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Chemical Compositions of Atmospheric Aerosol Particles in Relation to the Wind Direction in Winter Season, Sapporo, Japan

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Abstract

In the winter of 1986 and 1987, measurements of atmospheric aerosol particles in relation to the wind direction were carried out at the summit of Mt. Teine (1,024 m a.s.l.) and in the campus of Hokkaido University in Sapporo City, Hokkaido, Japan. To assess the characterization of the source of air pollution in relation to the wind direction, organic and elemental carbon, ionic components, and metal elements were sampled at both sites and analyzed at the Instrumental Analysis Center, Hokkaido University. Marine aerosols with sea-salt particles and soil particles showed a considerable difference in concentration by altitude and wind direction. These particles decreased with the amount of precipitation. The change of particle concentrations of anthropogenic aerosols by altitude was considerably higher in winter than their background level in summer season in 1986 (Lee et al., 1987).

Key Words: Chemical compositions of aerosol, Atmospheric aerosol particles, Wind direction, Characterization of the source of air pollution.

1. Introduction

Atmospheric aerosol particles arise from wind-blown dust, sea surface, refuse burning, smelters and a wide range of individual specific industrial sources. They have a great effect on the environment and human health, in relation to the meteorological situations in urban areas (EPA, 1973; Wark and Warner, 1976; Khemani et al., 1987). Therefore, it is important to survey the relation between meteorological parameters and the amount of aerosol concentrations (Sladkovic et al., 1986).

Recently, a number of studies have been made regarding the chemical compositions of airborne particles and aerosols in urban areas (Usero and Gracia, 1986; Sievering,1987). However, no theoretical or experimental investigations have been carried out to estimate the change of aerosol concentrations with respect to meteorological parameters, especially the wind direction.

The purpose of this paper is, therefore, to describe (1) the difference of aerosol concentrations by each wind direction and altitude, (2) comparison between winter values and summer background values (Lee et al., 1987), and (3) correlation between the anthropogenic aerosol concentrations and the altitude and wind direction.

2. Materials and Methods

2.1 *Sampling, preparation and analysis*

During a winter season, the air quality in an urban area such as Sapporo City has a great difference in atmospheric aerosol concentration, because of the existence of surface inversion layer and the variation of wind system in the boundary layer at approximately 400 m a. s. l. (Magono and Endoh, 1979; Endoh et al., 1981). From October 26, 1986 to January 24, 1987, the observation of aerosol particles was performed at following two sites: the campus of Hokkaido University (50 m a. s. l.) in Sapporo City and the Cloud Physics Observatory of Hokkaido University on the summit of Mt. Teine (1,024 m a. s. l.), which is located above the polluted atmospheric layer on Sapporo (Kikuchi, 1984).

The location of Sapporo City and sampling sites are shown in Figure 1. The sampling apparatuses of aerosol particles used in the present study consist of an aerovane (Environmental Pollution Control Center, Model WDP-100) and selection circuit unit assembly of each wind direction as shown in Figure 2. They are

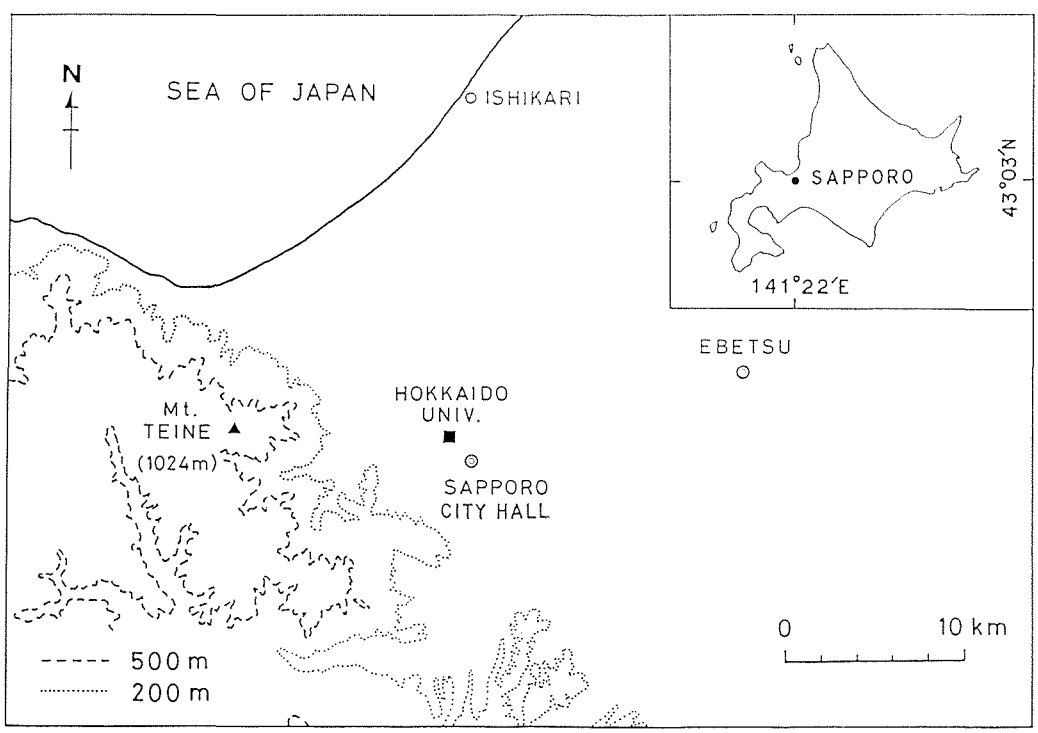


Figure 1. Location of Sapporo and sampling sites.

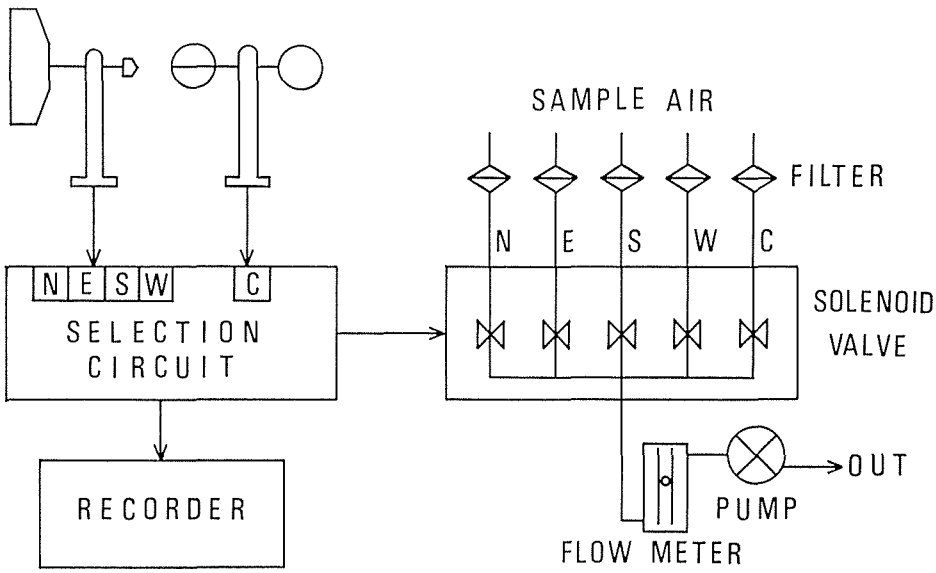


Figure 2. Sampling apparatus controlled by aerovane.

used in conjunction with a filter air sampler requiring an electrical solenoid valve to sample air when the wind direction is specific. The chemical contents of aerosol particles sampled and analyzed in this study in the winter period are the same as those in the summer period in 1986 by Lee et al. (1987); that is, organic and elemental carbon, ionic components (NH_4^+, NO_3^-, SO_4^{2-} and Cl^-) and metal elements (Al, Na, Cu, Ni, K, Zn, Ca, Mn, Mg and Cd). Details of the sampling procedure, preparation and filter species used with the wind controlled air sampler

Table 1. Analytical methods of individual items and instruments

Item	Analytical method	Instrument
T. C.	850°C, 1hr	NC-80 Carbon analyzer
E. C.	300°C, 30 min	(SUMIGRAPH)
NH_4^+	Indophenol method	
NO_3^-	Hydrazin reduction	UV-120-02
Cl^-	Mercury thiocyanate	Spectrophotometer
SO_4^{2-}	Barium perchlorate-Thorin	(SHIMADZU SFU)
Al	HNO_3	
Ca	+	
Cd	$HClO_4$	Flameless
Cu	+	Zeeman effect
K	HF	Atomic absorption
Mg	Dry	Spectrophotometer
Mn	+	(HITACHI 170-70)
Na	HCl	
Ni	Atomic absorption	
Zn	analysis	

are described in the previous paper (Lee et al., 1987).

Table 1 indicates analytical methods and instruments used for analyzing aerosol samples. They are the same as those adopted by Lee et al. (1987) for the summer period in 1986. Total and elemental carbon of aerosol samples are determined by NC Analyzer. Ionic components are determined by a UV Spectrophotometer at a specified wavelength range. Metal elements are analyzed by conventional atomic absorption methods using a Flameless Zeeman Effect Atomic Absorption Spectrophotometer. Analytical procedures and model of instruments used in these analyses are explained in more detail by Lee et al. (1987).

2. 2 *Meteorological conditions*

The results of measurements on the frequency of the monthly wind direction and the amount of precipitation during the winter period at both sites are shown in Figure 3. The prevailing wind direction in winter seasons at Sapporo City in general is westerly or northerly in the monsoon type (Kikuchi, 1984). However, as seen in the figure, the cumulative time of each wind direction did not show a remarkable difference at Hokkaido University. Especially, the calm condition and northerly wind prevailed in the observation period. On the other hand, at the summit of Mt. Teine, the westerly and southerly winds prevailed comparing with other wind directions. Especially, the calm condition and easterly wind appeared

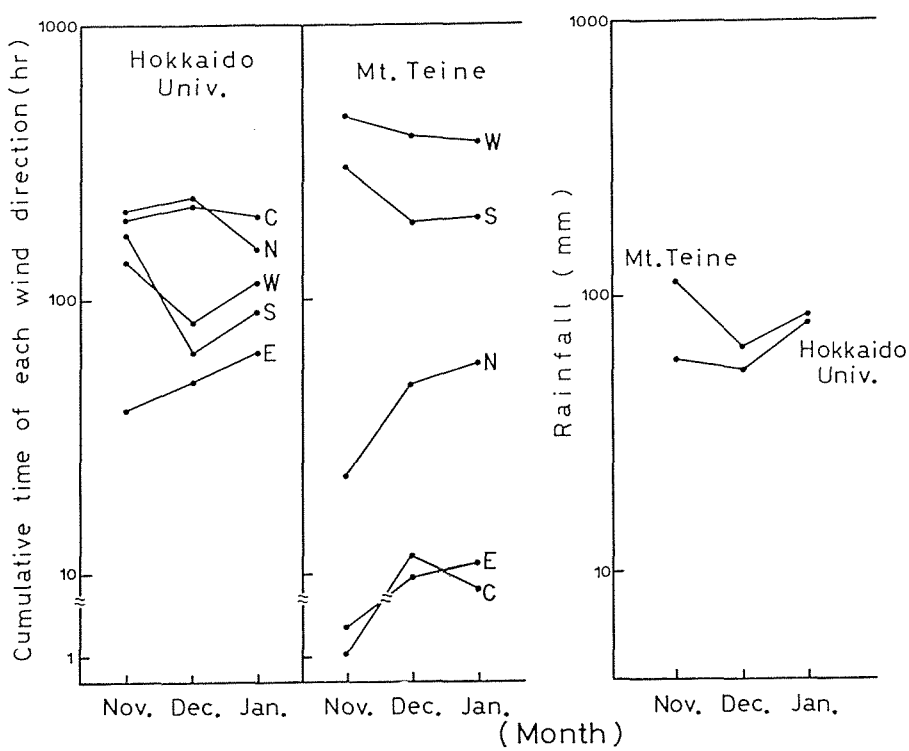


Figure 3. Frequency of monthly wind direction and precipitation amounts at both sampling sites.

only for several hours and ten hours. The precipitation amount was not so different, but that of November at Mt. Teine was more than that of Hokkaido University throughout the observation period.

3. Results and Discussion

The total mean values of chemical compositions by each wind direction during the winter period at both sites are shown in Figures 4 and 5. The arrangement of chemical components can be combined to form the representative origin source of three groups, namely, NH_4^+, Ca, Cu, Zn, Al, Mn and Ni from natural sources; Cl^-, K, Na and Mg from marine sources; and Cd, NO_3^-, TC, EC and SO_4^{2-} of anthropogenic sources, respectively (Ohta and Okita, 1983; Lee et al., 1987).

In the lowland site, Hokkaido University, the anthropogenic and marine aerosol (Carbon, SO_4^{2-}, Na, Mg etc.) concentrations are higher than others in all wind directions as shown in Figure 4. The total average concentrations of each chemical components, on the whole, increased in quantity as compared with data obtained during the summer period in urban area (Lee et al., 1987). In Figure 5, the aerosol concentrations at the mountain site generally show less than those in the lowland site without any difference by wind directions. Especially, the amount of soil aerosols in the mountain site during the winter period shows the least value. Therefore, it shows the largest difference of concentration against that in lowland area. This fact can be explained by a deep snow cover at the summit of Mt. Teine in winter. On the other hand, the lowland area was affected by suspended particles from the soil and street dust especially caused by studded tires of vehicles.

Figures 6 and 7 show the ratio of aerosol concentrations in the winter period against that in the summer period at both sites. The thick lines in concentric circles in both figures represent that the concentration ratio of winter/summer equals to 1. In winter as compared with summer, concentrations of anthropogenic and marine aerosols are higher in lowland areas, although natural and anthropogenic aerosols generally amount less in a westerly wind direction. These results can be explained by the appearance of pollutants in a high concentration from contaminant sources according to the sporadic westerly wind in summer period.

In the mountain area, the marine aerosol amount is higher than that in the summer period, since the winter monsoon mostly blows from the Japan Sea, that is, from westerly and northerly directions. Further, the concentration of anthropogenic aerosols on Mt. Teine is limited, compared with a high amount in lowland, Hokkaido University. This result indicates that the surface inversion layer and the stable layer of air temperature are frequently formed between the ground surface and the summit of Mt. Teine. The amount of crustal elements (Al, Zn etc.) and NH_4^+ which are originated in botanical and animal bacteria mainly show lesser values than in the summer period.

The concentration amounts ($\mu g/m^3$) of NH_4^+ and Al at both sites, mainly originated from natural and soil sources, are shown in Figure 8 by each wind direction and calm condition. They show a considerable difference in concentration

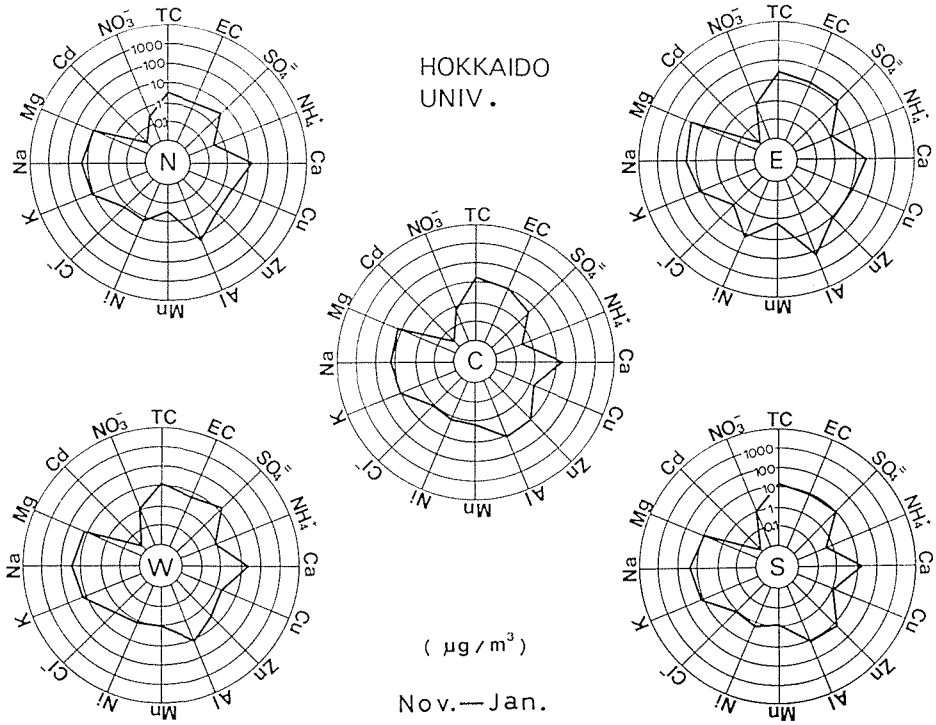


Figure 4. Total mean values of chemical compositions by each wind direction during the sampling period at the Hokkaido University.

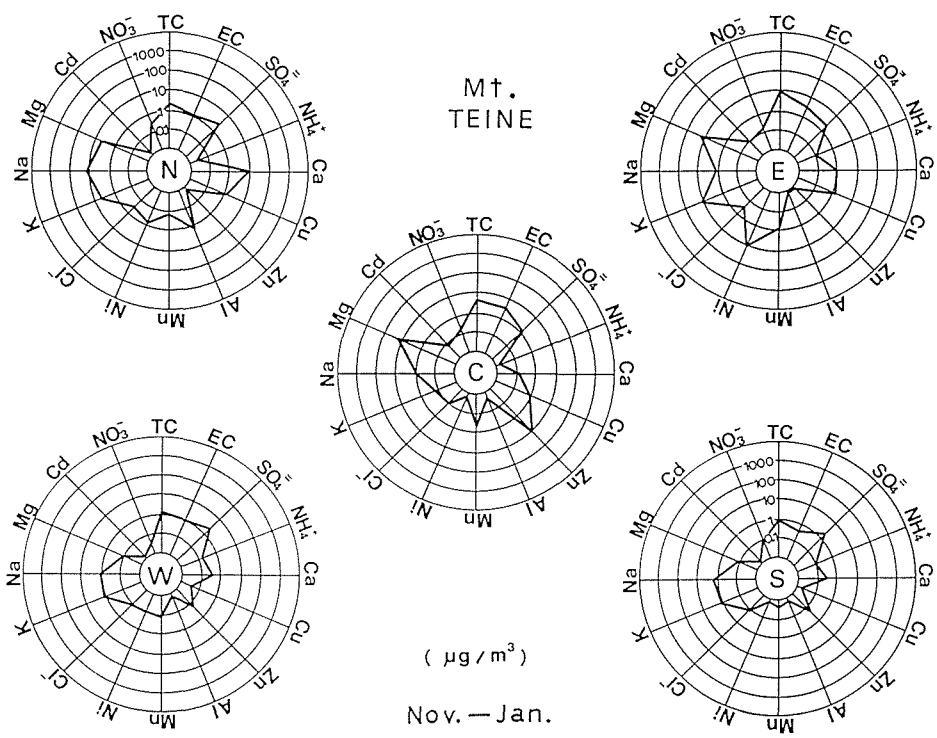


Figure 5. As for Fig. 4 but for the Mt. Teine.

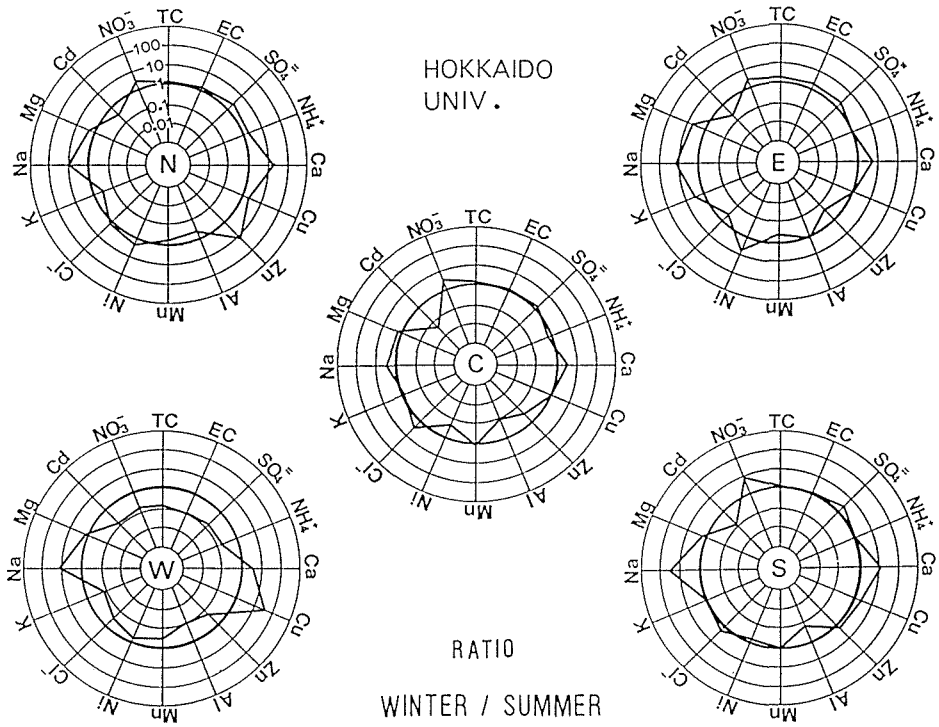


Figure 6. The ratio of aerosol concentrations in winter period to those in summer at the Hokkaido University.

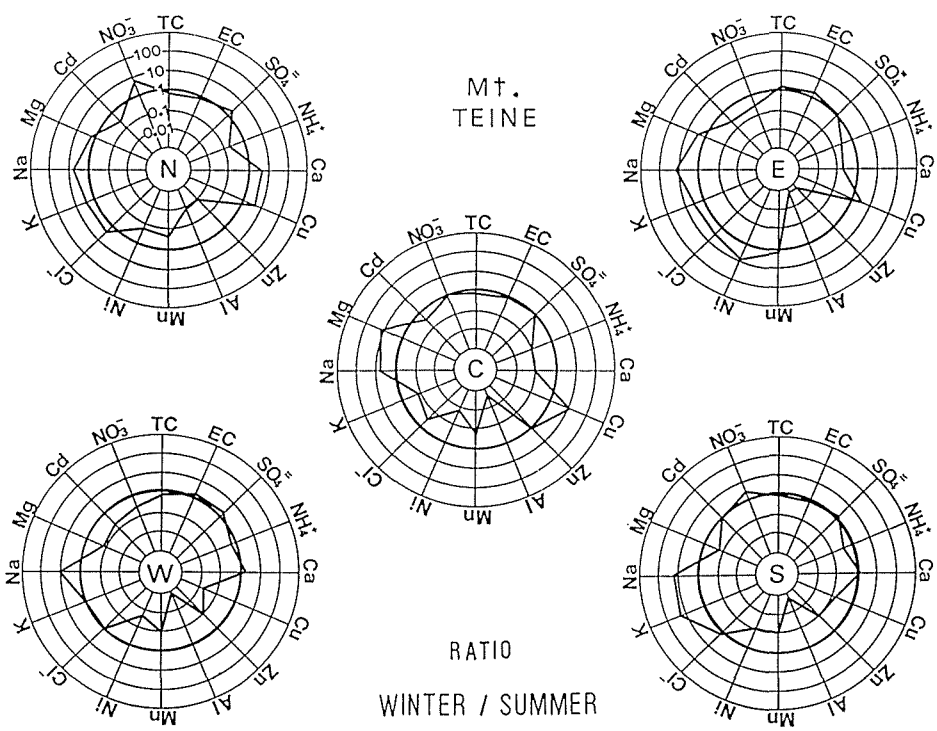


Figure 7. As for Fig. 6 but for the Mt. Teine.

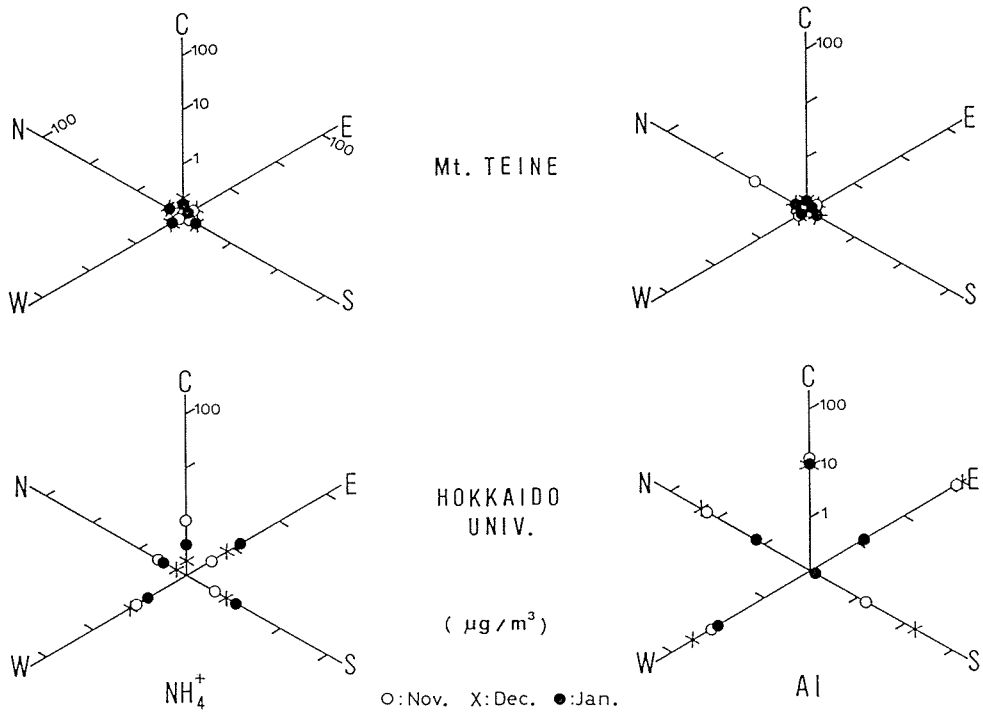


Figure 8. Vertical concentrations of NH_4^+ and Al by each wind direction.

by altitudes. This phenomenon can be explained by a scavenging effect of many times of snowfalls on the summit of Mt. Teine in winter (Murakami et al., 1983). Figure 9 indicates the concentration amounts of Na and Cl^-, mainly originated from marine sources. Generally, marine aerosol concentrations at the lowland site are higher than at Mt. Teine in all wind directions. This result is similar to that measured in the summer period by Lee et al. (1987). It is considered that large and giant particles such as sea-salt particles are difficult to be transported from the sea surface to the high altitude such as mountain top, owing to its own weight, and that they will be captured by surface obstacles such as trees and houses (Tanaka and Toba, 1969; Kikuchi and Yaura, 1970: Lee et al., 1987). However, even at the summit of mountains, the concentration of Na during the winter is higher than in the summer period. It proves the strength of winter monsoon winds.

The concentrations of total carbon and SO_4^{2-} which mainly are originated from anthropogenic sources have higher values in the lowland than at the summit of the mountain as shown in Figure 10. On the other hand, at both sites, differences of concentrations by wind directions have no great effect during the winter period, likewise. These results suggest that, in Sapporo City, the air pollution is much more caused by the combustion of gas or coal oil fuel which are used extensively throughout the whole urban area for heating in winter season than by local contaminant

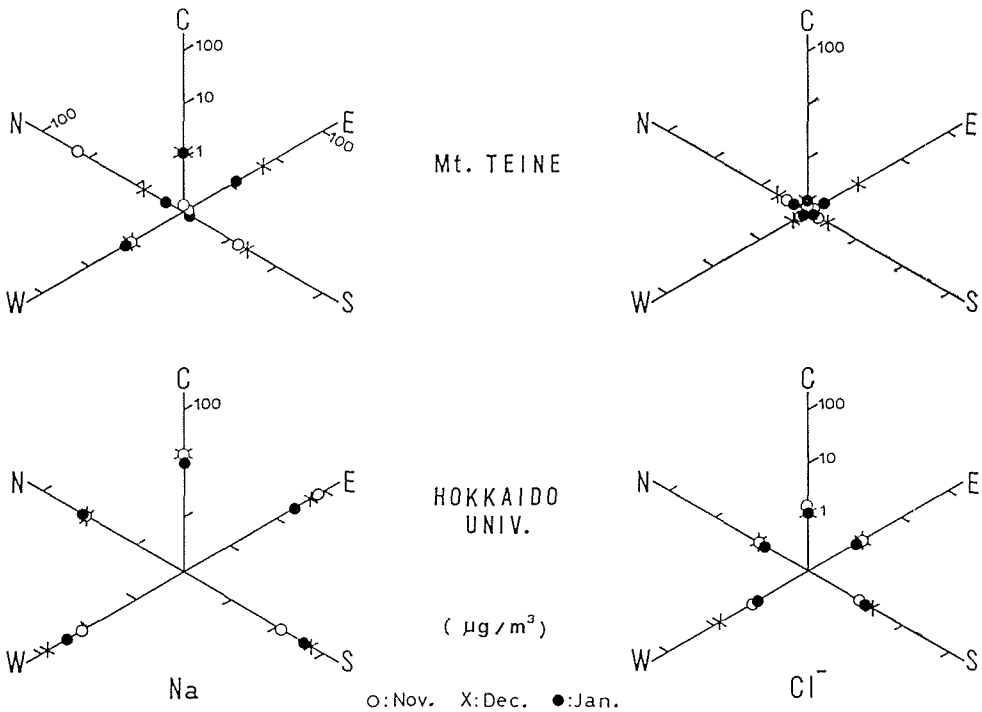


Figure 9. As for Fig. 8 but for Na and Cl^-.

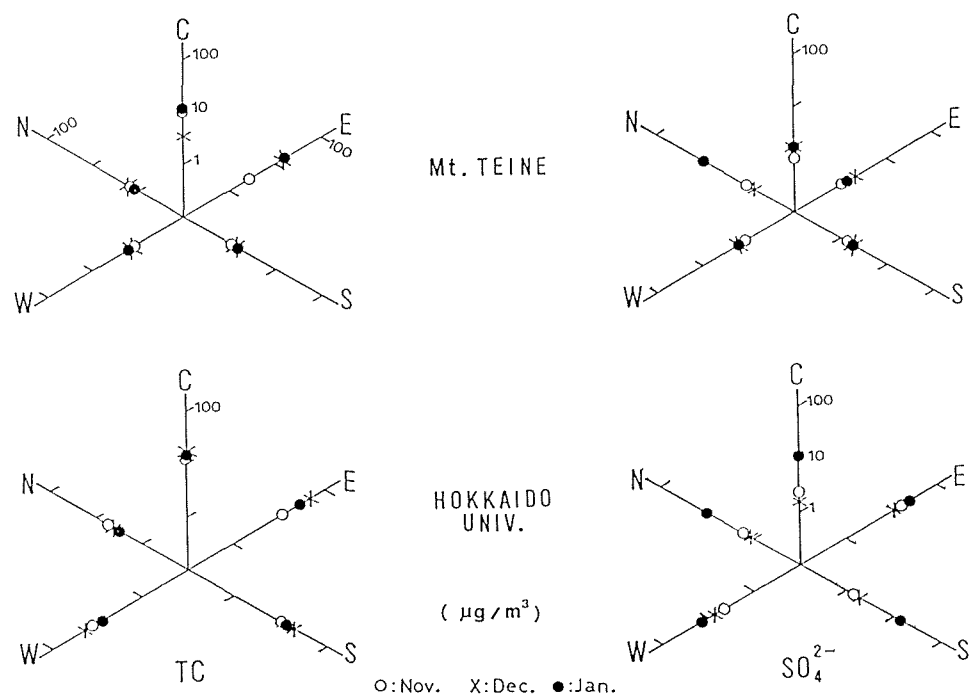
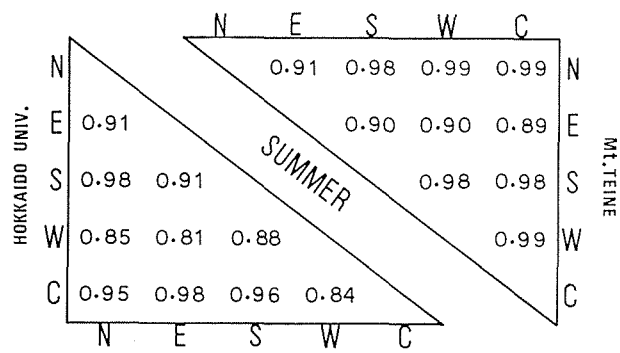


Figure 10. As for Fig. 8 but for Total Carbon and SO_4^{2-}.

sources.

Table 2 indicates the linear correlation coefficient (r) of anthropogenic aerosols (TC and SO_4^{2-}) by wind direction in summer and winter periods. In this calculation, each concentration of the TC and SO_4^{2-} measured nearly once a week is used as a statistical variable. In the summer, the variance of anthropogenic aerosol concentrations by wind direction is scarce at the summit of Mt. Teine. However, a correlation coefficient between southerly winds and easterly winds at Mt. Teine has a small value ($r=0.15$) in contrast to 0.95 in lowland area, Hokkaido University. This means that the difference of concentrations between southerly winds and easterly winds is considerably on the mountain than in lowland area. The westerly wind has a reversed correlation on the Hokkaido University campus in the lowland. In winter, the difference of concentration by wind direction on the mountain is more pronounced than in the lowland area. For example, each correlation coefficient between southerly winds and northerly winds at both sites shows 0.38 on the Mt. Teine and 0.78 at the Hokkaido University. Correlation coefficients of the TC and SO_4^{2-} concentration for each wind direction by altitude at both sites are shown in Table 3. Generally, except the northerly wind, the difference of anthro-

Table 2. The linear correlation coefficient of anthropogenic aerosol particles (TC and SO_4^{2-}) by wind direction in summer and winter periods.



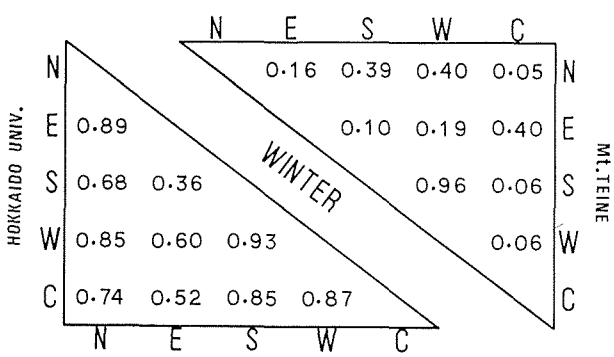


Table 3. The linear correlation coefficient of TC and SO_4^{2-} concentration in each wind direction by altitude at both sites

W.D. / SEASON	N	E	S	W	C
SUMMER	0.72	0.75	0.17	0.60	0.75
WINTER	0.98	0.54	0.08	0.06	0.45

pogenic aerosol concentrations by altitude in winter is greater than in summer. These results indicate, therefore, that the characteristic of the aerosol concentrations in Sapporo City in winter is influenced most remarkably by pollution related to the surface inversion layer of air temperature which is formed near the ground surface (Endoh et al., 1981). Correlation of vertical concentration by altitude in northerly winds is great without seasonal difference. It is assumed that the pollution source from northerly winds in the winter period does not differ clearly from in the summer period. In addition to this, in the northerly winds, the correlation by vertical altitude is high in winter, reflecting a strong monsoon wind from the north during the winter season.

4. Conclusions

The concentrations of anthropogenic and marine aerosol particles in winter period are higher than those in summer in lowland area measured by Lee et al. (1987). The concentration of anthropogenic aerosol particles on the summit of Mt. Teine is limited, in contrast to a large amount in lowland, Hokkaido University, since the surface inversion layer and the stable layer of air temperature are often formed between the lowland and the mountain. The amounts of the natural and soil aerosol particles show a great difference in concentration by altitude. This can be explained by the scavenging effect of heavy snowfalls on the summit of Mt. Teine in winter. It is considered that marine aerosol concentrations at lowland site are higher than in mountain area, since large and giant particles such as sea-salt particles are hard to be transported from the sea surface to the high altitude of the mountain because of their own weight and capture by surface obstacles. The TC and SO_4^{2-} aerosol particles which are mainly produced from anthropogenic sources, showed higher values on the Hokkaido University campus than on Mt. Teine. On the other hand, the difference of total mean concentrations by wind directions does not show any characteristic features in Sapporo City. The correlation of vertical anthropogenic aerosol concentrations by altitude under northerly winds is good without seasonal differences. From these results, it is concluded that the difference of total mean concentrations by wind directions was not great during the present sampling periods. However, the vertical change of anthropogenic aerosol particle concentrations by altitude was considerably higher in winter than in summer season in 1986.

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References

Endoh, T., Magono, C., Taniguchi, T., Murakami, M. and Yoshida, Y. (1981): Distribution of aerosols in vertical cross section above Sapporo. J. Env. Sci., Hokkaido Univ., **4**, 197–209 (In Japanese).

EPA (1973): Air Pollution Engineering Manual. AP-40, Chapter 2, 11–18, U. S. Environmental Protection Agency.

Khemani, L. T., Momin, G. A. and Naik, M. S. (1987): Influence of atmospheric pollutants on cloud microphysics and rainfall. Boundary-layer meteorology, **41**, 367–380.

Kikuchi, K. (1984): Meteorological Cyclopedia. Chapter 3, Sapporo Library No. 28, p. 85, 152–155 (In Japanese).

Kikuchi, K. and Yaura, S. (1970): Observations of giant sea-salt particles over the ocean from Tokyo to Syowa Station, Antarctica. Jour. Meteor. Soc. Japan, **48**, 377–380.

Lee, D. I., Taniguchi, T. and Kikuchi, K. (1987): Chemical compositions of atmospheric aerosol particles in relation to the wind direction at a high mountain and urban area in Sapporo, Japan. J. Env. Sci., Hokkaido Univ., **10**, 331–341.

Magono, C. and Endoh, T. (1979): 3-Dimentional observation of aerosol distribution above Sapporo City. Autumn Meeting of Meteorological Society of Japan, 36, p. 148 (In Japanese).

Murakami, M., Kimura, T., Magono, C. and Kikuchi, K. (1983): Observations of precipitation scavenging for water-soluble particles. Jour. Meteor. Soc. Japan, **61**, 346–358.

Ohta, S. and Okita, T. (1983): The composition of aerosols in Sapporo City. Autumn Meeting of Meteor. Soc. of Japan, p. 203 (In Japanese).

Sievering, H. (1987): Midwest/western/eastern U. S. precipitation and aerosol sulfate: differences attributable to natural source inputs? Atmos. Environ., **21**, 2525–2530.

Sladkovic, R., Reiter, R. and Potzl, K. (1986): Chemical components of the atmospheric aerosol at a high mountain station as a function of temporal trend and meteorological parameters, based on a 13-year measuring series. Atmos. Environ., **17**, 231–239.

Tanaka, M. and Toba, Y. (1969): Basic study on salt damage (3), Analysis of three dimensional distribution of sea-salt particles. Ann. Rep. Inst. Dis. Prev., Kyoto Univ., 12 B, 201–212 (In Japanese).

Usero, J. and Gracia, I. (1986): Trace and major elements in atmospheric deposition in the 'Campo de Gibraltar' region. Atmos. Environ., 1639–1646.

Wark, K. and Warner, C. F. (1976): Air Pollution, Its Origin and Control. Chapter 5, 153–154.

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