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Variations of tropospheric ozone concentration over Syowa Station, Antarctica

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ABSTRACT

Aircraft measurements of the tropospheric O_3 concentration were made over Syowa Station (69°00′S, 39°35′E), Antarctica from May 1989 to January 1990, except during the polar night months of June and July. The O_3 concentration increased with height for the whole period of the measurements. Lower tropospheric O_3 showed a prominent seasonal variation with maximum concentration in winter and minimum concentration in summer, which is very close to the result of continuous O_3 measurements initiated at Syowa Station in February 1988. In the upper troposphere, the concentration reached high values in winter, decreased gradually until late spring and then increased again. As a result, the height-dependent difference of the concentration was reduced from late autumn to early spring and enhanced in remaining seasons of the year. The seasonal variation of upper tropospheric O_3 was similar to those of the lower stratospheric O_3 concentration measured by ozonesondes and the total amount of O_3 by a Dobson spectrophotometer over Syowa Station. Inspection of TOMS (Total Ozone Mapping Spectrometer) data also suggested that the temporal variations of the upper tropospheric O_3 measured in this study are closely related to those of the lower stratospheric O_3 over Syowa Station. Comparison of the results of surface O_3 measurements in the southern hemisphere showed that the concentration increase going southward, and appearance of the minimum concentration of the seasonal variation is delayed by almost 1 month at the South Pole compared to Syowa Station and Cape Grim, Tasmania. Taking into account these results, it is suggested that the intrusion of stratospheric O_3 into the troposphere occurs over Syowa Station throughout the year, and that air with low O_3 concentrations is transported from subpolar or middle latitudes to the station through the lower troposphere from spring to early autumn, in addition to the photochemical destruction of O_3 near the station.

1. Introduction

Atmospheric O_3 is one of the most important trace gases in determining the radiation budget (Fishman et al., 1979a) and is a precursor for highly reactive radicals (Levy, 1971; Crutzen, 1988). Its concentration variations, due to both natural and anthropogenic causes, will therefore lead not only to changes in the chemical cycles of many gases such as CO, CH_4, CH_3Cl and H_2S but also to global climate change.

Measurements of tropospheric O_3 in Antarctica have been made by several researchers to elucidate its temporal and spatial variations (Wexler et al., 1960; Aldaz, 1965; Oltmans and Komhyr, 1976, 1986; Oltmans, 1981). The tropospheric O_3 variations are determined by many factors such as intrusion of stratospheric air with rich O_3 into the troposphere, transport of air with different O_3 concentrations from different places and destruction at the earth's surface. Recently, photochemical production and destruction of O_3 in the troposphere were also proposed to be important causes of these variations (e.g., Fishman et al., 1979b; Crutzen, 1988). It is, however, thought that the photochemical production of O_3 scarcely occurs in the antarctic region, because the air is fairly free from influence of the human activities. Tropospheric O_3 variations in the antarctic region have been interpreted in terms of air transport

process (Wexler et al., 1960; Oltmans and Komhyr, 1976, 1986; Galbally, 1981; Hogan, 1981, 1986; Oltmans, 1981; Robinson et al., 1983, 1984; Komhyr et al., 1988; Janach, 1989), but they are not fully understood yet. Further studies are therefore required for better understanding of the behavior of the tropospheric O_3 concentration in this region.

We initiated continuous measurements of surface O_3 concentration at Syowa Station (SYO; 69°00′S, 39°35′E), Antarctica in February 1988. In addition to this, we also made aircraft measurements of the tropospheric O_3 concentration over the station from May 1989 to January 1990, except during the polar night months of June and July.

In this paper, the results of our aircraft measurements are given, primarily in terms of height-dependent seasonal variations of the tropospheric O_3 concentration. Our data are also compared with measurements from ozonesondes and the Dobson spectrophotometer at SYO, and continuous measurements at SYO, the South Pole, Samoa (Oltmans and Komhyr, 1986) and Cape Grim, Tasmania (Douglas et al., 1985) and TOMS data from the Nimbus 7 satellite. Causes of temporal and spatial variations of the tropospheric O_3 concentration over the antarctic region are also discussed.

2. Measurements

The tropospheric O_3 concentrations were measured by using an ultraviolet photometer (Dasibi Model 1006-AHJ) mounted on a Pilatus Poter PC-6 aircraft. 18 flights were made over SYO for the period from May 1989 to January 1990 except in June and July when aircraft operation was difficult due to darkness. SYO is located on East Ongul Island in Lützow-Holm Bay, away from the Antarctic Continent by 4 km, as shown in Fig. 1. The surface of the island is composed of naked rock and is covered with snow except in summer (Nakazawa et al., 1991). The sea between the island and the continent was frozen during the period of this measurement.

The aircraft ascended to its maximum altitude of approximately 7 km and then descended, repeating a level flight for about 5 min at every 300 m height intervals. Ambient air was brought into the cabin from the wingtip through a Teflon tube (7 m in length, 6 mm in inner diameter), to avoid contamination from the engine exhaust. Atmospheric aerosols were removed by a Teflon filter attached between the O_3 meter and the tube. It was confirmed by laboratory test that O_3 loss in this air inlet system is negligibly small. The output of the O_3 meter, together with temperature of sample air in the cell of the meter, was sampled every 12 s during the 5 min-level flight and recorded digitally on printing paper as well as on a stripchart serving as back-up. The pressures and temperatures of ambient air were also monitored for determination of the heights at which the measurements were made. All instruments for these parameters were set up in a box kept at ~25°C by a temperature controller. The concentration values measured by the O_3 meter were adjusted to the condition under which the O_3 meter was calibrated, using the ideal gas law with temperatures and pressures of the sample air.

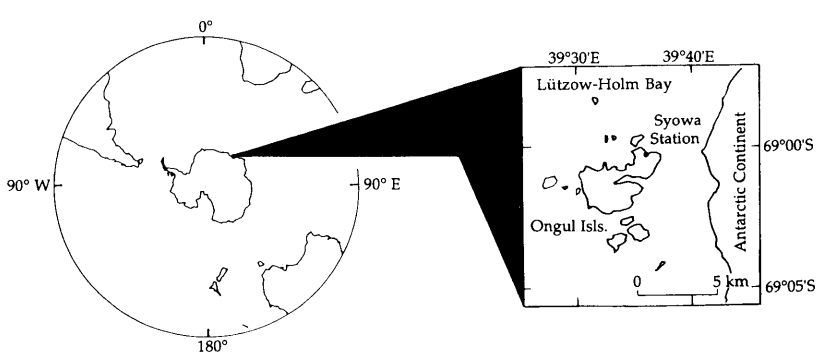

Fig. 1. Map of the Antarctic Continent and location of Syowa Station.

At SYO, two identical O_3 meters were used, in turn, for continuous O_3 measurements of 1 year each. The meter was returned to Japan for overhaul and output calibration after 1 year and of use then shipped to the station again. During the measurements at SYO, the zero offset of the meter output was checked every 10 days by using the air passed through O_3 scrubbers. All O_3 data presented in this paper were corrected for this effect. When the meter was replaced by the new one, both meters were operated simultaneously for 1 or 2 weeks for intercalibration. Results from the meters agreed with each other within 1 ppbv (parts per billion by volume). One of the two meters was used for the aircraft measurements.

Since details of the calibration procedures for the O_3 meters will be given elsewhere, only its outline is described here. A gas-phase titration (GPT) method was employed in this study. A chemiluminescent NO_x analyzer was first calibrated by using the GPT system in which NO standard gas with a concentration of 100 ppmv (parts per million by volume) was diluted to 200 ppbv by mixing with purified air. Then, an O_3 generator installed in the GPT system was calibrated by measuring the NO concentration in mixtures of the diluted standard gas and O_3 from the generator. When O_3 is added to the air with excess NO, NO decreases by a quantity equal to that of added O_3. After these procedures, the O_3 meter was calibrated by using the O_3 generator.

3. Results and discussion

The O_3 concentrations measured on 6 May and 23 December 1989 are shown in Fig. 2, as examples of the vertical O_3 profile over SYO. Fig. 2a represents the profile observed typically from late autumn to early spring. The vertical gradient of the concentration is gentle, since the lower tropospheric O_3 reaches high values in this period of the year, as mentioned below. On the other hand, during the period from late spring to summer when low and high values of the O_3 concentration appear in the lower and upper troposphere, respectively, the O_3 concentration increases gradually in the lower troposphere and rapidly in the upper troposphere with increasing height, as shown in Fig. 2b. The vertical gradient of the concentration is therefore steep for this period, especially for heights higher than 4 km.

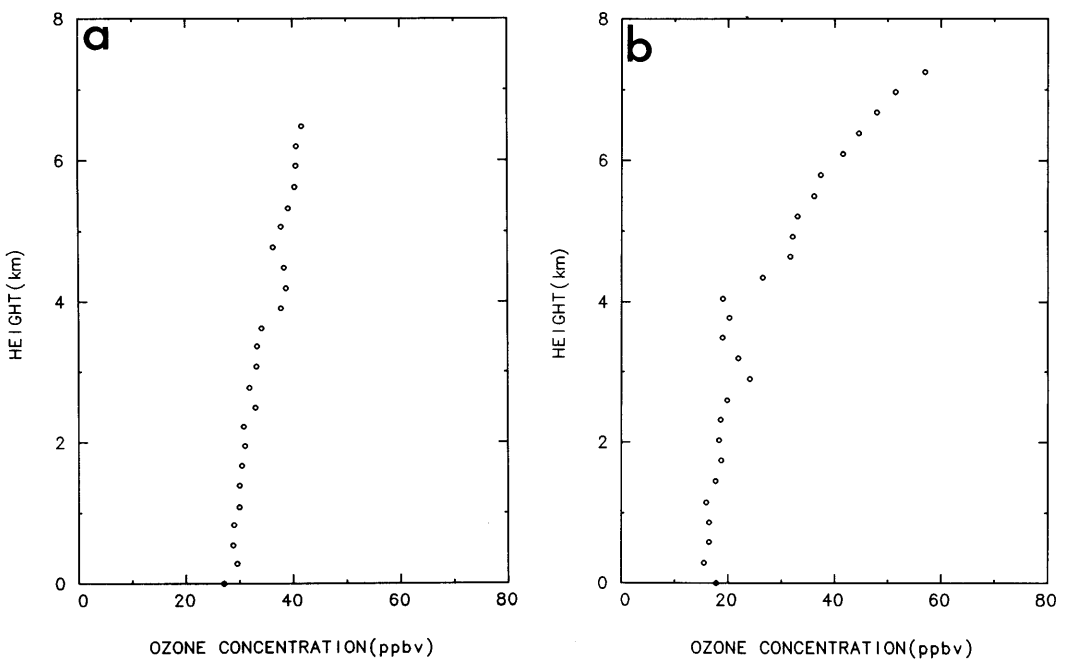

Fig. 2. Vertical profiles of the O_3 concentration over Syowa Station of (a) 6 May 1989 and (b) 23 December 1989.

Since inspection of the aerological data over SYO showed that all O_3 data were obtained in the troposphere, they were first grouped into 5 height sampling intervals of 0–2, 2–4, 4–6, 6.0–6.5 km and above 6.5 km. The data classified into respective intervals were then simply averaged. Here, the data taken at heights higher than 6.0 km were divided into two intervals because the aircraft used did not always reach altitudes higher than 6.5 km. The O_3 concentrations thus obtained are shown in Fig. 3, together with the daily mean values from continuous surface O_3 measurements at SYO for the period of February 1989–January 1990 (Aoki et al., unpublished data). Since the diurnal variation of surface O_3 is hardly observable in the antarctic region (Oltmans and Komhyr, 1976; Oltmans, 1981; Aoki et al., unpublished data), the measured concentrations are probably independent of time of the day when measurements are made.

Except for a few cases, the O_3 concentration increases with increasing height for the whole period of the measurements. In the lower troposphere, the concentration shows a prominent seasonal variation with maximum in winter and minimum in summer, which is very close to the results of surface O_3 measurements at SYO, as seen in Fig. 3. On the other hand, the upper tropospheric O_3 concentration shows high values in winter, decreases gradually from September to early November and then increases. As a result, the height-dependent difference of the concentration is reduced from late autumn to early spring and enhanced in remaining seasons of the year. In the height interval of 4–6 km, the seasonal variation is almost in phase with that in the lower troposphere, but the O_3 concentration is rather constant from spring to summer when the concentration decreases and increases in the lower and upper troposphere, respectively. Peak-to-peak amplitude of the seasonal variation is therefore smaller than those of other height intervals, implying that variations of the middle tropospheric O_3 are affected significantly by those in the upper and lower troposphere. It can also be seen in Fig. 3 that variabilities of the O_3 concentration are much larger in the upper troposphere than in the lower troposphere.

Fig. 4 shows a cross-sectional representation of the O_3 concentration up to 8 km over SYO, which was drawn from the results of our aircraft measurements and ozonesonde observations made

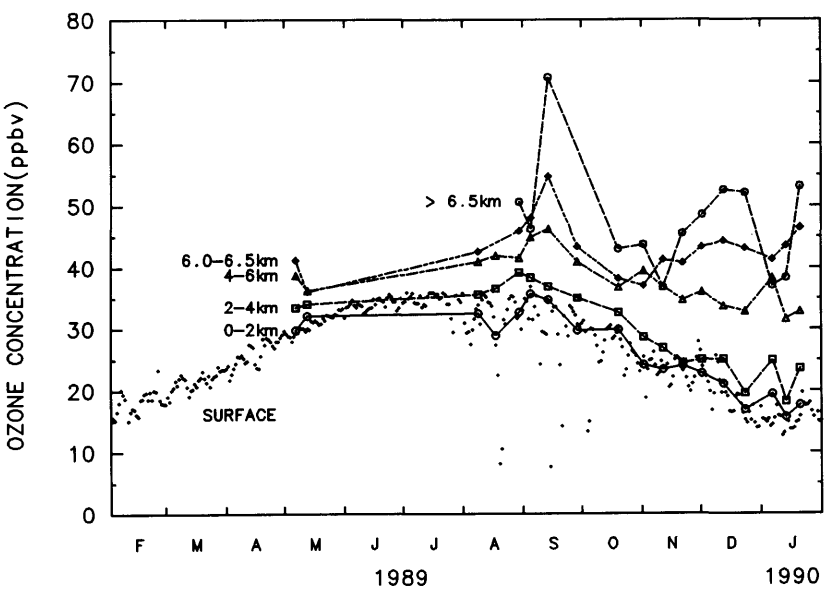

Fig. 3. Variations of the tropospheric O_3 concentration obtained by aircraft measurements over Syowa Station from May 1989 to January 1990 and daily mean values of surface O_3 concentration at the Station from February 1989 to January 1990.

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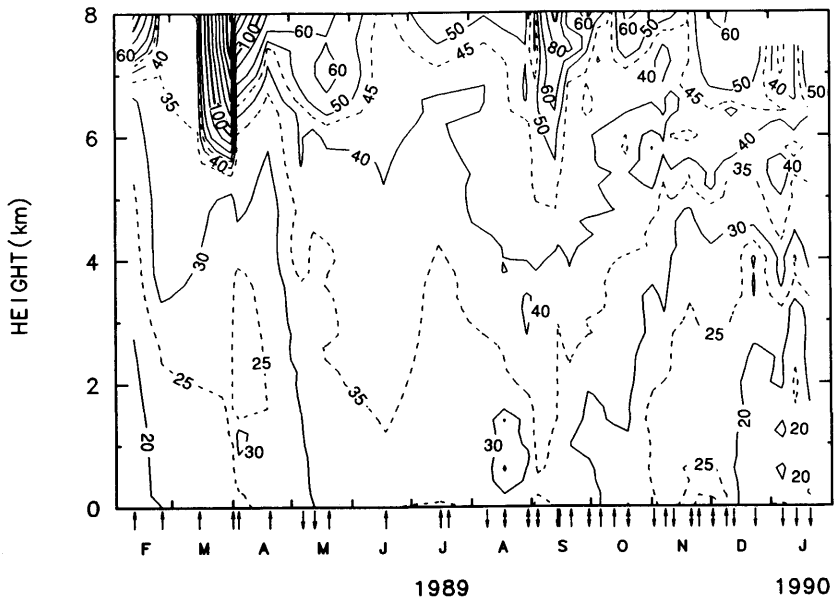

Fig. 4. Distribution of the O_3 concentration (ppbv) over Syowa Station from February 1989 to January 1990, drawn from the results of aircraft measurements and ozonesonde observations. The dates when ozonesonde observations and aircraft measurements were made are indicated by upward and downward arrows, respectively.

by the Japan Meteorological Agency. Comparison of respective concentrations measured by the Dasibi O_3 meter and ozonesondes from February 1988 to January 1990 revealed that the former are higher by 2.7 ppbv, on average, than the latter in a range of 10–60 ppbv. The result that the ozonesonde gives smaller values than the Dasibi O_3 meter was also reported by Chubachi (1985). However, this small concentration difference was not corrected in Fig. 4, because the tropospheric O_3 data from ozonesonde measurements are rather scattered from one sounding to the next, as compared with those from the O_3 meter. The characteristics of the tropospheric O_3 variations mentioned above are more clearly seen in this figure. It is also seen in this figure that remarkably high values of the O_3 concentration sometimes appear at higher than 6 km. These results imply that temporal and spatial variations of the O_3 concentration over SYO are determined by different processes for different seasons and heights.

We first examine the variations of the O_3 concentration in the upper levels. The tropopause heights measured by using rawinsondes and ozonesondes over SYO from January 1989 to January 1990 are plotted in Fig. 5. In general, the height of the first tropopause is low from summer to autumn and high from winter to spring, which is opposite to that in middle latitudes. The tropopause sometimes moves down to heights lower than 7.5 km, especially from late summer to autumn. Therefore, high values of the O_3 concentration found above 6 km during this period may be attributed to intrusion of stratospheric air which is richer in O_3 than tropospheric air or to the air affected strongly by it. In fact, the results of the ozonesonde observations show that heights of the first tropopause were 7.6, 5.6 and 7.2 km on 9 February, 31 March and 3 April 1989, respectively when the O_3 concentrations were very high in upper levels, as seen in Fig. 4. But, the O_3 concentrations in this height region were low on 23 February and 1 March 1989 when the tropopause was located at higher than 8 km.

On the other hand, the tropopause was scarcely observable at heights lower than 7.5 km in remaining seasons of the year. Nevertheless, high O_3 concentrations were found in the upper troposphere for example, in mid-September of

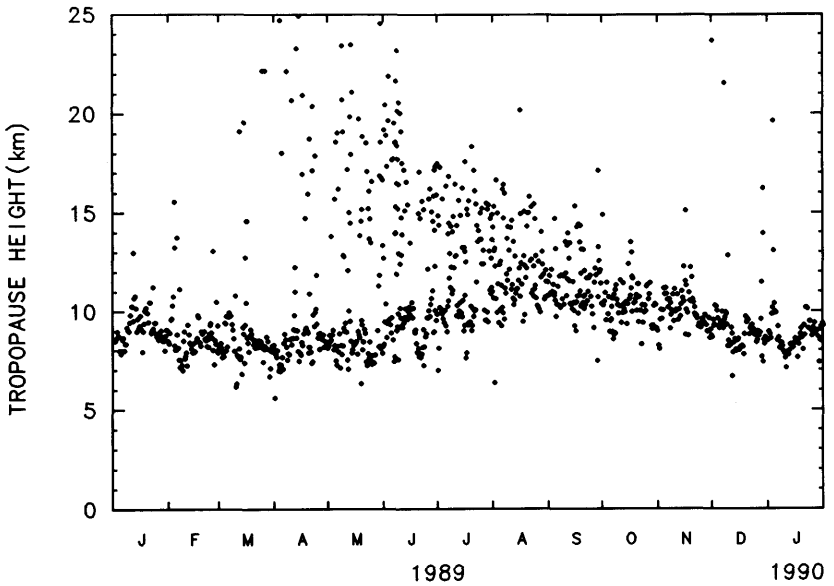

Fig. 5. Variation of the tropopause height over Syowa Station from January 1989 to January 1990.

1989 when the tropopause was always located at higher than 8.5 km. Therefore, we have to examine other processes for interpreting the variations of the upper tropospheric O_3 from winter to early summer. Fig. 6 is a cross-sectional representation of the O_3 concentration up to 30 km over SYO. This figure shows only ozonesonde data. At 10–20 km, the O_3 concentration decreases with time in September except for a transient high value in the lower level, it is rather constant with small fluctuations from early October to late November and then increases rapidly. Such a variation is also seen in the total amount of O_3 over SYO measured by the Dobson spectrophotometer, as shown in Fig. 7.

To elucidate a cause of variations of the total amount of O_3 during this period, we examined the TOMS data. The result showed that the total amount of O_3 over SYO decreases when SYO is located inside the ozone hole and increases rapidly with the disappearance of the hole in late spring. In this connection, the ozone hole usually shrinks and the station lies outside the hole early in November, but in 1989, the hole remained over SYO until mid-November. Inspection of the TOMS data also showed that high values of the total O_3 amount in mid-September are attributable to the fact that SYO was located temporarily outside the ozone hole by its southward movement. Taking account of the above finding, as well as the fact that the variations of the lower stratospheric O_3 and total amount of O_3 are similar to that of the upper tropospheric O_3 observed in our aircraft measurements, it is suggested that downward transport of stratospheric O_3 into the upper troposphere through the tropopause occurs rather quickly, at least over SYO. Horizontal transport of the stratospheric O_3 intruding into the troposphere in other polar or subpolar regions may also be partly responsible for the variations of the upper tropospheric O_3 over SYO.

As described above, the lower tropospheric O_3 concentration shows a prominent seasonal variation with a maximum in winter and a minimum in summer. This seasonal variation in the lower troposphere is out of phase with those of the total amount of O_3, the lower stratospheric O_3 and the upper tropospheric O_3, especially in the summer season. Therefore, it cannot be explained only by continuous downward transport of O_3 from the stratosphere to the ground throughout the year. To elucidate other processes important to variations of the lower tropospheric O_3 over SYO, we examined the seasonal variations derived

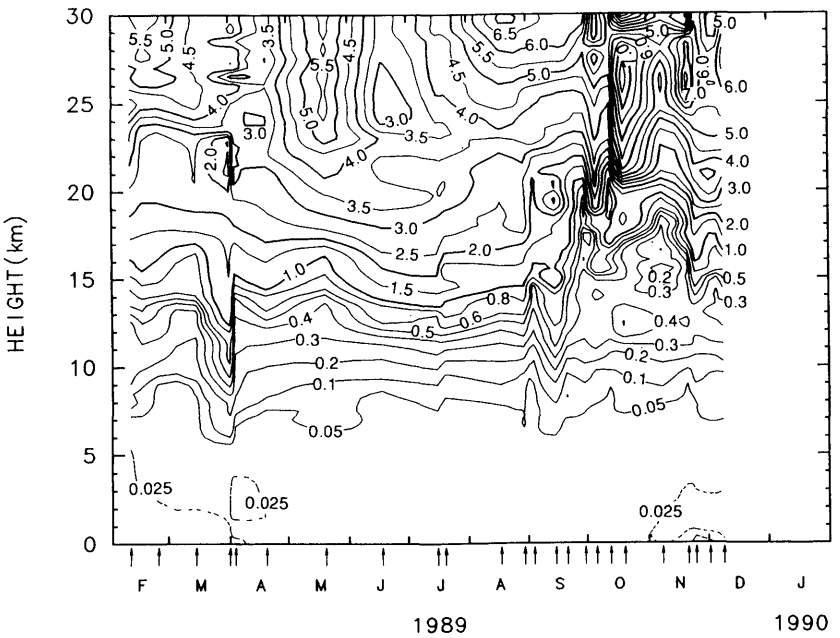

Fig. 6. Same as in Fig. 4 but for up up to 30 km. The concentration is expressed in ppmv.

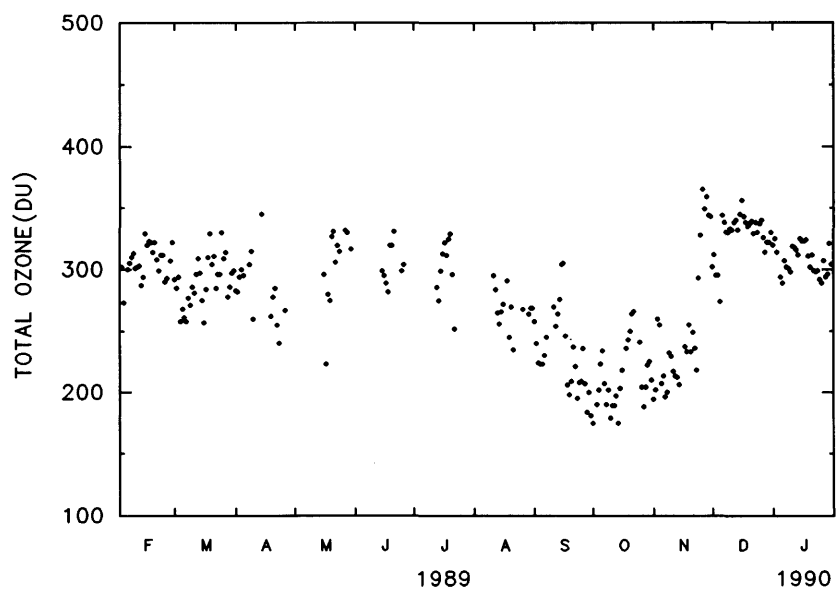

Fig. 7. Variations of the total amount of O_3 over Syowa Station measured by the Dobson spectrophotometer from February 1989 to January 1990. The amount of O_3 is expressed in Dobson Units.

from the surface O_3 measurements at SYO for February 1988–January 1990, the South Pole (SPO; 90°S, 25°W) for 1975–1986, Cape Grim (CGO; 41°S, 145°E) for 1982–1984, and Samoa (SMO; 14°S, 171°W) for 1976–1986, along with ozonesonde observations at the 700 hPa pressure level over SYO for 1977–1990 and our aircraft measurements at the 2–4 km height over SYO. Monthly mean O_3 concentrations were taken from Oltmans and Komhyr (1986) and the Atmospheric Environment Service (1987) for SPO and SMO, Douglas et al. (1985) for CGO, Japan Meteorological Agency (1982, 1987, 1988, 1989, 1990, private communication 1990) for ozonesonde observations. Values in each month of respective years were averaged. In this procedure, the O_3 concentrations from aircraft measurements were used to supplement the ozonesonde data after adjusting to the concentration scale of our Dasibi O_3 meter so that both results are compatible with each other. The O_3 concentrations thus obtained are shown in Fig. 8.

It is seen from this figure that the seasonal O_3 variations observed in the lower troposphere of the southern hemisphere are similar to each other and that the concentration increases with increasing latitude. The minimum concentration of the seasonal variation appears in January at SYO and CGO, but it is delayed by almost 1 month at SPO compared to that at SYO, as was found by Oltmans and Komhyr (1976) from a comparison of the results of continuous measurements at coastal and inland stations in the antarctic region. At 700 hPa level over SYO, corresponding approximately to the altitude of SPO, the O_3 concentrations appear to be slightly higher from autumn to early spring and lower in summer than those at SPO.

To see the details of the latitudinal distribution of the O_3 concentration in southern middle and high latitudes, seasonal differences of the O_3 concentration between the observational locations, calculated from the data plotted in Fig. 8, are shown in Fig. 9. The concentration difference between SPO and SYO largely disappears from early autumn to early spring and is enhanced in remaining seasons of the year. A similar variation is also seen in the relation between SPO and the 700 hPa level over SYO, but the concentration difference is mostly negative from early autumn to early spring, reflecting the fact that the O_3 concentration increases with increasing height over SYO throughout the year. The O_3 concentration at CGO approaches closely to that at SYO in early autumn, and the concentration difference between both stations is increased rapidly to the maximum in winter and then reduced gradually. As described above, the O_3 concentrations plotted in Figs. 8 and 9 were derived from the data taken for different periods for different measurements. Indeed, the seasonal O_3 variation is variable from year to year and the secular trend of the lower tropospheric

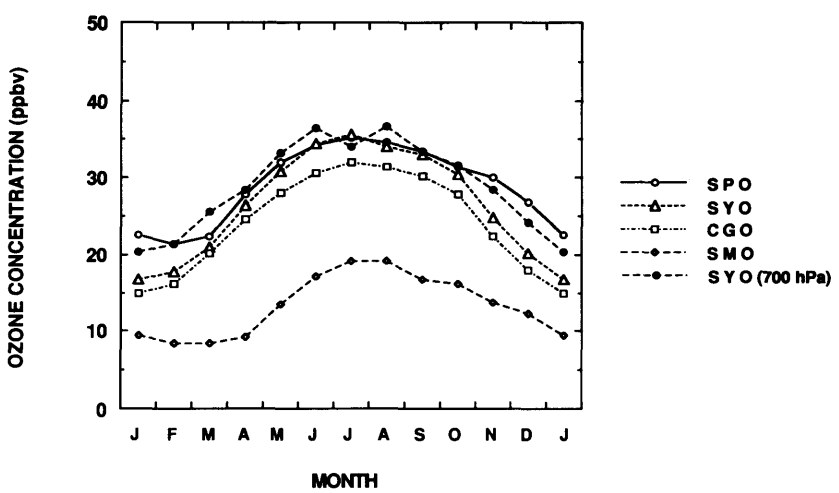

Fig. 8. Seasonal variations of the lower tropospheric O_3 concentration at the South Pole (SPO), Syowa (SYO), Cape Grim (CGO) and Samoa (SMO) and at 700 hPa pressure level over Syowa.

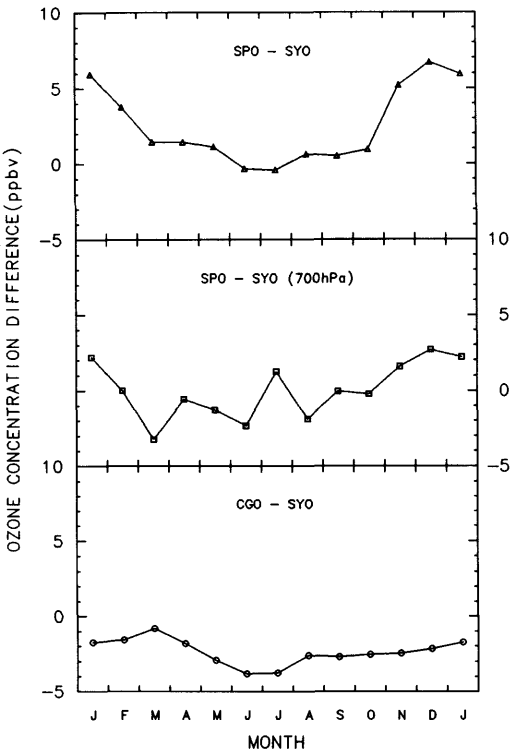

Fig. 9. Seasonal differences of the lower tropospheric O_3 concentration between the South Pole and Syowa (top), between the South Pole and 700 hPa pressure level over Syowa (middle) and between Cape Grim and Syowa (bottom).

O_3 concentration is also found (Oltmans and Komhyr, 1986; Komhyr et al., 1988). Moreover, the calibration method for the O_3 meter at SYO is different from the UV photometric method which is widely used at other locations. It is therefore impossible to make a strict comparison of the O_3 concentrations at different locations. Nevertheless, it may be deduced from above comparison that the lower tropospheric O_3 concentration is higher at SPO than at SYO from spring to summer, and that the O_3 concentration difference between SYO and CGO is enhanced especially in winter, since these differences are fairly large.

From these results, the following seasonally dependent process of air transport is thought to be one of the important factors for the variations of the lower tropospheric O_3 concentration in the antarctic region. From autumn to spring through winter, southward transport of the air with low O_3 concentrations from lower latitudes is suppressed, and the lower tropospheric O_3 concentration thus becomes high. Furthermore the height-dependent difference of the tropospheric O_3 is reduced due to continuous downward transport from the upper atmosphere with rich O_3. Consequently, the concentration difference between high and middle latitudes is increased during this period. From spring to autumn through summer, the air with low O_3 concentrations is transported from subpolar or middle latitudes into the antarctic region through the lower troposphere, in addition to downward transport of the upper tropospheric air. Since the former takes precedence over the latter, the lower tropospheric O_3 concentration is decreased. The vertical gradient of the tropospheric O_3 therefore becomes steep in the antarctic region and the concentration difference between middle and high latitudes is reduced. This transport process is also responsible for (1) lower concentrations at 700 hPa level over SYO than those at SPO in summer, and (2) the occurrence of a 1-month delay of the minimum concentration of the seasonal O_3 variation at SPO than in coastal region of the Antarctic Continent.

There are still two other possibilities for interpreting low O_3 concentrations in the lower troposphere from spring to early autumn. It has been pointed out by some researchers (e.g., Crutzen, 1988) that O_3 is destroyed by photochemical reactions in unpolluted troposphere with low NO_x concentrations. Crutzen and Gidel (1983) also suggest, from the simulation with their 2-dimensional photochemical model, that photochemical O_3 destruction occurs even in polar summer when the solar altitude is low compared with lower latitudes. Thus, the decrease of the lower tropospheric O_3 concentration from spring to early autumn may be attributed not only to transport of the poor-O_3 air from lower latitudes but also to photochemical O_3 destruction. Such a suggestion is also made by Schnell et al. (1991). In this connection, it is suggested from the seasonal variation of ^7Be concentration at SPO (Larsen and Sanderson, 1988, 1989) that the downward transport from the upper atmosphere to the ground is more effective in summer than in remaining seasons of the year. If this is the case, photochemical destruction of O_3 has to be significant to decrease the lower tropospheric O_3 concentration.

Galbally and Roy (1980) also propose that the destruction rate of O_3 is greater on land surface than on surfaces of snow and sea. As described above, snow melts and naked rock is exposed at SYO in summer. However, our results of surface O_3 measurements on fast ice, apart from SYO in this season, show close agreement with those of SYO, and the time when snow begins to melt at SYO is not in accordance with the time when the O_3 concentration decreases. Therefore, this effect is thought to be not important for producing low concentrations of the seasonal O_3 variation, at least near SYO.

The O_3 transport process for interpretation of the seasonal variation of the tropospheric O_3 in the antarctic region is also proposed by Oltmans and Komhyr (1976, 1986), Oltmans (1981) and Komhyr et al. (1988). They suggest that the stratospheric O_3 intrudes into the troposphere from the tropopause gap near the jet stream in middle latitudes is transported to the antarctic region through the upper troposphere, and that this process is more effective than downward transport of the stratospheric air in the antarctic region. Such a transport process is different from that described in this paper. In this connection, ozonesonde observations on board the icebreaker "Shirase" between low and high latitudes of the southern hemisphere in November and December of 1987–1990 (Matsubara et al., 1991, personal communication) show that the stratospheric O_3 from the tropopause gap tends to move equatorward rather than poleward. The same indication is also found in results of numerical simulations of air parcels (Yamazaki et al., 1989).

For better understanding of temporal and spatial variations of the tropospheric O_3 in the antarctic region, further measurements with aircraft and ozonesondes throughout the year are required in the southern hemisphere, especially from the middle latitudes to the antarctic region, in addition to continuous measurements of surface O_3 concentration. Knowledge from such measurements may be also useful for elucidating the transport of other trace gases such as CO_2 and CH_4 to the antarctic region.

4. Acknowledgements

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