ON THE ORIGIN AND NATURE OF ANTARCTIC AEROSOLS

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Abstract: In order to obtain a better understanding of the properties of the aerosols in "clean" air, a series of aerosol measurements was carried out at Syowa Station, Antarctica, from February 1977 to December 1978. From these measurements, it was found that the Aitken particle concentration, Mie particle concentration and Mie particle size distribution exhibited the characteristic annual variations caused by different mechanisms. The possible mechanisms which could cause these annual variations were discussed. It is suggested that the annual variation of Aitken particle concentration is caused by the seasonal variation of the influx of solar photons into the earth's atmosphere. It is also suggested that the annual variation of Mie particle concentration is mainly caused by the seasonal variation of influx of the maritime air into Antarctica, and that the annual variation of Mie particle size distribution is mainly caused by the seasonal variation of distance from the open sea. It is also suggested that in summer the photochemical growth of Aitken particles affects the regularity of the annual variations of the concentration and size distribution of Mie particles.

1. Introduction

In order to provide a better understanding of the concentrations and properties of Antarctic aerosols, a series of systematic aerosols measurements was undertaken at Syowa Station (69.0°S, 39.6°E), Antarctica, for two consecutive years from February 1977 to December 1978.

The instruments and techniques applied in the measurements were as follows:

The concentration and mean size of Aitken particles ($0.002 \leqq r \leqq 0.2\ \mu$m) were measured with a Pollak counter of MRI-type (ITO, 1976). The counter was equipped with a diffusion pipe for the measurement of diffusional loss of particles from which information on the mean size of the particles could be obtained. It was also equipped with an electric current balancing circuit (a kind of null method circuit) which was needed for the precise measurement of very low concentration

of the particles. The size distribution of Aitken particles was measured by a diffusional decay technique which was reported in a previous paper (ITO, 1980a). The volatility of Aitken particles was measured by passing sample air through a heated quartz tube prior to counting with the Pollak counter. The technique is similar in principle to that used by TWOMY (1968). The concentration and size distribution of Mie particles ($0.2<r\leqq 1\ \mu$m) were measured with an optical particle counter (Dan Co., Ltd., Japan) which is similar in principle to a Royco light-scattering particle counter. Sequential sampling of aerosol particles with an impactor and an electrostatic precipitator (Thermo-System Inc., U.S.A.) was done, and the particles were subjected later to physical and chemical investigation of individual particles by means of electron microscopy. Some of the results obtained from these measurements have been reported (IWAI, 1979; IWAI *et al.*, 1978; ITO, 1980b, c; ITO and IWAI, 1981; IWAI *et al.*, 1981; ONO *et al.*, 1981; KOIDE *et al.*, 1981; ITO, 1982).

This note is an extended abstract to give recent results obtained through further analysis of the data from the above measurements.

2. Aitken Particles

The median value of hourly mean concentration of Aitken particles obtained from February to December 1978 was 223 cm^{-3}. This concentration is comparable to those in clean atmosphere outside Antarctica.

The monthly mean concentrations obtained at Syowa Station during 1977 and 1978 showed a regular annual variation having a maximum in March and a minimum in June or July (ONO *et al.*, 1981). A similar annual variation was also

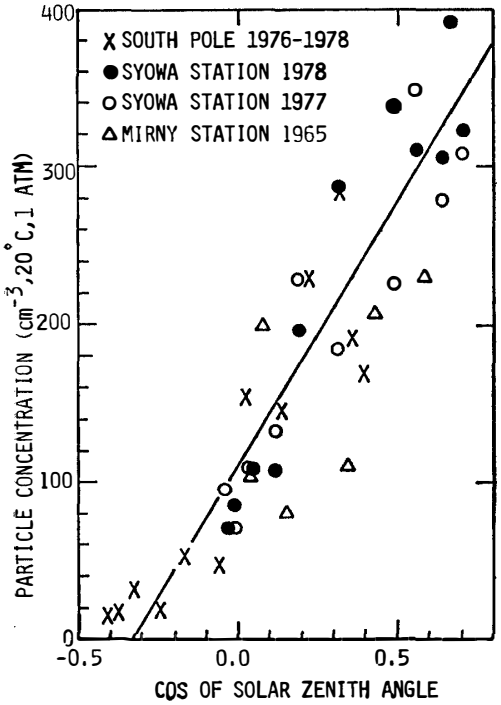

Fig. 1. Monthly mean concentration of Aitken particles as a function of solar zenith angle at each location at the northing time on the middle day of the month.

observed at Mirny Station in 1965 (VOSKRESENSKII, 1968) and at South Pole Station every year from 1975 to 1978 (MURPHY and BODHAINE, 1980). From these observations, it has become clear that as a common feature throughout Antarctica, the concentration of Aitken particles shows a quite regular annual variation having a maximum in the sunlit months and a minimum in the polar night months. This feature may suggest that solar radiation plays an important part in supplying the Antarctic atmosphere with Aitken particles.

The relation between solar radiation and Aitken particle concentration can be shown more clearly by Fig. 1. In this figure the monthly mean concentration is plotted against the cosine of solar zenith angle at each station at the northing time on the middle day of the month. The concentration at the South Pole was reproduced from the table reported by MURPHY and BODHAINE (1980), by taking an average for three years from 1976 to 1978. The concentration at Mirny was reproduced from the tables reported by VOSKRESENSKII (1968), by taking a weighted mean of the monthly mean concentrations determined separately for two kinds of air mass, that is, the continental Antarctic air and the maritime polar air. All concentrations plotted were assumed to have been measured at room temperature and were converted to values at sea level pressure. Since the concentrations in March in both 1977 and 1978 at Syowa Station were too high to plot in the same figure, those were excluded. The straight line in the figure is the least square fit for the plotted data, except those at Mirny.

Figure 1 shows that the monthly mean concentration of Aitken particles, which varies seasonally and spatially in the Antarctic atmosphere, can be related to a single parameter, that is, the solar zenith angle. The cosine of the solar zenith angle, when multiplied by the solar constant, represents the influx of solar photons which enter the earth's atmosphere. It is thought that the solar photons play an important part in the atmospheric processes of gas-to-particle conversion. Therefore, Fig. 1 can be considered to show that the seasonal and spatial variations of monthly mean concentrations of Aitken particles can be attributed to the variation of production rate of the photochemical particles. It is suggested that in Antarctica the most significant source of Aitken particles is the photochemical reactions which cause the gas-to-particle conversion in the Antarctic troposphere.

In Fig. 1, the concentration keeps a close correlation to the solar zenith angle even in the polar night months when the influx of solar photons is almost negligible. The cause of this phenomenon can be understood by considering that the monthly mean of aerosol concentration implicitly has the character of a kind of spatial mean taken over a wide area. Even in the polar night month, the photochemical production of Aitken particles takes place in the sunlit atmosphere over the Subantarctic region. As a result of macroscale eddy diffusion, the Subantarctic air mixes with the Antarctic air. It does not take more than several days until the air of Subantarctic origin arrives in the South Pole through such mixing. The residence time of Aitken particles in the troposphere is of the order to several days. Therefore, it seems that the Aitken particles which exist at the South Pole in the polar night months originate in the Subantarctic atmosphere.

3. Mie Particles

The median value of hourly concentrations of Mie particles obtained at Syowa Station from February to December 1978 was 0.77 cm^{-3}. The size distribution averaged for the above period could be approximated as a power law distribution $dn/d \log r \propto r^{-\beta}$, where $\beta = 4.17$. At places other than Antarctica, the representative value of the concentration is higher than 1.0 cm^{-3}, and that of β is equal to 3.0 or less. A large value of β means that the aerosol has suffered a loss of larger particles, or a supply of smaller particles, in the size range of Mie particles. Thus, Antarctic aerosols, when compared with aerosols in the clean atmosphere outside Antarctica, can be characterized by the low concentration of Mie particles and a size distribution with a large β-value resulting probably from the preferential loss of large particles.

The monthly mean concentration obtained at Syowa Station during 1978 showed an annual variation having a maximum in September and a minimum in February. The β-value of the monthly averaged size distribution showed an annual variation having a maximum in December and a minimum in May. The causes of these annual variations of concentration and size distribution will be discussed below.

Figures 2 and 3 show the relation between the particle concentration and the influx of the maritime air flowing into the Antarctic region. The abscissae of both figures represent the monthly mean of southward components of the wind at the 850 mb level over Syowa Station. The ordinate of Fig. 2 represents the monthly

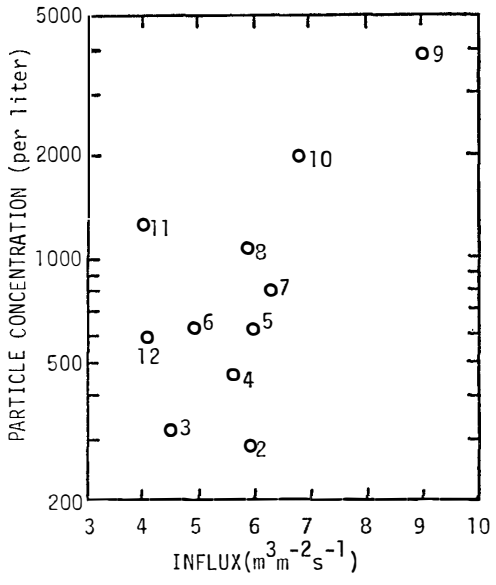

Fig. 2. Monthly mean concentration of particles larger than 0.37 μm in diameter as a function of monthly mean influx of the maritime air. The numbers in the figure indicate the months.

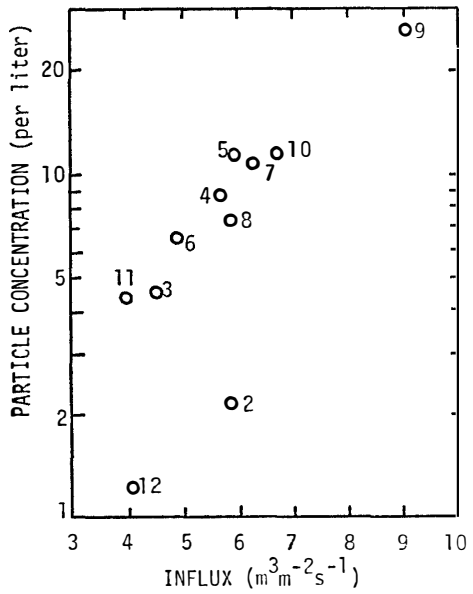

Fig. 3. Same as Fig. 2 except for the concentration of particles larger than 1.1 μm.

mean concentration of Mie particles whose diameter is larger than 0.37 μm, whereas Fig. 3 is for particles larger than 1.1 μm which are the larger size component of Mie particles.

In both figures, most of the plotted points approximately form lines which indicate a tendency for the monthly mean concentration of particles to appear to have a larger value in a month with a larger influx of maritime air. This is more clearly seen in Fig. 3. In Fig. 3, the plotted points, except two for February and December, are distributed along the line passing through the data points for September and November. The low concentration in February or December in Fig. 3 may be attributed to the loss of particles due to the cloud physical removal, from the fact that the cloud coverage and precipitation in the region between 55°S and 65°S increase in the austral summer (VAN LOON, 1972).

A similar phenomenon of low concentration in summer months may also be expected in Fig. 2, but it is not clearly seen. It might be masked by other effects which may cause the increase of particles in summer. In summer, aerosol particles may grow in size due to the condensation of vapor substances produced by photochemical reaction in the atmosphere. For particles larger than 0.1 μm in diameter in the natural atmosphere, the number concentration of particles is higher for those of smaller size than those of larger size, and the growth rate is larger for smaller particles than for larger particles. Therefore, the increase in concentration due to the photochemical growth of particles in summer is more appreciable in the size 1.1 μm, as shown in Fig. 3. Thus, it is reasonable that the relation between the range of total Mie particles as shown in Fig. 2 than in the size range larger than concentration and the influx of maritime air, or the phenomenon of irregularly low concentration in summer, appears rather clearly in Fig. 3 and less clearly in Fig. 2.

From Figs. 2 and 3, it can be concluded that the annual variation of monthly mean concentration of Mie particles in the Antarctic atmosphere is mainly caused by variation of the influx of maritime air, although this regularity is disturbed in

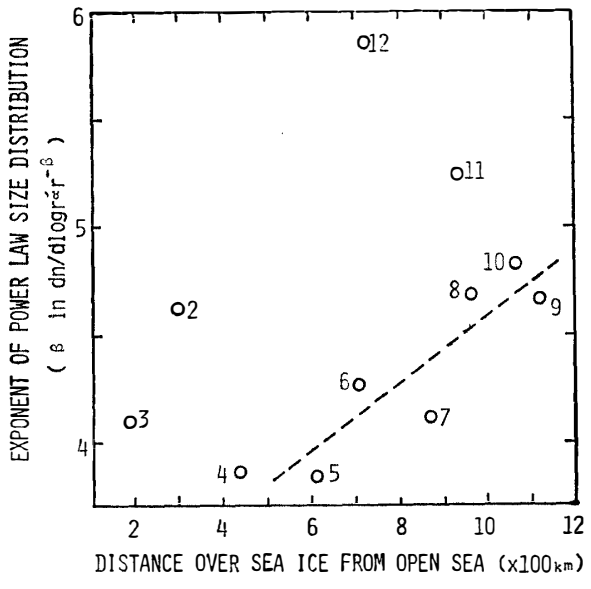

Fig. 4. The exponent of the monthly averaged size distribution, when approximated in a form of $dn/d\log r \propto r^{-\beta}$, as a function of distance over sea ice from open sea. The numbers in the figure indicate the months.

summer by the effect of photochemical growth of particles and probably by the effect of cloud-physical removal which becomes active in summer.

A similar explanation can be given for the annual variation of size distribution of Mie particles. Figure 4 shows the relation between the size distribution and northward extent of the sea ice region around Antarctica. The abscissa of Fig. 4 represents the meridional distance from the northern edge of the sea ice region to the fringe of the polar ice cap. The distance was calculated from the monthly area of the sea ice region around Antarctica, listed in SCHWERDTFEGER (1970). The ordinate of Fig. 4 represents the β-value of the monthly average size distribution of Mie particles.

In Fig. 4, the data for seven months from April to October roughly form a line. The line indicates a tendency for the monthly averaged size distribution to appear to have a larger β-value in a month with larger northward extent of the sea ice region around Antarctica.

According to the discussion of Figs. 2 and 3, the main components of Mie particles at Syowa Station in the months from April to October are thought to be brought with maritime air originating from the Subantarctic oceanic region. Therefore, the dependence of the β-value on the northward extent of the sea ice region, as seen in Fig. 4, suggests that the Mie particles in the maritime air suffer a change in their size distribution during their traveling over the sea ice region. Since β increases with the increase in the northward extent of the sea ice region, the deformation of size distribution seems to be caused by the preferential losses of large particles in the Mie particle size range. Such kinds of particle loss can be attributed to the inertial impaction of particles on the snowy surface with extreme roughness in the sea ice region.

On the other hand, in the remaining four months in Fig. 4, February, March, November, and December, the β-value of size distribution appears to have a large value which does not obey the regularity, that is, the β-value depends on the northward extent of the sea ice region. Since these four months are sunlit months with plenty of solar radiation available, the irregularly large β-value in these months can be attributed to the supply of the small Mie particles which have grown from Aitken particles by the condensation of vapor substances produced by photochemical reactions in the Antarctic atmosphere.

Therefore, Fig. 4 can be considered to show that the annual variation of size distribution of Mie particles at Syowa Station is caused mainly by the variation of northward extent of the sea ice region over which the maritime air has to travel, although this regularity is disturbed in summer by the effect of photochemical growth of Aitken particles.

4. Concluding Remarks

In the present work, it is clarified that the Aitken particle concentration, Mie particle concentration, and Mie particle size distribution in the Antarctic atmosphere exhibit the characteristic annual variations caused by different mechanisms. The annual variation of Aitken particle concentration is caused by the seasonal varia-

tion of influx of solar photons which relates to the photochemical production of particles in the Antarctic atmosphere. It is suggested that Aitken particles in the Antarctic atmosphere seem to be produced mainly by photochemical reaction in the Antarctic atmosphere and are partly brought by maritime air coming from the Subantarctic oceanic area. The annual variation of Mie particle concentration is mainly caused by seasonal variation of southward flux of the maritime air coming from the open sea whereas that of the Mie particle size distribution is mainly caused by seasonal variation of extent of the sea ice region around Antarctica. Mie particles in the Antarctic atmosphere seem to be brought mainly by the maritime air from the Subantarctic oceanic region and partly by the photochemical growth of Aitken particles in summer.

From previous works (ITO and IWAI, 1981; ONO *et al.*, 1981; ITO, 1982) which were based on the measurement of size distribution of Aitken particles, the volatility measurement of Aitken particles, the morphological identification of individual aerosol particles by means of electron microscopy, and the analysis of day-to-day variations of aerosol concentrations, it was concluded that two types of aerosol originating from different sources are present in the Antarctic atmosphere. With the intrusion of maritime air in cyclonic storms, mostly in the winter months, aerosols composed of sea salt particles and ammonium sulfate aerosols contained originally in the clean maritime air are dominant. On the other hand, in the summer months, aerosol in the chemical form of sulfuric acid droplets is predominant. Those sulfuric acid aerosols are formed by photochemical oxidation of sulfur-bearing gases in the sunlit troposphere and are transported down into the surface boundary layer under certain circumstances by meteorological processes.

The present results are accepted as further evidence for the previous conclusions mentioned above.

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