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Chemical Compositions of Atmospheric Aerosol Particles in Relation to the Wind Direction at a High Mountain and Urban Area in Sapporo, Japan

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Abstract

Sampling of aerosol particles using filter papers at the top of Mt. Teine (1,024 m a.s.l.) and on the campus of Hokkaido University in Sapporo City, Hokkaido, Japan was carried out in relation to the wind direction from August to October, 1986. The concentrations of Carbon, SO_4^{2-}, NO_3^-, NH_4^+, Cl^- and metal elements (Al, Na, Cu, Ni, K, Zn, Ca, Mn, Mg and Cd) were measured to reveal the characterization of source of the air pollution according to wind direction considering the meteorological parameters at both sites. Soil elements show a higher concentration than those of anthropogenic or marine origin, and which decreased with rainfall. The marine aerosol amounts in the urban area were higher than those on the mountain site. All concentrations of each element measured were in no way affected by the wind direction and the difference of altitudes during summer period.

Key Words: chemical compositions of aerosol, characterization of the source of air pollution, meteorological parameters, wind direction.

1. Introduction

Suspended particles and aerosols were produced from many sources including wind-blown dust, fossil-fuel combustion, metal ore processing and refuse incineration (Dutkiewicz et al., 1987). Pollutants produced from these various processes have some effects on human health (visibility, respiration and disease etc.) as well as the effect on vegetation or materials (EPA, 1973), and they will be changed according to the meteorological situations (Stoker, 1972). Especially, these phenomena are seen more frequently in urban areas as compared to rural areas. To determine the variation of aerosol concentrations by the wind direction, it is important to understand the present situation of air pollution in the urban and rural areas. 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 et al., 1975 ;

Usero and Gracia, 1986 ; Gregory et al., 1987). A detailed study, however, considering each wind direction has not been made till the present. Hence, in this study, aim to determine a background level of atmospheric aerosols by wind directions as against the winter, in as much as aerosol amounts in summer have shown the least values in Sapporo City (Ohta and Okita, 1983).

2. Materials and Methods

2. 1 *Sampling collection*

A characteristic of air pollution in Sapporo City which has a population of about 1,560,000 people is a typical urban style characterized by burning of fossil fuel for heating in winter seasons, the effluents of motor vehicles and so on. In addition, a serious social problem has arisen from street dust caused by studded tires of all vehicles in winter seasons (Kikuchi, 1984). Representative aerosol compositions occurring in this area showed soil dust, sea-salt, NH_4^+, NO_3^-, SO_4^{2-}, organic, elemental carbon and so on (Ohta and Okita, 1983). To obtain the difference between chemical compositions aerosol particles, air samplings using filter papers were out carried from August 8 to October 26, 1986 on the campus of Hokkaido University (50 m a. s. l.) situated in the middle of Sapporo City and Cloud Physics Observatory, Hokkaido University at the top of Mt. Teine (1,024 m a. s. l.) separated by 10 km to the west from the city center as shown in Figure 1.

An air sampler controlled by a selection circuit equipped with an aerovane (Environmental Pollution Control Center, Model WDP-100) is shown in Figure 2. The vane assembly sends a signal to the control unit when the wind blows from

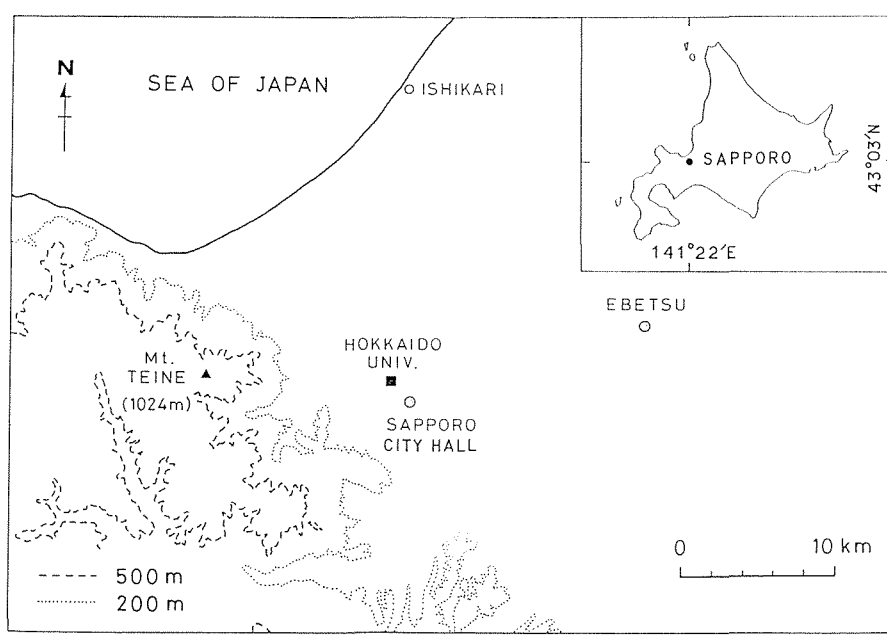

Figure 1. Location of Sapporo and sampling sites.

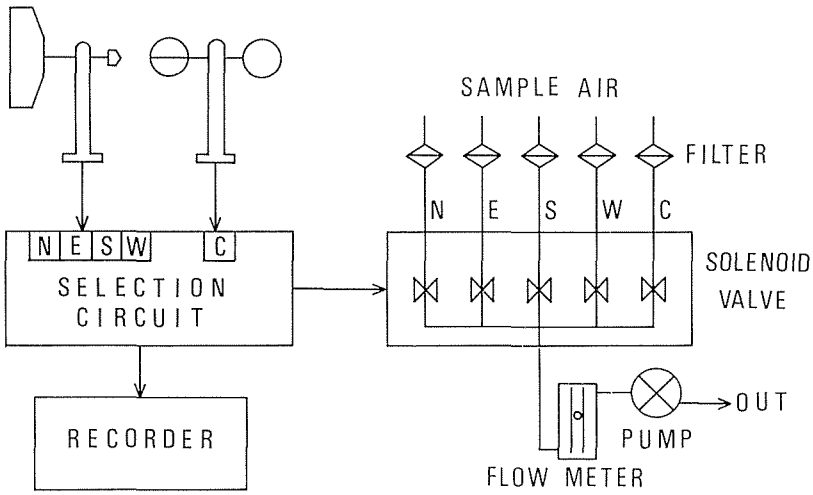

Figure 2. The wind direction controlled air sampling apparatus.

a selected direction sector of the aerovane. The vane and selection circuit unit assembly are used in conjunction with filter air sampler requiring an electrical solenoid valve to sample air when the wind is coming from a specific direction. Solenoid valve C (calm) is opened automatically, when wind speed is less than 0.5 m/sec. The quadrants of wind direction selected are from 45 to 134.9°(E), 135 to 224.9°(S), 225 to 314.9°(W), and 315 to 44.9°(N). Filters used in low-volume air sampling by each wind flow direction consist of two types; one is AF 07 P teflon filters for measuring ion components and metals contained in atmospheric aerosols, the other is for carbon measurements before aerosol sampling, Pallflex 2500 AQST quartz fiber filters are heated in an electric furnace at 850°C in air for an hour to remove carbonaceous contaminants.

2. 2 *Sample preparation and analysis*

Table 1 indicates analytical methods and instruments used to analyze aerosol samples. After sampling, several discs 1 cm in diameter are cut from the quartz fiber filter. One half of the discs are used to determine total carbon. The other half is heated in an electric furnace at 300°C in air for 30 min. to remove organic carbon, and then they are analyzed to determine elemental carbon. The difference in the amount of the total and elemental carbon gives the amount of organic carbon. The carbon determination is made on a combination with a NC—analyzer (Sumitomo Chemical Industry Inc., Model NC-80) and gas chromatography apparatus (Hitachi Inc., Model 163 FID) equipped with a nickel catalyst methanizer and flame ionization detector (Ohta and Okita, 1984). The amount of ammonium ion, nitrate, chloride and sulfate collected on the teflon filter are determined by an UV Spectrophotometer (Shimadzu Inc., Model 120-02 SFU) at 640 nm, 530 nm, 460 nm and 520 nm (APHA, 1977; APNC, 1975; Persson, 1966). Metal elements are analyzed by conventional atomic absorption methods (APHA, 1977) using Flameless Zeeman

Table 1. Analytical methods of individual item and instrument using analysis

Item	Analytical method	Instrument
T.C.	850°C, 1 hr	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	

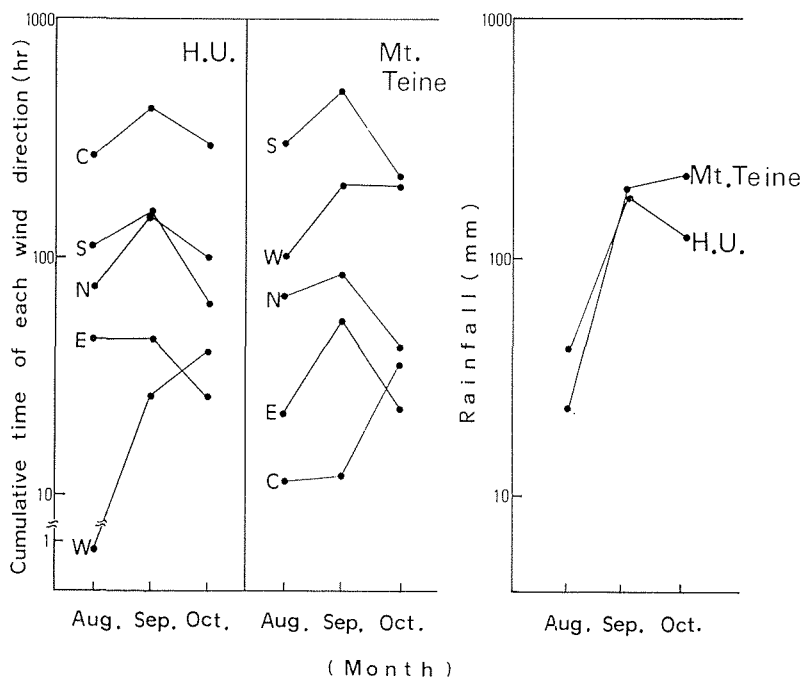

Figure 3. The frequency of monthly wind direction and precipitation amounts at sampling sites.

effect Atomic Absorption Spectrophotometer (Hitachi Inc., Model 170-70).

2.3 *Meteorological conditions*

During the sampling periods the frequency of monthly wind direction and precipitation amounts at both sites are measured. Figure 3 shows the results. There was a great difference between the cumulative time of wind direction at Hokkaido Univ. (H. U.) and that at Mt. Teine. In the low land site, the data of Hokkaido Univ., show a windless situation as compared with southerly winds at Mt. Teine. At both sites, the precipitation amounts of September and October are more than that of August.

3. Results and Discussion

Monthly mean values of the carbon and ion components in the aerosol are shown in Figures 4, 5 and 6. Although the cumulative time of the westerly wind has the least value (app. 50 min.) as shown in Figure 3, the mean concentrations of the westerly wind during August in the low land area, Hokkaido Univ., are higher than those measured for other wind directions. These results indicate that the higher concentrations are formed sporadically from the contaminant sources according to the westerly wind. In Figure 4, solid and open histograms show elemental and organic carbon, respectively. The concentrations of total carbon and EC/TC ratio at Hokkaido Univ. are higher than those at Mt. Teine. Northerly carbon average value shows less than other wind directions at Hokkaido University. As seen in the Figure 5, no data of NO_3^- are seen in October except for the easterly wind. The variation ranges of SO_4^{2-} concentration by each wind directions are higher at an urban area at Hokkaido Univ. as compared to Mt. Teine. SO_4^{2-} concentration in September at both sites show low values as compared with other months. In addition, this phenomenon is similar to NH_4^+ concentration indicated in Figure 6. Cl^- concentration at Hokkaido Univ. shows somewhat higher values than at Mt. Teine without any difference by wind direction. Figure 7 shows the total mean values of chemical compositions by each wind direction

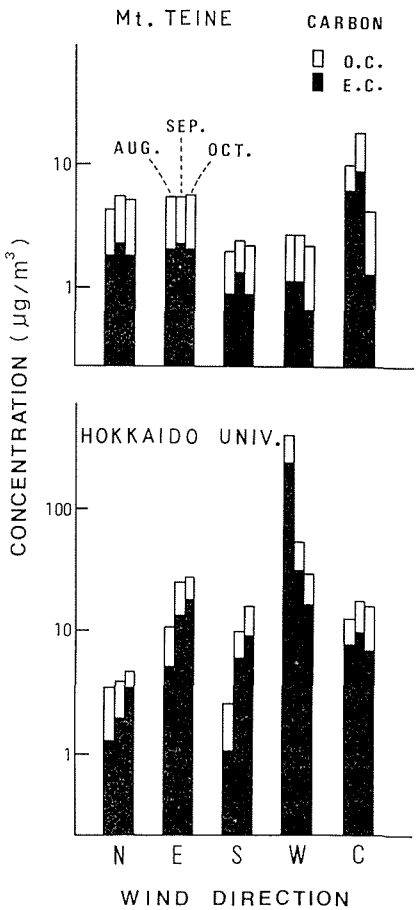

Figure 4. Monthly amounts of the organic and the elemental carbon in calm and each wind direction at sampling sites.

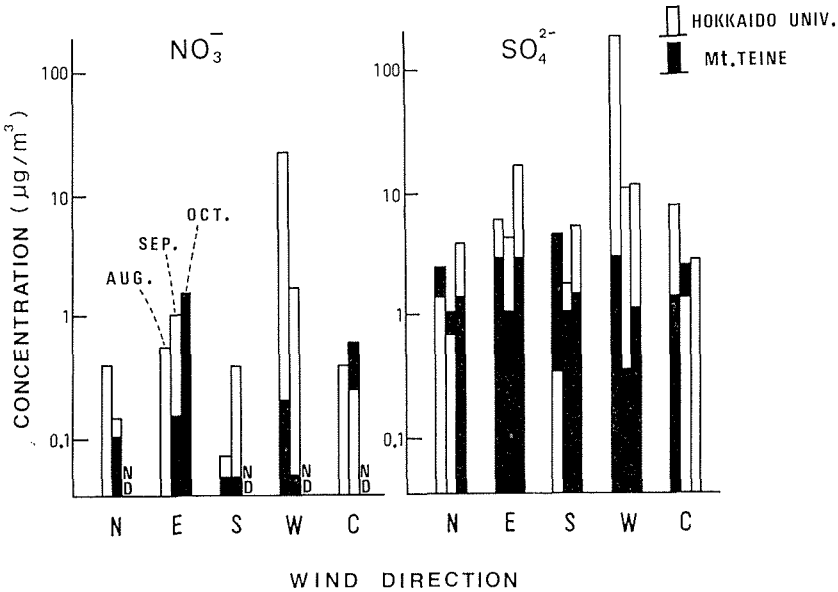

Figure 5. The same as Fig. 4 for NO_3^- and SO_4^{2-}.

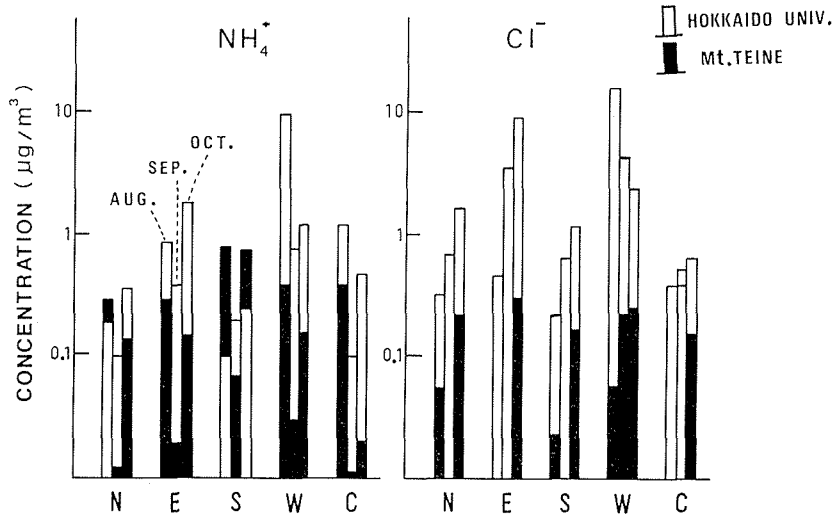

Figure 6. The same as Fig. 4 for NH_4^+ and Cl^-.

during the sampling period at Hokkaido University. In this figure, the arrangement of carbon, ion components and metal elements can be combined to form the representative origin source of three groups; NH_4^+, Ca, Cu, Zn, Al, Mn, and Ni from natural sources, Cl^-, K, Na, and Mg from marine aerosol source, and Cd, NO_3^-, TC, EC and SO_4^{2-} are of an anthropogenic source, respectively. In urban areas, the total average concentrations of each element respectively show no differences according to wind directions. But higher concentrations are recog-

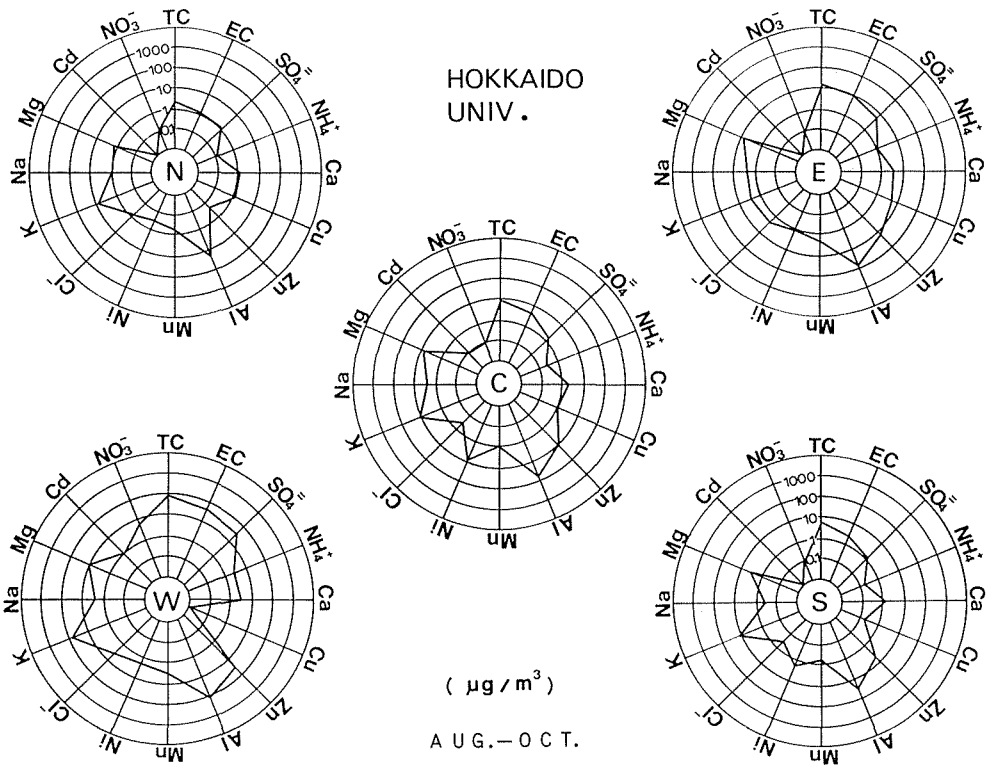

Figure 7. Total mean values of chemical compositions by each wind direction during the sampling period at the Hokkaido University.

nized when the wind directions are from the W rather than from other directions. The average total concentrations of crustal origin elements (Al, Zn etc.), marine aerosols (Na, K, Mg etc.) and anthropogenic aerosols (TC, EC, SO_4^{2-} etc.) for all wind directions approximately show greater levels than others. The results given in Figure 8 indicate the same as in Figure 7 at the Mt. Teine. In general, the average total concentrations of each elements at the mountain site show lesser values than in the urban area. Amounts of crustal origin elements showing higher concentrations than others have no relation with each wind directions at both sampling sites. In addition, aerosol concentrations at Mt. Teine during the sampling period do not show distinct differences by wind directions.

Figure 9 indicates, by each wind direction, concentration amounts ($\mu g/m^3$) at both sites of the NH_4^+ is dissolved from botanical and animal bacterias mainly and Al is formed by crustal and soil dusts mainly (natural origin sources), respectively. They show no great difference of concentration amounts by wind directions. Without any differences by wind directions at both sites, NH_4^+ and Al concentrations in September and October are lower than in August. These results suggest that the scavenging effect is influenced by precipitation amounts in September and October (Murakami et al., 1983).

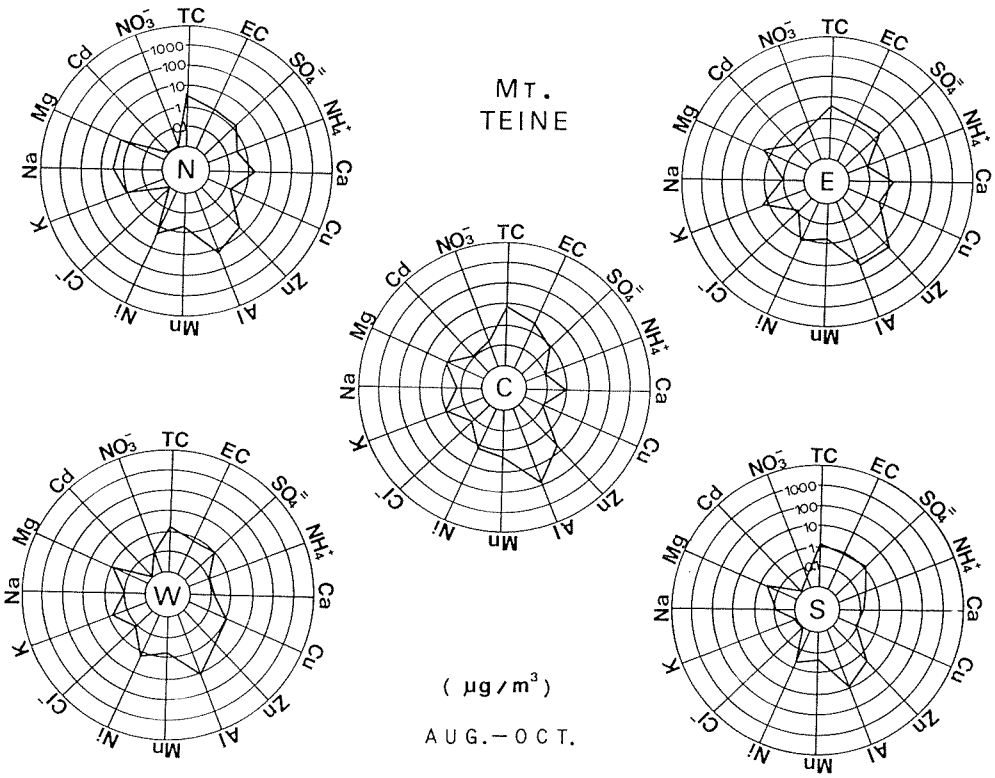

Figure 8. The same as Fig. 7 for the Mt. Teine.

Concentration amounts of Na and Cl^- usually originated from marine aerosol sources are shown in Figure 10. In the low land site, Hokkaido Univ., in August, Cl^- concentration with W wind is higher than others. As for the source of Cl^-, it is suspected that sea-salt may have been transported from the Japan Sea where is situated to the northwest of the city. On the other hand, at Mt. Teine, Na and Cl^- concentrations in October are higher than in August and September without any differences by each wind direction. It is assumed that in October, pressure pattern distributions of winter monsoon have formed strong convective instability in the layer between mountain and low land sites. Generally, marine aerosol concentrations at the low land site, Hokkaido Univ. are higher than at Mt. Teine. It is considered that large particles such as sea-salts are hard to be transported from the sea surface to the top of the mountain owing to have its own weight and capturing by surface obstacles (Tanaka and Toba, 1969; Kikuchi and Yaura, 1970).

As shown in Fig. 11, the concentrations of TC and SO_4^{2-} (anthropogenic origin) at both sites have no great differences by wind directions as well as vertical altitudes. This means that in this urban area, carbon and SO_4^{2-} amounts are mainly produced from coal, oil fuels and man-made pollutants are absolutely rare during the summer season. On the other hand, in the westerly winds, the difference of vertical concentration at both sites appears clearly, since high concentration with W wind in

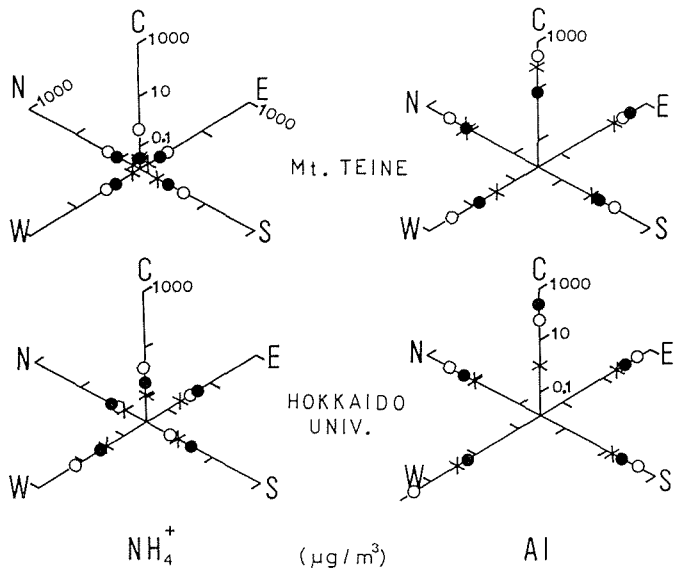

Figure 9. Vertical concentrations of NH_4^+ and Al by each wind direction.

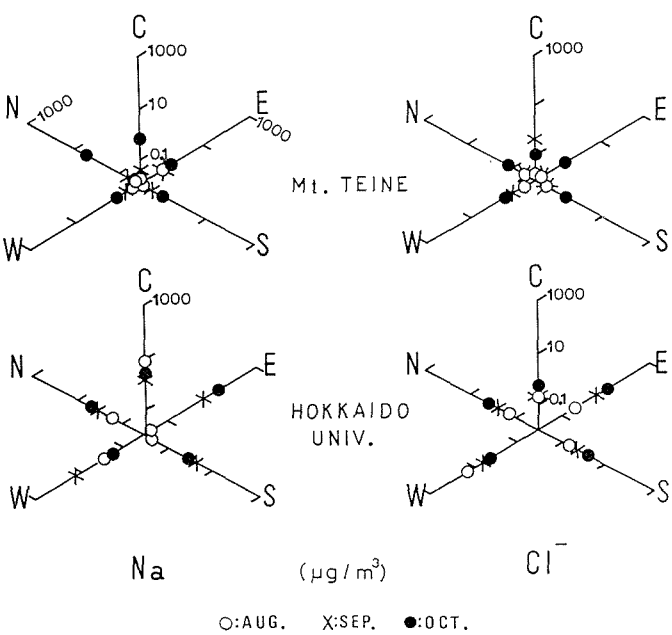

Figure 10. The same as Fig. 9 for Na and Cl^-.

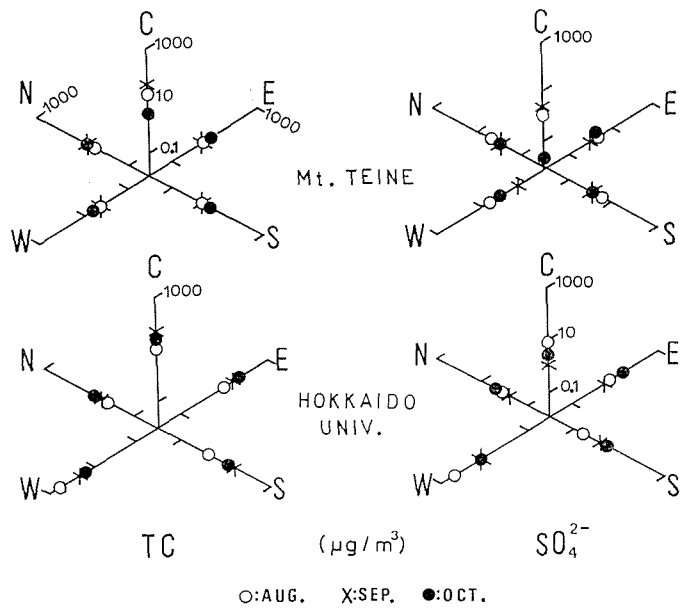

Figure 11. The same as Fig. 9 for Total Carbon and SO_4^{2-}.

surface layer at urban area came into existence during sampling period.

4. Conclusions

Long—term sampling of aerosols by wind directions was performed from August to October, 1986 at the surface layer in urban area of Hokkaido University and Mt. Teine in Sapporo City. In this study, we found that vertical variations of marine aerosols (K, Na, Mg) has much higher than the others. On the other hand, the elements of a predominantly natural origin (Al, Mn, Ni) show a very little variation of vertical concentrations at both sites. Components that are usually associated with anthropogenic emissions, such as carbon, SO_4^{2-}, NO_3^- and Cd showed respectively low differences of vertical concentrations with each wind direction, since the anthropogenic pollution sources have decreased during summer in this urban area. From these results it was found that difference of concentrations by wind directions is not great because the absolute value measured during the summer is very smaller than in other seasons.

We therefore presume that vertical variations by each wind direction should be obtained if it were observed during the winter, since the air quality of urban area such as Sapporo City in winter is influenced most markedly by pollution related to the temperature inversion layer which is formed from near the ground surface (Endoh et al., 1981).

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References

Air Pollution Natural Conference, Japan (1975): Air Pollutants Handbook, Vol. 1, Measurements, p. 269 (in Japanese).

APHA Intersociety Committee (1977): Methods of Air Sampling and Analysis, 2nd Ed., 507–511, 538–541, 592–601, 839–841.

Dutkiewicz, V. A., Parekh, P. P. and Husain, L. (1987): An evaluation of regional elemental signatures relevant to the northeastern United States. Atmos. Environ., 21, 1033–1044.

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.

Gregory, S., Carl, E. and Glen, E. (1978): Chemical element balances and identification of air pollution sources in Washington, D.C. Atmos. Environ., 12, 1143–1153.

Hammerle, R. H. and Pierson, W. R. (1975): Sources and elemental composition of aerosol in Pasadena, California, by energy-dispersive X-ray Fluorescence. Environ. Sci. & Tech., 9, 1058.

Kikuchi, K. (1984): Meteorological Cyclopedia, Chapter 3, Sapporo library No. 28, 152 (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.

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

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

Ohta, S. and Okita, T. (1984): Measurements of particulate carbon in urban and marine air in Japanese areas. Atmos. Environ., 18, 2439–2445.

Persson, G. A. (1966): Automatic colorimetric concentration of sulphate for measuring sulphur dioxide in ambient air. Int. J. Air. Wat. Pollut., 10, 845–852.

Stoker, H. S., Spencer, L. S. (1972): Environmental Chemistry, air and water pollution, Part 1, 78–79.

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., 12B, 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.

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