of marine aerosols (Na and Cl) showed the highest values among those of other observation periods at the urban site. This result is well proved as the effects of strong northwesterly winter monsoon from the Japan Sea. The element of Fe had a high value in March especially. It is considered that studded tires of vehicles were mainly used in the urban areas during this period. The effect of street dusts by studded tires has given rise to a serious social problem in Sapporo City. Figure 8 shows a representative example of SEM - EMAX analyses obtained from aerosol particles, which were collected in March 28, 1987 at the urban site. It was observed that the shape of individual particles containing the element of Fe was a hard type around its surface in contrast to other elements which were obtained by Lee et al.(1988a,b).

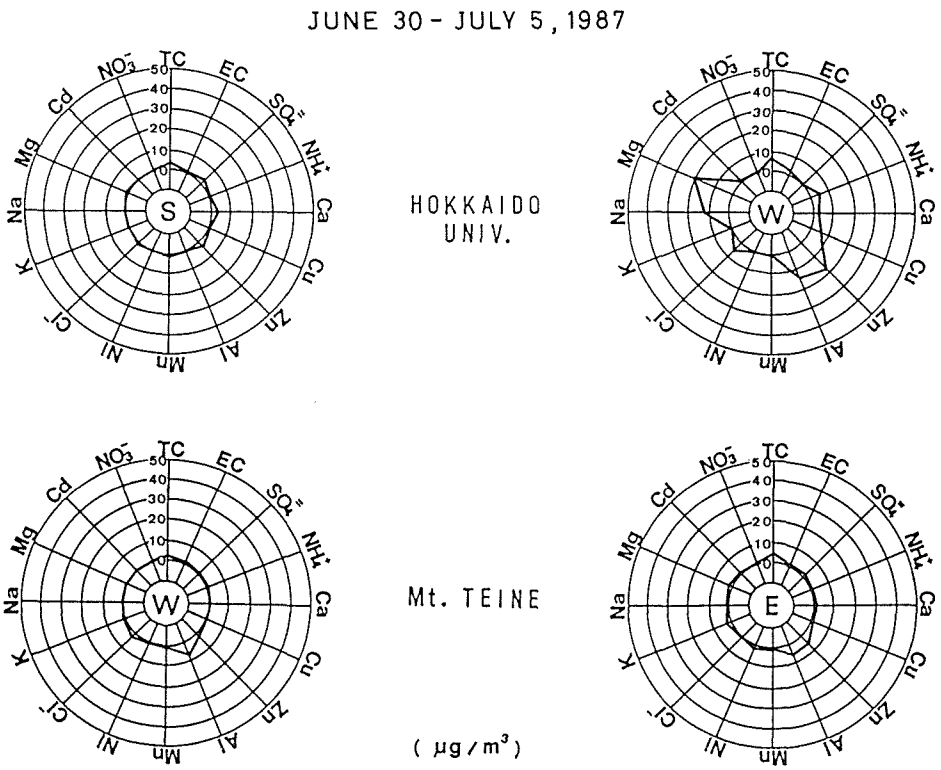


Figure 4. Total mean values of each chemical composition in the southerly and westerly winds at the urban site and the westerly and easterly winds at the mountain site (June 30 - July 5, 1987).

Table 4. Concentration rate (%) for each wind direction at both sites in June 30–July 5, 1987.

WIND DIRECT.	HOKKAIDO UNIV.	Mt. TEINE
N	9.2	19.4
E	23.2	22.7
S	6.7	7.7
W	37.4	11.6
C	23.6	38.6

Table 5. Same as Table 4 but for August 10–17, 1987.

WIND DIRECT.	HOKKAIDO UNIV.	Mt. TEINE
N	5.9	20.6
E	5.3	22.6
S	21.5	18.6
W	63.5	13.3
C	3.8	24.9

Subsequently, the frequencies of elements contained in aerosol particles at the mountain site, Mt. Teine, are shown in Table 7 during each observation period, 1987. Generally, the percentage frequencies of Si and Al (soil particles) for each pressure pattern showed higher values than the others. This result has a close similarity to that obtained at the urban site as shown in Table 6. However, the percentages of the element of Na showed low values at the mountain site as compared with those at the urban site. This may be

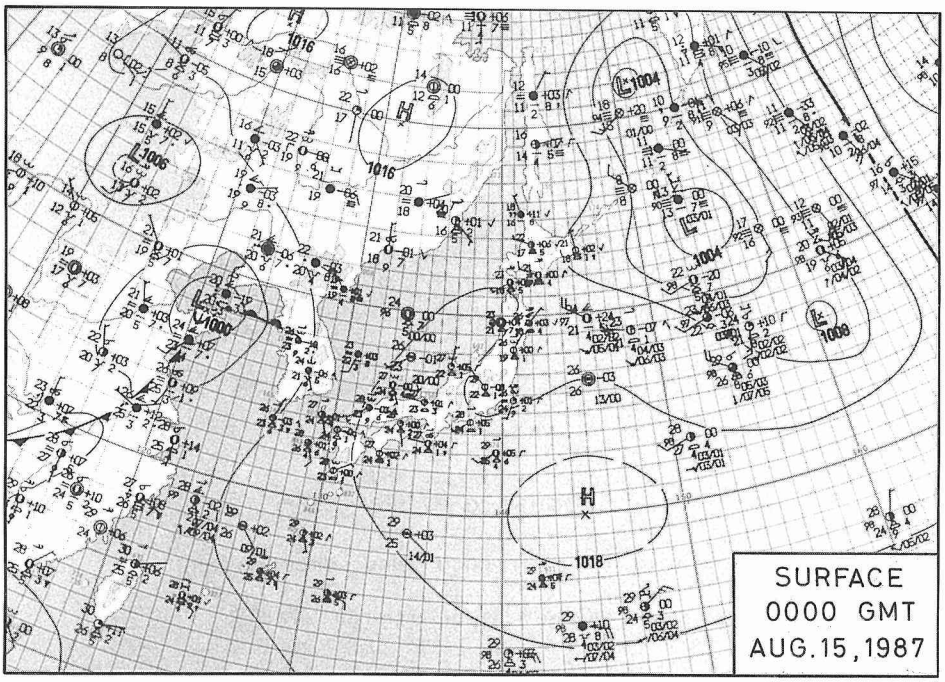


Figure 5. Same as Figure 2 but for August 15, 1987 (After J.M.A.).

explained by the fact that southerly and westerly winds prevailed at the summit of Mt. Teine during the sampling period, January 27, 1987. It can be also proved that large particles such as Na are too heavy to be transported from the sea surface to the high altitude because of their own weight as described in previous papers (Lee et al., 1988 a,b). Especially, on the mountain site on March 28, 1987, the percentage frequencies of the elements of Fe and Ca which are the main components of street dusts from studded tires (Mohri et al., 1982) have negligible values as compared to those in the urban site as a result of atmospheric stability effect often brought about by travelling high pressure.

The frequency distributions of elements contained in each aerosol particle were as same as described before. However, they indicated the percentage in number of particles containing a certain element, but not the mass percentage of the element. The consideration regarding the frequency distribution of elements alone contained in aerosol particles is not sufficient to identify the components of the aerosol particles or to determine their origins. In order to investigate the origin of aerosol particles, the relative weight fraction of three elements presented in each particle was considered. Especially, in this study, the elements of Si, Na and S were selected as standards for soil particles, sea-salt particles and sulfate particles as combustion products originating from human activity, respectively. Weight concentration ratio could be obtained by measuring the X-ray intensities of the elements (Si, Na and S), since a good correlation between X-ray intensity ratio and weight concentration ratio was obtained, as a result of an experimental study using an ultra -

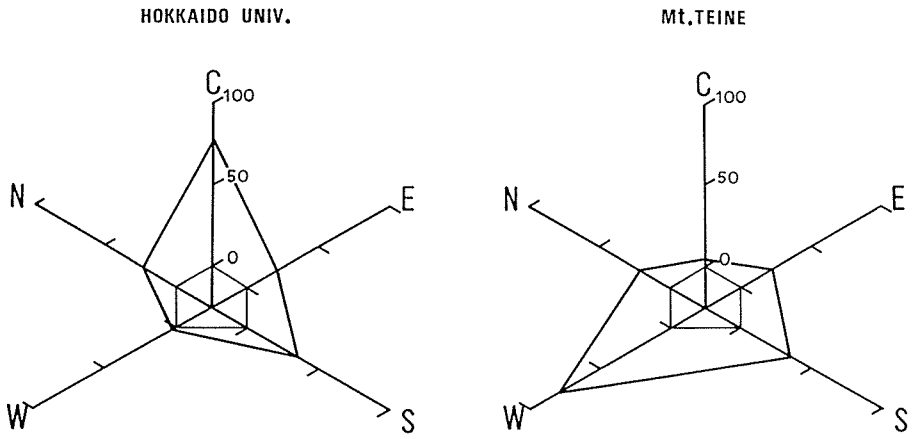


Figure 6. Same as Figure 3 but for August 10 - 17, 1987.

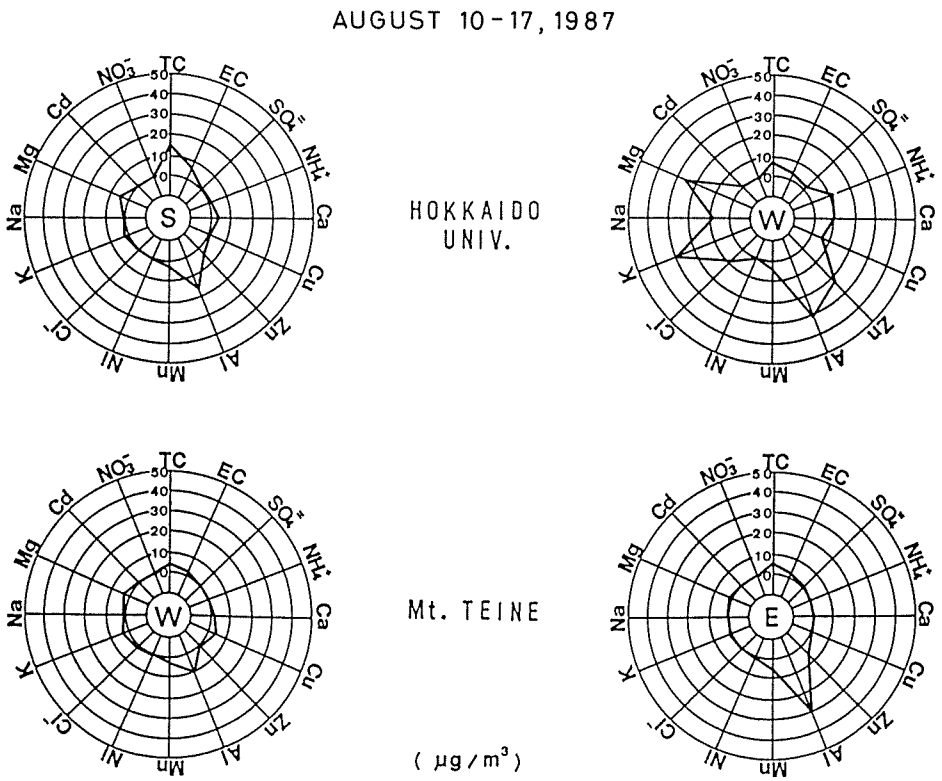
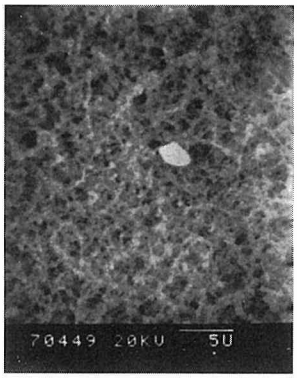


Figure 7. Same as Figure 4 but for August 10 - 17, 1987.

Table 6. Frequency (%) of elements contained in the aerosol particles at the urban site, 1987.

Date (Pressure pattern)	Elements (%)															No.
	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	
Jan. 27 (Winter monsoon)	58	35	53	63	0	60	65	43	43	3	3	0	43	0	0	5	40
Feb. 24 (Japan Sea-low)	15	28	55	60	5	40	18	40	48	8	0	2	48	0	0	4	40
March 28 (Travelling high)	14	38	70	83	32	70	28	46	66	2	4	2	72	2	2	12	50
July 3 (Bai-u front)	15	18	68	78	3	20	18	38	45	3	0	0	43	2	0	10	40
Aug. 15 (Pacific high)	15	23	78	80	5	60	28	45	68	5	0	0	53	2	0	5	40



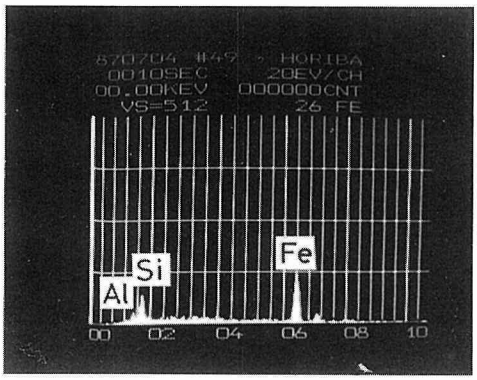


Figure 8. A typical example of aerosol particles observed on March 28, 1987 at the urban site.

marine blue reagent ($Na_xAl_xSi_{(12-x)}O_{24}Na_yS_z$) (Murakami and Kikuchi, 1982).

Figures 9, 10 and 11 show triangle diagrams of the Si : Na : S ratios of aerosol particles collected at both sites on January 27, February 24, and March 28, 1987, respectively. In these figures, solid circles and open circles indicate the relative weight fractions of Si, Na and S present in the aerosol particles for two hours from 10:00 to 12:00 (day time) or from 22:00 to 24:00 (night time), respectively. The relative weight fraction of Si for soil particles is predominant at both sites, although all of Hokkaido Island is mostly covered by a deep snow in winter. It may be expected that the aerosol particles rich in Si are soil particles originating from the other southern islands and the Asia continent. This fact was also reported by Murakami and Kikuchi (1982). This is well proved by mean wind direction (approximately westerly winds) in sampling time as shown in Table 8, which indicates data of the prevailing wind direction and wind speed (m/s) at both sites during the sampling

Table 7. Same as Table 6 but for the mountain site, 1987.

Date (Pressure pattern)	Elements (%) Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	Cr	Mn	Fe	Ni	Cu	Zn	No.
Jan. 27 (Winter monsoon)	13	5	50	73	8	53	55	50	63	5	8	0	30	3	3	5	40
Feb. 27 (Japan Sea-low)	5	20	78	83	5	33	13	40	43	8	0	0	33	0	0	3	40
March 28 (Travelling high)	8	13	60	83	8	38	20	40	40	8	0	0	30	2	0	15	40
July 3 (Bai-u front)	10	8	53	68	0	20	25	53	53	8	0	0	43	3	0	3	40
Aug. 15 (Pacific high)	8	13	70	80	8	33	13	40	50	8	3	0	50	2	3	5	40

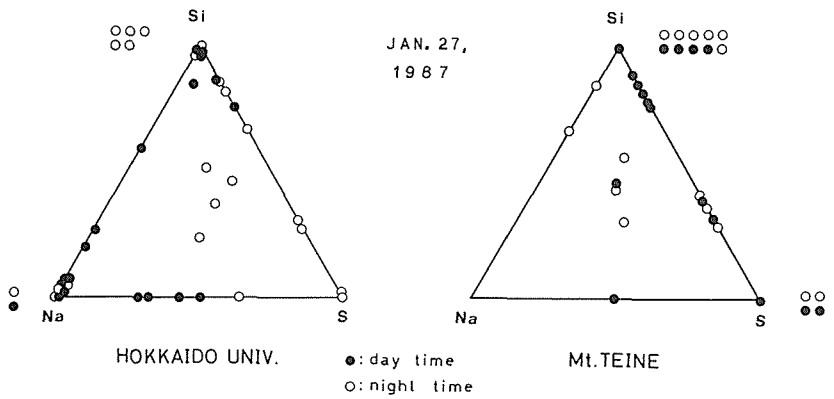


Figure 9. Triangle diagrams of the Si : Na : S ratios of aerosol particles collected at both sites on January 27, 1987.

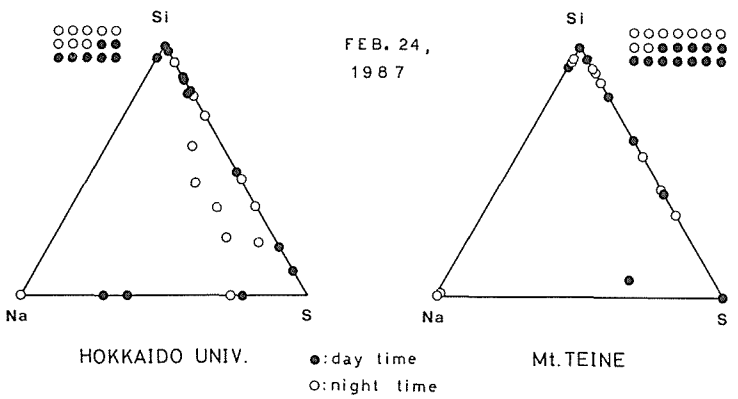


Figure 10. Same as Figure 9 but for February 24, 1987.

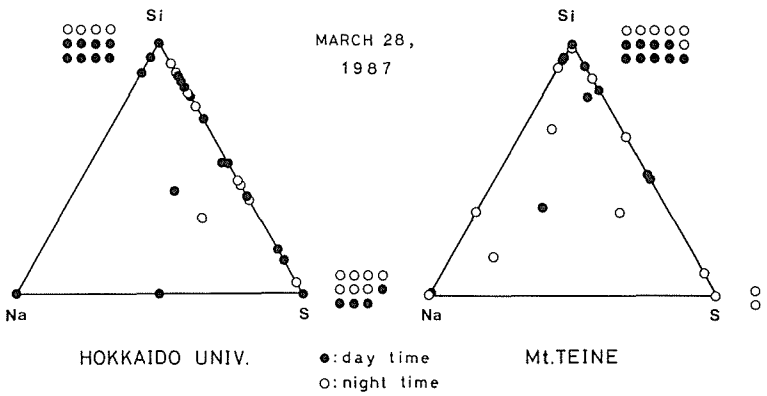


Figure 11. Same as Figure 9 but for March 28, 1987.

Table 8. Average wind direction and wind speed (m/s) at both sites for each sampling time, 1987.

Site	Hokkaido University				Mt. Teine			
Date	10 : 00−12 : 00		22 : 00−24 : 00		10 : 00−12 : 00		22 : 00−24 : 00	
Jan. 27	NW	(5.5)	C	(0.5)	SW	(8.2)	SW	(11.2)
Feb. 24	SW	(10.0)	NE	(2.2)	SW	(12.0)	E	(4.3)
March 28	S	(9.2)	S	(4.6)	NW	(16.0)	SW	(10.4)
July 3	E	(5.0)	S	(7.0)	SE	(2.4)	S	(9.2)
Aug. 15	E	(2.0)	E	(2.2)	SE	(3.0)	SE	(4.1)

time, 1987. For instance, in relation to wind systems, Na for sea-salt particles was rich in the urban site of January 27, 1987. It can be understood by the effect of northwesterly winter monsoons from the Japan Sea during the day time in the urban site. However, on March 28, 1987, the ratio of Si was larger than others in spite of southerly winds at the urban site. It can be explained that street dusts are generated by the use of studded tires in Sapporo during this period. Especially, the ratios of S for anthropogenic aerosols in the urban site were higher than on the mountain site on March 28, 1987. This fact may be explained by the observational facts that the surface temperature inversions frequently occur at travelling high pressure pattern in winter. It is well proven by observations of the vertical concentrations of aerosol particles using a tethered balloon in Sapporo City, and explained by the surface temperature inversions seen frequently (Ishioka et al., 1988).

In summer period (July 3 and August 15, 1987), triangle diagrams of the Si : Na : S ratios of aerosol particles collected at both sites are shown in Figures 12 and 13. As shown in these figures, generally, the weight fractions of Si element were higher than the others without any differences by pressure patterns in the summer season. A distinct difference of each Si : Na : S ratio was not clear, since only aerosol particles under easterly and southerly winds were mainly collected at both sites during the summer season. However, these results showed similar tendencies to those obtained in the previous paper during summer period (Lee et al., 1987).

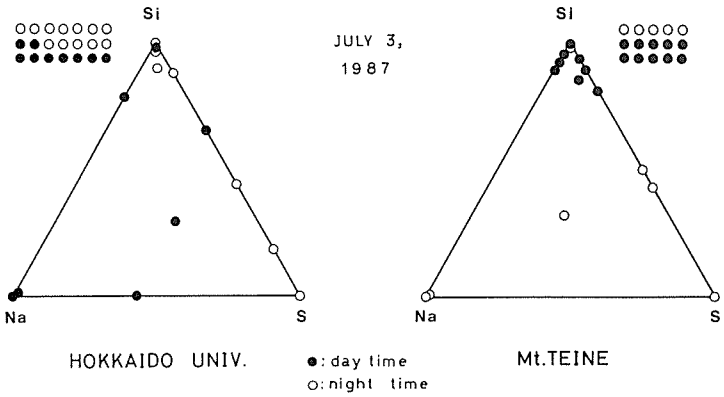


Figure 12. Same as Figure 9 but for July 3, 1987.

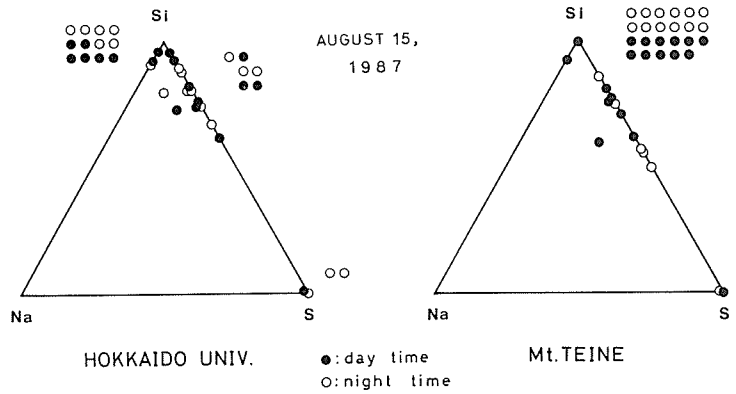


Figure 13. Same as Figure 9 but for August 15, 1987.

4. Conclusions

In order to study the variation of aerosol concentrations by wind systems, the aerosol samplings in relation to the atmospheric pressure patterns were carried out for each wind direction at the urban and mountain sites. In this study, short-term aerosol samplings in summer period were performed under two pressure patterns of Bai-u front pressure (June 30 - July 5, 1987) and Pacific Ocean high pressure (August 10 - 17, 1987). In general, aerosol concentrations for each wind direction in relation to the pressure pattern in summer showed lower values than those in winter. The concentrations of marine aerosols (Na and Mg) such as sea - salt particles and soil particles (Al and Zn) were high in westerly and southerly winds at the urban site. The scavenging effect by heavy rainfalls was remarkable in the period of Bai-u front pressure pattern as compared with Pacific Ocean high pressure pattern.

As the representative results of SEM -EMAX analyses under each pressure pattern,

the effects of strong northwesterly winds from the Japan Sea were appeared clearly under the winter monsoon pressure pattern with high percentage frequencies of marine aerosols (Na and Cl). The element of Fe showed a high value in March, especially. It was considered that studded tires of vehicles were the main cause in the urban area during this period. However, the percentages of Na and Fe showed lower values at the mountain site as compared with those at the urban site as a result of atmospheric stability effect often formed by travelling high pressure.

In the measurements of Si : Na : S ratio, generally, the element of Si for soil particles showed high values. The ratio of S for anthropogenic aerosols to other aerosols in the urban site was higher than in the mountain site on March 28, 1987. This fact may depend upon the fact that the surface temperature inversion and stable layer aloft are frequently observed under travelling high pressure patterns. Therefore, it is concluded that anthropogenic aerosol particles observed under high pressure patterns may have a remarkable influence on the formation of high pollution level of air quality in Sapporo, Japan.

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