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The Coarse Particle Aerosols in the Free Troposphere Around Japan

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Airborne measurements of solar aureole intensities were carried out around Japan with a spectral scanning radiometer. Horizontal and vertical stratifications and volume spectra of tropospheric aerosols were retrieved from the measured solar aureole intensities. The results show that bimodal volume spectra of aerosols with a saddle point radius around 0.5 μm are generally observed in the troposphere, both within and above the planetary boundary layer. A submicrometer particle mode of the spectra, known as the accumulation mode, dominates the coarse particle mode within the planetary boundary layer, especially in summer, whereas coarse particles were observed frequently in the free troposphere. Synoptic analyses with air mass trajectories, isentropic surfaces, and weather charts suggest that the windblown dust particles injected into the atmosphere in arid regions of eastern Asia are the origin of the coarse particles observed in the free troposphere around Japan. Windblown dust particles are elevated to the free troposphere by ascending air motion in a warm front and then transported by the westerlies. The arid regions to the east of the Tibetan Plateau, where disturbances due to the frontogenesis occur frequently, are capable of supplying coarse particles to the free troposphere over the northwestern Pacific Ocean throughout the year.

1. INTRODUCTION

In spite of large spatial and temporal variabilities in the physical and chemical properties of tropospheric aerosols, the aerosols' measured size distributions show some common characteristics associated with the source and transport mechanism [*Shaw*, 1977; *Patterson and Gillette*, 1977]. Soil- or sea-salt-derived particles are apt to have micrometer-sized radii (the coarse particle mode), whereas secondary aerosols generally converted from gaseous constituents are apt to have submicrometer-sized radii (the accumulation mode). This fact has been a strong motivation for measuring the aerosol size distribution in order to estimate aerosol origin.

Measurements of the aerosol size distribution are also important for the study of aerosol impact on the climate system of the Earth, because the effects are quite size dependent. Submicrometer particles efficiently affect the radiation budget of the Earth-atmosphere system by scattering solar radiation [*Yamamoto and Tanaka*, 1972; *Coakley et al.*, 1983] and by modulating cloud properties through their role as cloud condensation nuclei [*Twomey et al.*, 1984; *Wigley*, 1989; *Radke et al.*, 1989]. On the other hand, coarse particles are important in geochemical cycling and are also used as a tracer material for studying atmospheric transport. Extensive studies have been performed for the transport of Saharan dust particles to the European and the North Atlantic regions [*Prospero and Carlson*, 1972; *d'Almeida*, 1986] and of Asian dust particles to the North Pacific region [*Duce et al.*, 1980; *Uematsu et al.*, 1983; *Merrill et al.*, 1985, 1989]. These dust particles are the main deposition material on remote ocean floors because of long-range atmospheric transport.

In the present paper we will describe the results of airborne measurements of aerosol sizes and analyses of the spatial structure and the transport process of tropospheric aerosols around Japan. Since Japan is highly populated and is located downstream of the Asian dust origin, measurements of the size distribution of aerosols are interesting for studying the characteristics of aerosols which consist of both accumulation and coarse particle modes. Particular attention will be paid to coarse particles in the free troposphere, because a notable number of coarse particles have often been observed in the free troposphere not only over the continent [*Blifford and Ringer*, 1969; *Fitch and Cress*, 1981, 1983] but also over the central Pacific far from the continent [*Patterson et al.*, 1980].

Rather than employ the conventional method of using particle counters and impactors, we employed an optical remote sensing technique for the estimation of aerosol size distribution by inverting solar aureole intensities measured on board aircraft [*Nakajima et al.*, 1983, 1986]. Although many observations of aerosol size distribution have been performed in the planetary boundary layer, few have been performed above the planetary boundary layer. A major reason is the fact that the sampling air volume must be quite large in order to collect a sufficient number of aerosols using conventional methods. Our method does not suffer from this limitation, because we directly retrieve the size distribution of columnar aerosols. Also, the remote sensing technique we employed does not change the aerosol properties by air sampling. This is an important advantage especially under high humidity conditions [*Tanaka et al.*, 1990]. The results obtained by airborne observations are interpreted using trajectories and isentropic surfaces derived from weather charts and radiosonde data of the Japan Meteorological Agency.

2. RETRIEVAL OF AEROSOL SIZE DISTRIBUTION FROM THE SOLAR AUREOLE

The solar aureole is the region of enhanced brightness seen around the Sun, which is caused by the strong forward scattering of sunlight due to aerosols. Since the phase function, especially its forward part, depends on the size distribution of aerosols, we can infer the aerosol sizes from the aureole measurements. The use of aureole measurements to retrieve the size distribution of aerosols has an advantage over the use of spectral extinction measurements

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of direct solar radiation [*Yamamoto and Tanaka*, 1969; *King et al.*, 1978], because it is easier to estimate particle sizes in a wider range by inverting the differential scattering coefficient (volume scattering phase function) than by inverting the spectral extinction coefficient [*Nakajima et al.*, 1986].

For airborne measurements it is quite difficult to determine the optical thickness of aerosols above the planetary boundary layer from observation of direct solar radiation because of errors involved in the calibration constants and in the observed intensities. These errors are affected strongly by temperature variation under typical flight conditions. Therefore we use the normalized aureole intensity given by equation (1), which is less affected by the errors mentioned above:

$$R(\Theta) = E(\Theta)/mF\, d\Omega \tag{1}$$

where $E(\Theta)$, F, m, and $d\Omega$ are the observed monochromatic intensity of the solar aureole as a function of the scattering angle Θ, the intensity of the direct solar radiation, the optical air mass, and the solid view angle of the radiometer, respectively. The normalized aureole intensity $R(\Theta)$ becomes the differential scattering coefficient $\beta(\Theta) = \omega_a \tau_a P(\Theta)$ for the single-scattering approximation, where ω_a, τ_a, and $P(\Theta)$ are the single-scattering albedo, the optical thickness, and the normalized phase function of aerosols, respectively.

The retrieval of aerosol size distribution from the aureole intensity distribution consists of two processes. The first process is to extract the phase function of aerosols by removing the multiple-scattering effect. The second process is to find the aerosol size distribution by solving the integral equation relating the phase function with the size distribution $dV/d \ln r$, where V is the cumulative volume distribution with respect to the particle radius. The final results for volume spectrum and optical thickness of columnar aerosols are simultaneously determined by iterating the above two processes so as to adjust the theoretical normalized intensities to the observed ones. Calculated and observed aureole intensities coincide with each other within several percent. Formulations for the iteration are the same as those given by *Nakajima et al.* [1983, 1986]. Although aerosols are not spherical particles in most cases, we assume that the phase function of aerosols is expressed by that of optically equivalent spherical particles. This assumption is fairly reasonable for the interpretation of the solar aureole, because differences in the forward part of the phase function are very small between spherical and nonspherical particles [*Asano and Sato*, 1980; *Pollack and Cuzzi*, 1980; *Koepke and Hess*, 1988]. The complex refractive indices of aerosols are assumed to be $1.45–0i$ and $1.50–0.01i$ for the data obtained in remote areas over the northwestern Pacific Ocean and an urban area over Nagoya, respectively [*World Meteorological Organization* (WMO), 1983]. Optical thicknesses of air molecules for the respective wavelengths were calculated using the formulation of *Fröhlich and Shaw* [1980], slightly modified by *Young* [1981].

3. Instrumentation and Observations

3.1. *Instruments*

A spectral scanning radiometer (aureolemeter) was used to measure both direct solar and aureole radiances with a high precision. Details of the instrument are described by *Nakajima et al.* [1986], so that only a brief description will be given here.

The airborne aureolemeter is a spectral scanning radiometer with a light detector of a positive-intrinsic-negative silicon photodiode and seven interference filters for wavelengths of 332, 368, 500, 675, 777, 862, and 937 nm. The viewing field of the aureolemeter has a solid angle of 1.52×10^{-3} sr, which can be aimed at any direction by an altazimuth dc servomotor system. The position of the Sun is continuously monitored by a two-dimensional silicon position sensitive detector. The minimum measurable angle from the Sun with a sufficient accuracy is about 5°. Normalized aureole intensities at 13 selected scattering angles from 5° to 30° were used in the analyses. The aureolemeter was calibrated several times a year in cloudless conditions using a modified Langley plot method proposed by *Tanaka et al.* [1986]. The scatter in the calibration constants during the observational period was within 2% for the wavelengths of 500, 675, 777, and 862 nm and within 5% for the other wavelengths.

In addition to the aureolemeter, upward and downward looking spectral pyranometers were used to measure the radiative fluxes at the same wavelengths as the aureolemeter. The measured albedos of the underlying atmosphere-surface system were used in the analyses by assuming the law of Lambertian reflection. The details of the flux measurements are described by *Tanaka et al.* [1990].

3.2. *Observations*

Measurements of the solar aureole were carried out as a part of the Japanese Middle Atmosphere Program [*Kato*, 1987] around the northwestern Pacific Ocean to the south of Japan by a Swerlingen Merlin IV aircraft and over Nagoya, a typical urban area in Japan, by a Cessna 206 aircraft. Figure 1 shows flight courses of the observation.

Over the Pacific Ocean, measurements were performed along north-south lines at several altitudes up to the level of 480 mbar. The flights covered the latitudes from 29.6°N to 34.5°N along the longitude of 135°E on February 16, 1984, from 19.9°N to 34.5°N along 140°E on August 7–8, 1984, and from 27.9°N to 33.1°N along 140°E on December 16–17, 1985. Over Nagoya (35.2°N, 136.2°E), measurements were performed around noon under cloudiness conditions on January 14, November 7–8, 1984, October 9, 1985, and July 28 and 30, 1986. The aircraft ascended and descended continuously at altitudes up to 490 mbar within an area of about 30 km in length and 10 km in width.

4. Size Distribution and Transport Process of Aerosols

4.1. *Results From Observations Over the Northwestern Pacific Ocean*

Among three series of aureole measurements over the northwestern Pacific Ocean the data of August 7 and 8, 1984, can give a latitudinal variation of summertime aerosol stratification [*Nakajima et al.*, 1986]. It is found that the retrieved optical thickness of aerosols decreases with increasing distance from the Japan Islands and height from sea level. The latitudinal variation of the optical thickness of aerosols is large in the lower troposphere and decreases with increasing height. The optical thickness of $\lambda = 500$ nm varies from 0.2

(34°N) to 0.04 (20°N) below the height of 900 mbar and from 0.1 (34°N) to 0.03 (20°N) at 700 mbar. At the maximum flight levels above 600 mbar the value of optical thickness is almost constant, for example, about 0.03 at λ = 500 nm, independent of latitude.

Retrieved volume spectra of aerosols contained in the air column above the 700-mbar level are represented in Figure 2 for several latitudes. Since the measured aureole intensities were restricted with respect to the scattering angle and wavelength, size distributions are reliable only for radii between 0.1 μm and 3 μm. Each line in the figure was obtained by averaging several volume spectra retrieved from the aureole data around the indicated latitude. The variability of the volume spectrum at each latitude is ±10% at most for particle radii around 0.5 μm and about ±50% at most for extreme radii around 0.1 μm and 3μm, respectively. These variabilities are ascribed to those of ±10% in the measured aureole intensities. A volume spectrum with a saddle point radius of around 0.5 μm prevailed near the Japan Islands. The accumulation mode of the spectrum decreased at first with distance southward from the Japan Islands. A sudden decrease of the coarse particle mode occurred around the latitude of 25°N, and a monomodal volume spectrum was observed in the lower latitudes. This monomodal distribution is closely similar to those obtained at the uppermost level, as shown by a dashed line in Figure 2. In this level, the volume spectrum of aerosols is a monomodal function of

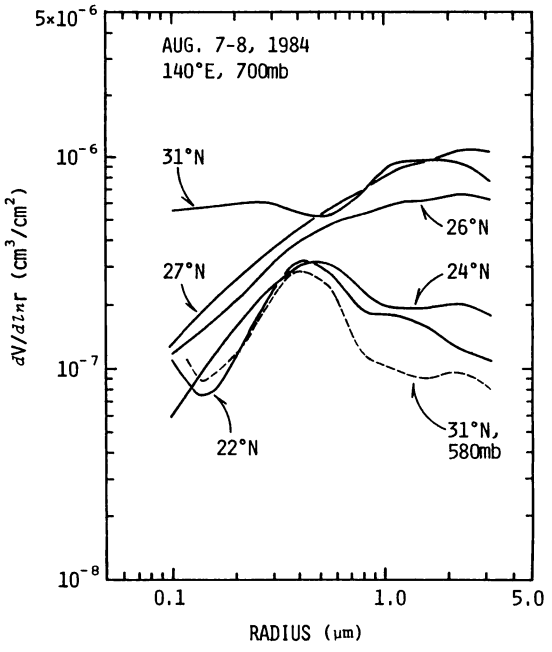

Fig. 2. Volume spectra of columnar aerosols observed at 700 mbar at several latitudes (solid lines) and at 580 mbar at 31°N (dashed line) along 140°E on August 7 and 8, 1984.

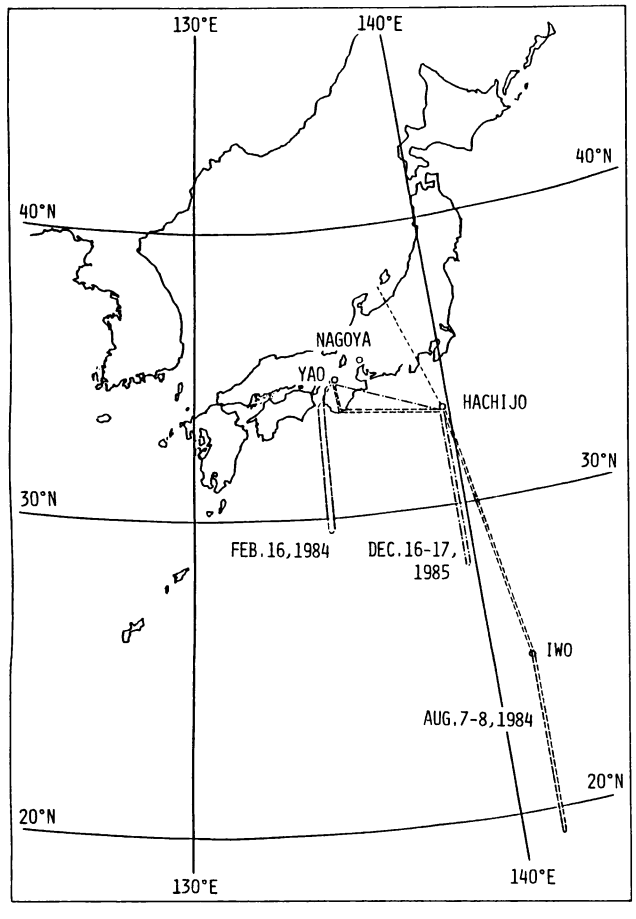

Fig. 1. Flight course of the northwestern Pacific Ocean observations and the location of Nagoya, Japan.

radius with a mode radius of 0.4 or 0.5 μm in almost all latitudes of observation. These monomodal volume spectra are quite similar to those obtained over Nagoya at the maximum flight levels around 500 mbar 3 months later and are attributed to the enhanced stratospheric aerosols due to the El Chichon eruption in the spring of 1982 [*Nakajima et al.*, 1986].

In order to study the role of long-range transport on the horizontal variability of the aerosol size distribution in the free troposphere, air mass trajectories were examined at three locations on the flight leg along 140°E. Figure 3 shows the results of the trajectory analyses on the isobaric surfaces of 700 mbar, in which stippled bands indicate trajectories for five days. In the northern part of the flight leg the air mass abundant in coarse particles was transported from the Asian continent by a westerly wind and then went south of the anticyclone over Japan, whereas a clean air mass in the southern part of the flight leg was transported from the central Pacific Ocean by an easterly wind.

Vertical air motions were not taken into account in these analyses, because atmospheric conditions were nearly barotropic and the trajectories for different levels were closely similar to each other. The baroclinity of the atmosphere is very weak in the northwestern Pacific region in this period. It is certain that the air mass origin in the northern part of the flight leg is quite different from that in the southern part. In order to see the synoptic structure of the atmosphere when the observed air mass was lying over the Asian continent, we show in the same figure (Figure 3) the weather chart of 700 mbar at 1200 UT (2100 LST) on August 3. An ascending air motion was located at the east side of the trough as shown by the Beaufort wind scales. This ascending air mass may have brought soil-derived coarse particles to the northern part of the northwestern Pacific Ocean. Dust storms over the Asian continent have been reported even in summer, although their frequency is rather low [*Zhang*,

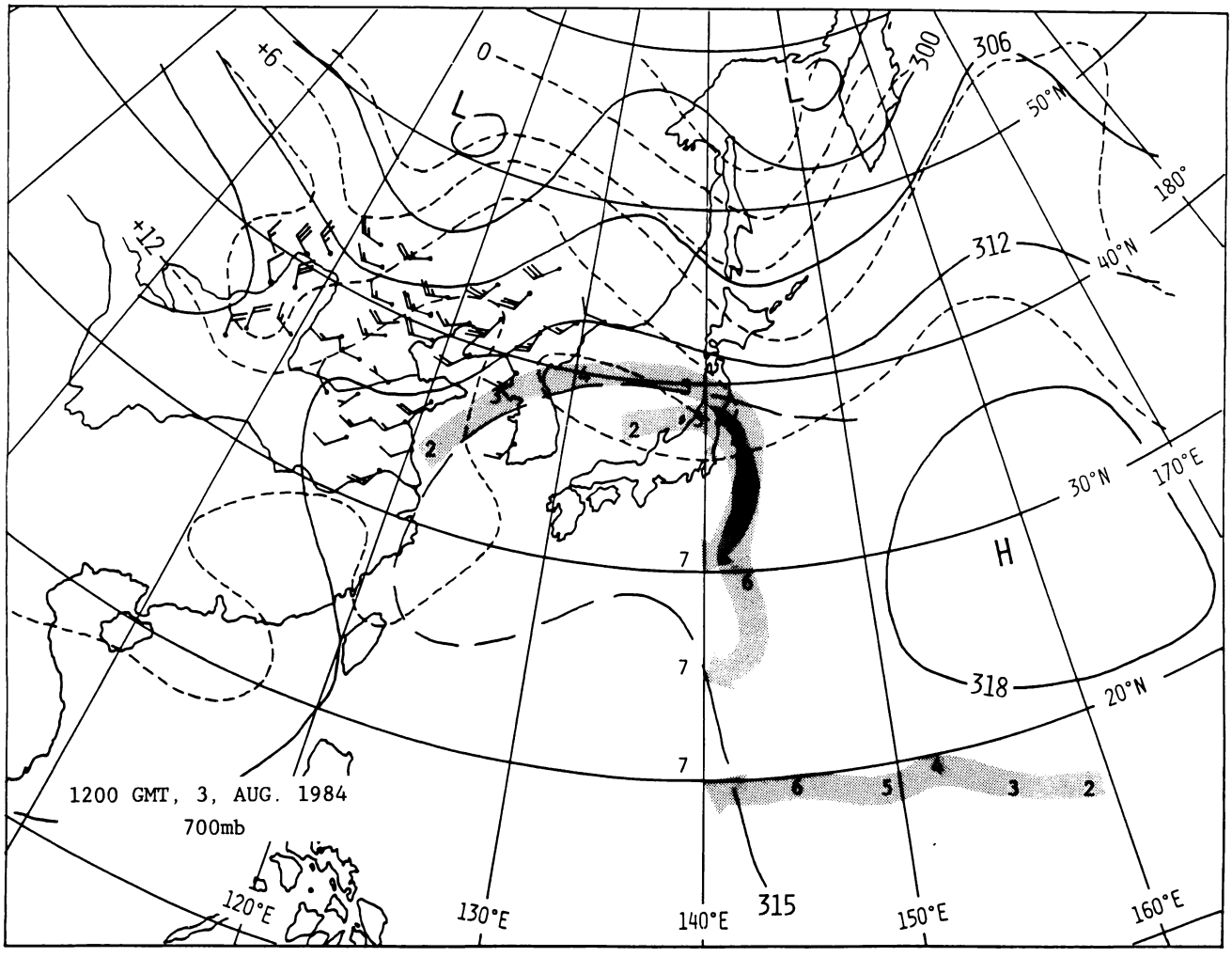

Fig. 3. Isobaric trajectories at 700 mbar (stippled bands) for the period of August 2–7, 1984, and weather chart of 700 mbar on August 3, 1984. Numbers on the stippled bands indicate daily air mass positions.

1984; *Merrill et al.*, 1989], and aerosols originating from the Asian continent have also been observed in Hawaii in all seasons [*Parrington et al.*, 1983]. Occasionally, quite large particles with radii $r > 35$ μm have been observed in the central North Pacific and attributed to long-range transport from the Asian continent [*Betzer et al.*, 1988]. Removal of aerosols by gravitational fallout is not always vital for such transport processes of coarse particles, because the fallout velocity of coarse particles with typical radii $r = 4 \sim 5$ μm is about 300 ~ 400 m/d [*Kasten*, 1968]. On the other hand, it is difficult to ascribe the observed coarse particles to sea-salt particles injected into the free troposphere. The top of the planetary boundary layer along the cruising course was found below 700 mbar on August 7 and 8 from radiosonde observations at Hachijojima (33°06′N, 139°47′E) and Chichijima (27°05′N, 142°11′E) weather stations of the Japan Meteorological Agency, and coarse particles were very scarce within the planetary boundary layer around latitudes from 20°N to 25°N on these days. Thus the coarse particle mode seen in Figure 2 originated from the Asian continent, whereas the accumulation mode originated from substances characteristic in the urban industrial area along the coast of Japan facing the Pacific Ocean.

In the wintertime we have observed two different volume spectra as shown in Figures 4*a* and 4*b*. The volume spectra abundant in coarse particles were retrieved on February 16, 1984, at high altitudes in the latitudes from 29.6°N to 34.5°N along 135°E. Figure 4*a* shows the volume spectra of columnar aerosols observed at 560 and 480 mbar by solid and dashed lines, respectively. From the difference between these two lines it is evident that the atmospheric layer between 560 and 480 mbar contained aerosols abundant in coarse particles while fairly monomodal stratospheric aerosols due to the El Chichon eruption prevailed in the layer above 480 mbar. The size distribution of this coarse particle mode is fairly similar to that of the Yellow Sand (the eastern Asia dust storm) observed in spring at Nagasaki, Japan, except that the concentration is lower by 1 order of magnitude [*Nakajima et al.*, 1989; *Tanaka et al.*, 1989]. Two stippled bands in Figure 5 show isobaric trajectories for two days composed from 500- and 700-mbar weather charts, respectively, which suggest that the air mass holding coarse particles was transported again from the Asian continent. It is found that the two trajectories differ little, that is, the air mass in the lower level came from farther south than that in the upper level. In this period, strong baroclinity of the atmosphere was observed over eastern Asia from the weather charts of 850, 700, and 500 mbar, so that we must

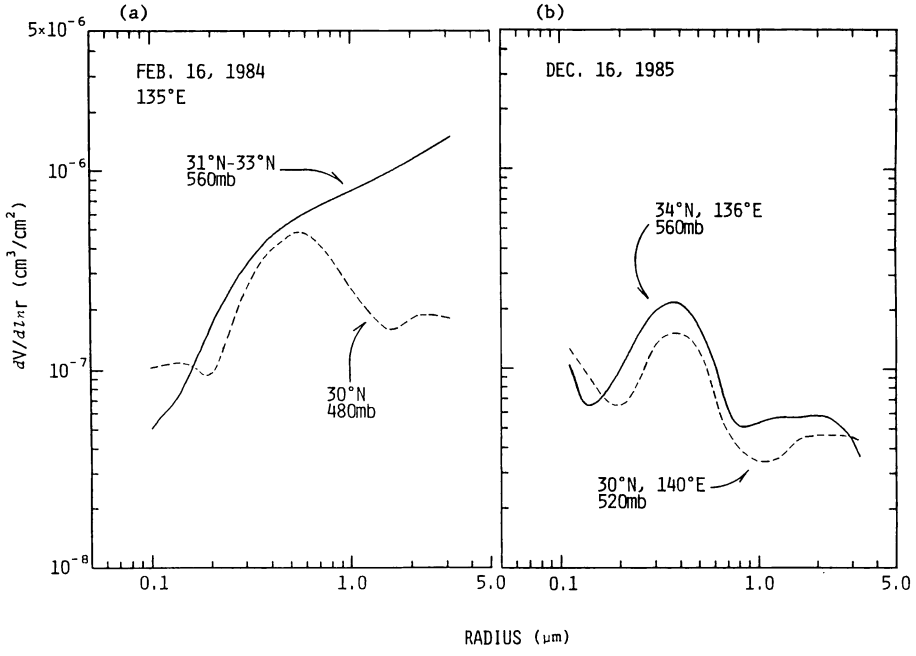

Fig. 4. (*a*) Volume spectra of columnar aerosols observed at 560 mbar (solid line) and 480 mbar (dashed line) on February 16, 1984, and (*b*) those at 560 mbar (solid line) and 520 mbar (dashed line) on December 16, 1985.

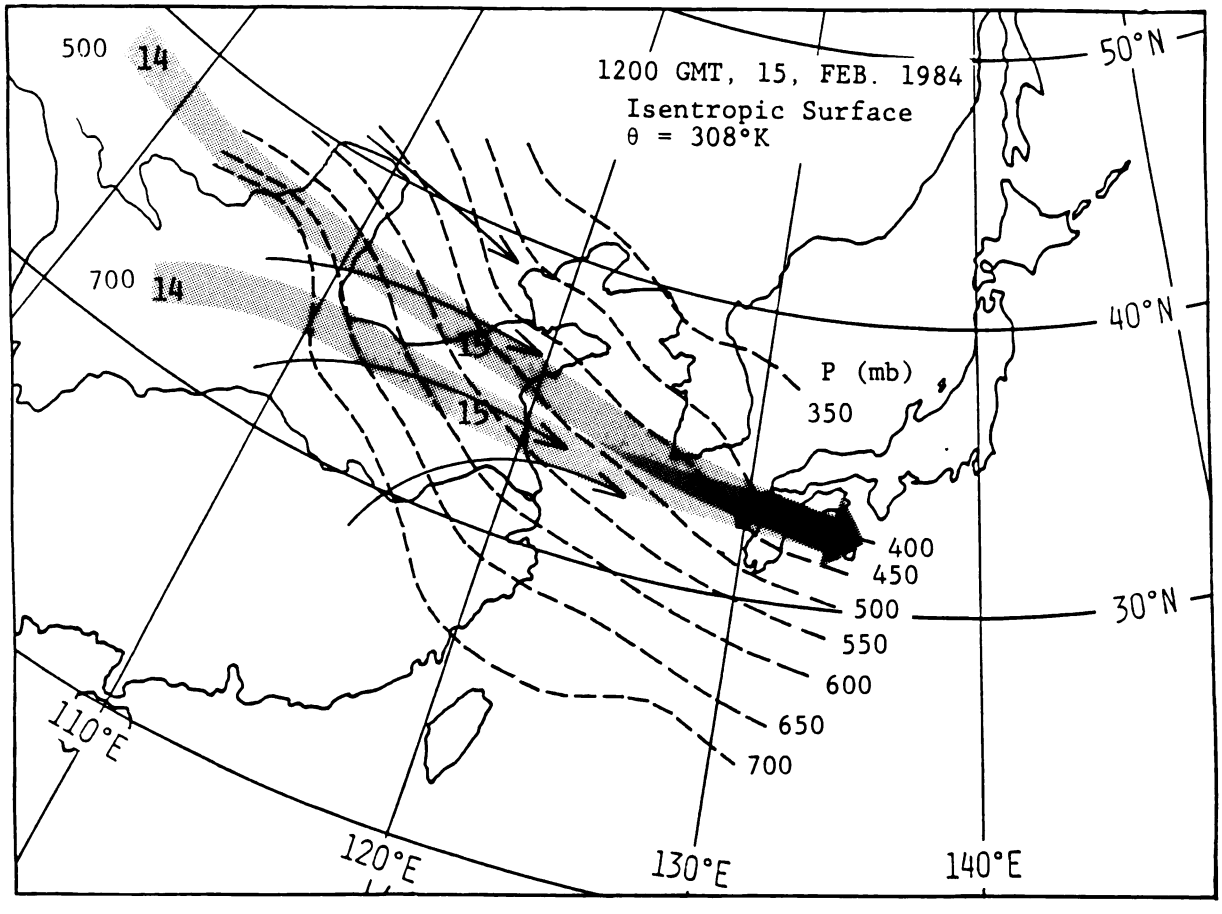

Fig. 5. Isobaric trajectories at 700 and 500 mbar (stippled bands) for the period of February 14–16, 1984, and the isentropic surface of 308°K shown by isobaric contours (dashed lines) on February 15, 1984. Solid lines with an arrow indicate air mass motions on that surface estimated from radiosonde data and the weather charts of 700 and 500 mbar. Numbers on the trajectories (stippled bands) indicate daily air mass positions.

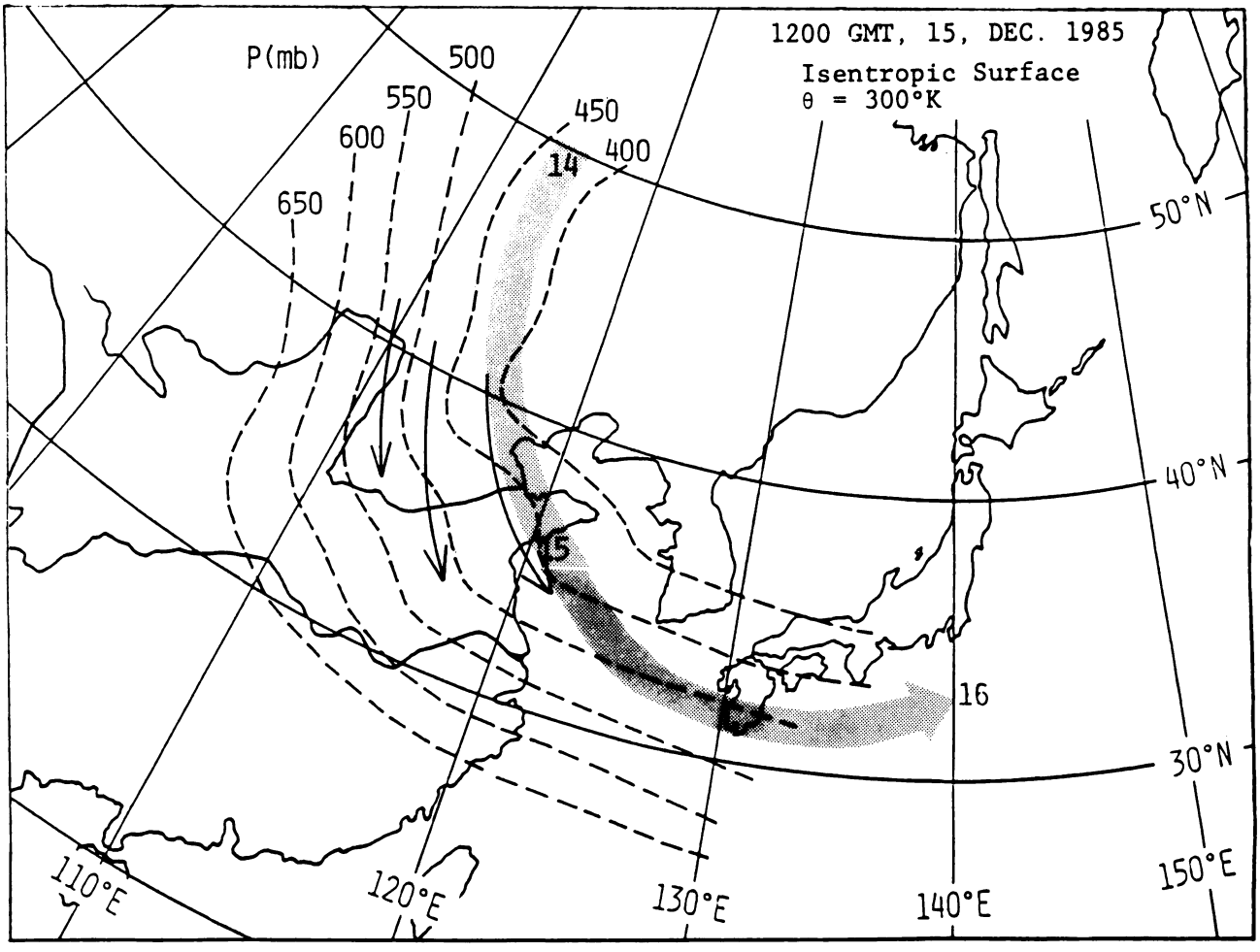

Fig. 6. The isobaric trajectory at 500 mbar (stippled band) for the period of December 14–16, 1985, and the isentropic surface of 300°K shown by isobaric contours (dashed lines) on December 15, 1985. Solid lines with an arrow indicate air mass motions on that surface estimated from radiosonde data and the weather chart of 700 mbar. Numbers on the trajectory (stippled band) indicate daily air mass positions.

take this effect into account. The 308°K isentropic surface, which is the typical potential temperature of the observed air mass, is also shown in Figure 5 by isobaric (dashed) contour lines for 1200 UT (2100 LST) on February 15, indicating a gradient of atmospheric stratification from southwest to northeast. Ascending motions of the air mass on this isentropic surface were inferred from the wind data of 700- and 500-mbar isobaric surfaces, as illustrated by solid lines with an arrow.

In contrast with the above results, no coarse particles were observed at both flight levels of 560 and 520 mbar on December 16 and 17, 1985, as shown in Figure 4*b*. Observational flights were carried out from 28°N to 34.5°N along the longitude of 140°E, but no obvious latitudinal dependence was found on the loading and the size distribution of aerosols. The shape of the monomodal volume spectrum was quite similar to that observed at the highest flight levels on February 16, 1984, and is ascribed to the stratospheric aerosols. The 500-mbar isobaric trajectory, shown by a stippled band in Figure 6, indicates that the observed air mass came from Siberia, the eastern part of the USSR. A large cyclone stayed over Sakhalin for several days around the observational period. A wind behind the cyclone from northwest to southeast is generally accompanied by de-

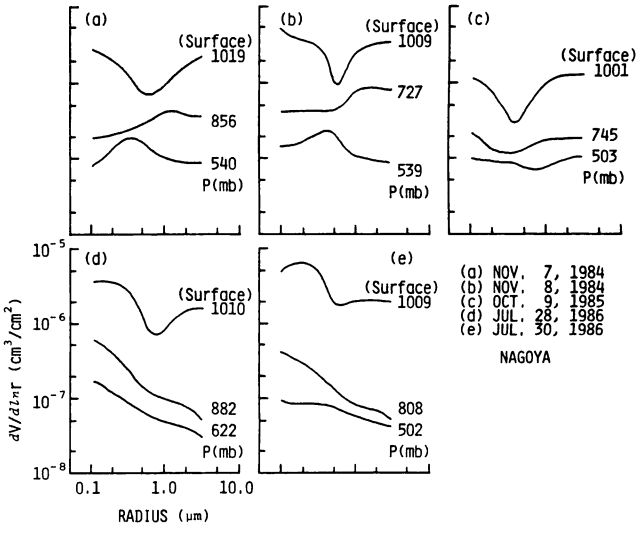

Fig. 7. Volume spectra of columnar aerosols observed at the ground surface, an altitude just above the planetary boundary layer, and the uppermost flight level over Nagoya on (*a*) November 7, 1984, (*b*) November 8, 1984, (*c*) October 9, 1985, (*d*) July 28, 1986 and (*e*) July 30, 1986.

scending air motion in the northern hemisphere because of the baroclinity of the atmosphere. Under this condition, therefore, few coarse particles are contained in the atmosphere even though the air mass passed over the arid region of eastern Asia. Figure 6 also shows the isentropic surface of 300°K at 1200 UT (2100 LST) on December 15, 1985, by isobaric (dashed) contour lines and the air mass motion at the surface by solid lines with an arrow which certainly indicate a prevalence of descending air motion.

4.2. *Results From Observation Over Nagoya*

Measurements of the solar radiation were carried out over Nagoya, paying attention to the vertical stratification of tropospheric aerosols. The volume spectra of aerosols in air columns above the ground surface, the planetary boundary layer (mixed layer), and the maximum flight level are shown in Figure 7 for observations in autumn and summer. Typical bimodal volume spectra with saddle point radii of about 0.5 μm prevailed in the lowest layer. The accumulation mode of the planetary boundary layer aerosols consists mainly of the secondary particles such as sulfate and nitrate produced through gas-to-particle conversion and coagulation processes, while the coarse particle mode consists mainly of soil-derived aerosols [*Whitby*, 1978]. In the summer (July 28 and 30, 1986), bimodal volume spectra dominated by the accumulation mode generally prevailed in the planetary boundary layer, while above the planetary boundary layer such bimodal volume spectra disappeared because of the disappearance of the coarse particle mode, and power law type spectra dominated by small particles instead prevailed. In the autumn (November 7 and 8, 1984, and October 9, 1985), in contrast, the accumulation mode disappeared, and a Jungelike power law spectrum ($dV/d \ln r$ = const) predominated above the planetary boundary layer. At the uppermost level the influence of volcanic aerosols due to the El Chichon eruption was clearly detected around a mode radius of 0.4 μm until the autumn of 1984, which decreased in concentration gradually during the observation period [*Nakajima et al.*, 1986].

Differences in the vertical stratification of aerosols observed over Nagoya, as shown in Figure 7, are generally ascribed not only to local characteristics of the urban atmosphere and the ground surface but also to synoptic scale conditions of the atmosphere. As a result of trajectory analyses for November 4–7, 1984, and October 6–9, 1985, strong westerlies were found for these periods. It is therefore reasonable to ascribe the source of the free tropospheric aerosols in these periods to the Asian continent. On the other hand, the 700-mbar chart on July 28, 1986, indicates that an anticyclone was located around the center of Japan. Similar situations were also found from the surface to the 500-mbar level and continued for several days. Since the pressure gradient was fairly small, the horizontal transport of aerosols due to a large-scale wind system was not so strong in this period. It is reasonable to speculate that the aerosols in the planetary boundary layer observed in the summer season were made from substances of ground surface origin (both soil particles and gases) and were spread throughout the planetary boundary layer through enhanced thermal convection. Since the development of the convective layer usually occurs by taking an air mass from the upper stable layer into the convective layer, transport of substances from the planetary boundary layer to the free troposphere is highly restricted, except for extremely unstable conditions accompanied by penetrative convection. A rapid decrease of the aerosol concentration beyond the top of the planetary boundary layer thus occurs.

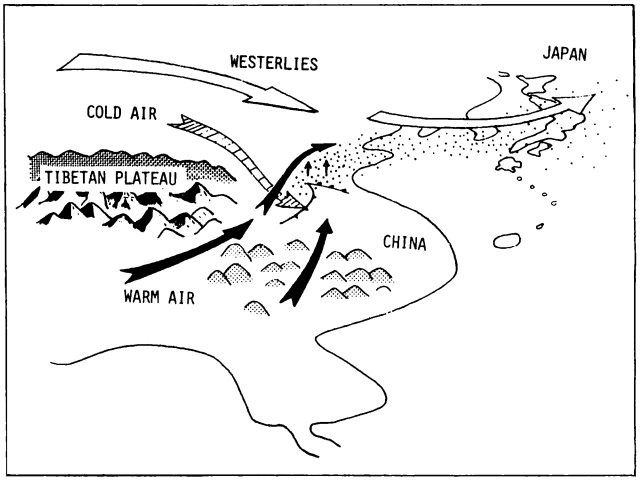

Fig. 8. Schematic illustration of the disturbance due to frontogenesis to the east of the Tibetan Plateau and transport of aerosols to Japan and the northwestern Pacific Ocean.

5. DISCUSSION

A well-known example of the transport of soil-derived dust particles to the northwestern Pacific Ocean is the Yellow Sand event in springtime (so-called Kosa in Japan). The dust storm moves generally with a cold front, and a large area of eastern China and Japan is covered by the heavy dust deposition [*Hirose et al.*, 1983; *Tanaka et al.*, 1989]. From lidar observations, *Iwasaka et al.* [1983] found two layers of dust, at 0.5–2.5 km within the planetary boundary layer and 4–8 km in the free troposphere, and ascribed the lower layer to the Yellow Sand dust transported from the basin along the Yellow River. Therefore the Yellow Sand event probably occurs when the cold surge to the east of the Tibetan Plateau is strongly enhanced and a cold front reaches around Japan.

Such a mechanism of the Yellow Sand event is considered to be different from that of coarse particles in the higher free troposphere we observed. From the weather charts at 1200 UT (2100 LST) on February 15, corresponding to the case shown in Figure 4*a*, strong baroclinity of the atmosphere was found in the arid region in the upper reaches of the Yellow River to the east of the Tibetan Plateau. A frontal surface was inferred to be in this area, where the cold air mass from the northwest was descending under the ascending warm air from the southwest. Such a weather condition is thought to be important for the generation and spreading of the coarse particle aerosols into the free troposphere, as pointed out by *Merrill et al.* [1985]. Soil-derived particles are first generated by enhanced turbulent motions and then transported to Japan and the Pacific Ocean by westerlies. This mechanism is shown schematically in Figure 8. The area to the east of the Tibetan Plateau has a potential to create disturbances at any time of the year due to confluence of a cold air mass from the northwest with a warm air mass from the southwest which have previously been divided by

the Tibetan Plateau. This is because the westerlies generally prevail throughout the year and there exists a strong baroclinity in the mid-latitude continental region of the northern hemisphere [*Holton*, 1979]. On the other hand, the source area of dust particles extends from the Takla-Makan desert to the basin along the middle course of the Yellow River all year round [*Zhang*, 1984]. Therefore soil materials may be transported vertically not only in the spring dust storm season but also whenever conditions such as aridity, lack of vegetation on the surface, low moisture content, and baroclinity of the atmosphere are satisfied.

An alternative coarse particle source is the arid region in western China, of which the Takla-Makan desert is a part. However, the observed air mass with the 308°K potential temperature was found over the Takla-Makan desert at the same level (at about 500 mbar) as over Japan. Furthermore, no ascending air motion was found on the weather chart, and a strong thermal convection to elevate the desert aerosols into the free troposphere such as in the Saharan desert is not expected in this season in mid-latitudes. Therefore it is difficult to consider the Takla-Makan desert as a source area of coarse particles observed on February 16.

A similar mechanism of injecting soil-derived aerosols into the free troposphere was revealed by *Reiff et al.* [1986] in the transport process of Saharan dust to Europe from trajectory and satellite imagery analyses. Aerosols are lifted on the isentropic surface of a baroclinic atmosphere in front of the cyclone over the northeastern Atlantic Ocean. This mechanism is consistent with the results of *Fitch and Cress* [1981, 1983], who performed numerous airborne measurements of aerosols over Europe and found that coarse particles varied greatly in concentration while accumulation mode particles were generally scarce above the planetary boundary layer. They suggested that such coarse particle aerosols are transported from the Saharan desert area.

As described above, the clean free troposphere is often disturbed by soil-derived particles, especially in mid-latitudes. The atmosphere has strong baroclinity over the mid-latitude continent, so that coarse particles are carried up to the higher troposphere of 5 ~ 6 km. Once aerosols are injected into the free troposphere, they continue to disperse over a wide region by the prevailing wind system until the dry or wet removal mechanism operates.

6. Concluding Remarks

We have carried out airborne measurements of solar aureole intensities with the spectral scanning radiometer for the purpose of investigating aerosol size distributions and their spatial structures. Horizontal and vertical variations of aerosol optical thickness and volume spectra in the troposphere around Japan and the northwestern Pacific Ocean were revealed by inverting the measured solar aureole intensities. Synoptic analyses with air mass trajectories, isentropic surfaces, and weather charts were also performed in order to interpret the obtained results. Conclusions of the present study are summarized as follows:

1. Bimodal volume spectra of aerosols with a saddle point radius around 0.5 μm, i.e., the accumulation and coarse particle modes, are present in the troposphere. The accumulation mode dominates the coarse particle mode within the planetary boundary layer, especially in summer, whereas coarse particles frequently prevail in the free troposphere above the planetary boundary layer throughout the year.

2. It is suggested that submicrometer particles present in the lower troposphere around Japan are generated from substances characteristic in the urban industrial area by the gas-to-particle conversion process. On the other hand, coarse particles in the free troposphere are estimated to originate from an arid region of the Asian continent and to be transported to the northwestern Pacific Ocean.

3. The Tibetan Plateau seems to play an important role for the injection of soil-derived coarse particles into the free troposphere. A cold air mass from the northwest and a warm air mass from the southwest divided previously by the Tibetan Plateau join at an arid region to the east of the Tibetan Plateau. There exists a strong baroclinity in the mid-latitude continent, so that a disturbance occurs and soil-derived particles are blown upward. Coarse particles thus generated are carried up to a higher level of the free troposphere by ascending motion on the stream from the southwest to the northeast and then transported to the northwestern Pacific Ocean by westerlies.

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