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SEASONAL VARIATION OF TROPOSPHERIC OZONE AND THE IMPACT OF TRANSPORT OVER SYOWA STATION, ANTARCTICA

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Abstract: To interpret the height-dependent seasonal variations of the tropospheric O_3 mixing ratio observed over Syowa Station, Antarctica, atmospheric transport processes were examined using a 3-dimensional trajectory analysis with U.S. National Centers for Environmental Prediction (NCEP) data. Results from the analysis suggest that the seasonal O_3 variations are affected significantly by a combination of the following processes: (1) in the upper troposphere over Syowa, the downward transport of air rich in O_3 from the stratosphere is enhanced, while the transport of air poor in O_3 from lower levels is suppressed from summer to early autumn; (2) in the lower troposphere over Syowa, lower tropospheric air poor in O_3 is transported horizontally from lower latitudes and the transport of air with high O_3 mixing ratios from upper levels is reduced from late spring to early autumn; (3) vertical air mixing is enhanced in the Antarctic troposphere in cold seasons; and (4) in the middle troposphere, the upward transport of lower tropospheric air takes precedence over air transport from upper levels throughout the year except in summer. These transport processes are partly related to some meteorological features at southern high latitudes, such as the circumpolar trough and katabatic circulation.

1. Introduction

Tropospheric O_3 variations are determined by many factors such as intrusion of stratospheric air with high O_3 mixing ratios into the troposphere, destruction at the earth's surface and photochemical production and destruction in the troposphere. Since it is thought that the photochemical production of O_3 is minimal in the Antarctic troposphere due to the fact that the Antarctic air is fairly free from the influence of human activities, the seasonal variation of tropospheric O_3 in the Antarctic region has been interpreted in terms of advection from other regions by atmospheric transport processes (WEXLER *et al.*, 1960; OLTMANS and KOMHYR, 1976, 1986; GALBALLY, 1981; OLTMANS, 1981; SCHNELL *et al.*, 1991; MURAYAMA *et al.*, 1992; OLTMANS and LEVY, 1994). The numerical experiments (LEVY *et al.*, 1985; ROEGOFS and LELIEVELD, 1997) have also suggested that the atmospheric transport is one of the most important factors for tropospheric O_3 variations in the Antarctic region. However, details of these transport processes such as their routes, seasonality and height dependence are not yet fully understood.

MURAYAMA *et al.* (1992) found, from their aircraft measurements of tropospheric O_3 over Syowa Station (69°00′S, 39°35′E) (referred to as SYO), Antarctica, that the seasonal variation of the O_3 mixing ratio is dependent on height. From the comparison of their measurement results with the ozonesonde and total amount of O_3 data over SYO, TOMS (Total Ozone Mapping Spectrometer) data and surface O_3 data at the South Pole, Samoa (OLTMANS and KOMHYR, 1986) and Cape Grim (DOUGLAS *et al.*, 1985), they also speculated that the height-dependent seasonal variations of tropospheric O_3 over SYO are determined mainly by the following processes: (1) intrusion of stratospheric O_3 into the troposphere over the station, (2) transport of air with low O_3 mixing ratios from lower latitudes to the station through the lower troposphere from spring to early autumn, and (3) photochemical destruction of O_3 near the station from spring to early autumn.

An evidence of an impact of long-range transport on the atmospheric measurements at SYO was provided by MURAYAMA *et al.* (1995) who performed a 3-dimensional backward trajectory analysis using meteorological data from the U.S. National Centers for Environmental Prediction (NCEP, formerly NMC), to interpret the seasonal variations of the atmospheric CO_2 mixing ratio observed at different heights over SYO. They suggested from their analysis that height-dependent seasonal air transport processes could influence those seasonal CO_2 variations.

There are a number of processes governing atmospheric transport over the Antarctic region. The activities of cyclones in the circumpolar trough zone and the katabatic circulation over the Antarctic continent are very important factors for air transport in the Antarctic troposphere. The katabatic outflow reaches the south side of the circumpolar trough zone, the air is lifted up there and then returned to upper levels over the Antarctic continent. Therefore, the activities of the circumpolar trough and the katabatic circulation are associated with each other. Both activities are enhanced in cold seasons and are largely suppressed in summer. The large-scale mean diabatic circulation due to the Brewer-Dobson cell is one of the important processes determining downward transport from the stratosphere to the troposphere over this region. However, the seasonality of this downward transport is not fully understood (YAMAZAKI *et al.*, 1989).

For a better understanding of the transport processes governing the seasonal variation of tropospheric O_3 over SYO, we carried out a similar 3-dimensional trajectory analysis. In this paper, the seasonal variation of atmospheric transport derived from this analysis is compared with that of tropospheric O_3 over SYO and the relationship is discussed.

2. Ozone Data

To obtain the average seasonal variations of the tropospheric O_3 mixing ratio at different heights over SYO, monthly mean O_3 values at the ground surface taken from continuous measurements at SYO for the period 1988–1990 (AOKI and YAMANOUCHI, 1994), those at 350 and 500 hPa levels from our aircraft measurements for the period May 1989–January 1990 (MURAYAMA *et al.*, 1992) and from ozonesonde observations for the period 1988–1990 (JAPAN METEOROLOGICAL AGENCY, 1995) over SYO were examined. The surface and aircraft data are obtained from measurements using a Dasibi ozonemeter.

Since the frequency of ozonesonde observation is significantly lower at 350 and 500 hPa (2.8 times per month on average) than that of ground-based continuous measurement, we used mixed data from aircraft and ozonesonde measurements (*i.e.*, measurements by Dasibi ozonemeter and ozonesonde) for 350 and 500 hPa. The relationship of the measured values between our Dasibi O_3 meter and the ozonesonde has been examined by MURAYAMA *et al.* (1992). In this paper, the ozonesonde data were adjusted to the mixing ratio scale of our Dasibi O_3 meter using this relationship so that both sets of data are compatible with each other.

3. Trajectory Analysis

Details of the trajectory analysis used in this study are described by MURAYAMA *et al.* (1995), YAMAZAKI *et al.* (1989) and YAMAZAKI (1992). Therefore, only a brief description and supplemental explanations are presented here. In the trajectory analysis, we used the 3-dimensional twice-daily U.S. NCEP meteorological data given at horizontal grid points every 2.5° in latitude and longitude at 12 standard levels between 1000 and 50 hPa. Vertical wind velocities (ω) were calculated from the horizontal wind field using a mass continuity equation under the assumption that $\omega=0$ at the 30 hPa pressure level. The time step for the calculation was 1 hour, and 3-dimensional wind components at the position for an air parcel at each time step were obtained by interpolating the wind data at contiguous grid points linearly in time and space.

Atmospheric transport processes were examined at every 50 hPa pressure level between 350 and 850 hPa over SYO. For this purpose, 143 (11×13) air parcels were initially assigned in a small horizontal space with dimensions of 0.4° (longitude) by 0.2° (latitude) at each pressure level over SYO (See Fig. 1 in MURAYAMA *et al.*, 1995). In this study, backward trajectories of these parcels for 20 days were calculated for each pressure level, to examine the meridional air transport between southern middle and high latitudes, as well as the vertical air transport. In this connection, MURAYAMA *et al.* (1995) performed a 30-day backward trajectory analysis to interpret the seasonal variation of the CO_2 mixing ratio over SYO. Such long-term trajectory analysis was performed to assess the atmospheric transport to SYO not only from different locations in the southern hemisphere but also from the northern hemisphere. Because the inter-hemispheric CO_2 mixing ratio difference is much larger than the spatial variation of the mixing ratio in the southern hemisphere, it is assumed that transport from the northern hemisphere could also influence CO_2 variability in the Antarctic region. On the other hand, in the case of O_3, in the southern hemisphere its spatial variation is large in the vertical and latitudinal scale compared to that of CO_2. Therefore, it might be concluded that the relative contribution to the Antarctic tropospheric O_3 variation of intra-hemispheric transport in the southern hemisphere is much larger than that of inter-hemispheric transport. With respect to this, LAW *et al.* (1992) showed, from the results of their tracer model, that the transport time from southern midlatitudes to the south polar region is shorter than 10 days even in summer when wind speed is low. Furthermore, the life time in the troposphere of O_3 is shorter than that of CO_2, especially in the tropical region. From these facts, in this paper, we have performed shorter-term trajectory analysis compared to that of MURAYAMA *et al.* (1995) and will discuss distributions of the parcels advected backward for 10 and 20 days.

The arrival time of the parcels at each level over SYO was set to 1200 GMT every 2 days, to obtain an average picture of the air transport over the period from 1 October 1988 to 31 July 1990. In this way, we obtained 15 or 16 distributions of air parcels arriving at each selected level over SYO per month. For each individual month, these 15 or 16 distributions were then superimposed on one another. The parcel distributions thus obtained for each month were used to interpret the height-dependent seasonal variations of the O_3 mixing ratio over SYO in terms of the atmospheric transport.

Long-term trajectory calculations cannot avoid errors due to integration of the uncertainties of wind data and interpolation of the sparse data. For this reason, trajectories are best used as an indication of the general airflow rather than the exact pathway of an air parcel (HARRIS *et al*., 1992; HARRIS and KHARL, 1990; MERRILL, 1989). From the backward trajectory analysis using the same model as in this study for the air parcels assigned initially at close positions in the upper troposphere over SYO, YAMAZAKI *et al*. (1989) showed that the parcels spread over the whole extra-tropical troposphere in the southern hemisphere within 20 days. Such distribution of air parcels also sometimes appeared in this study, though the dispersion of the parcels depends on the specific synoptic conditions. YAMAZAKI *et al*. (1989) extracted the character of the general air transport to SYO from the distribution of the great number of the parcels spread widely by statistical procedures. KIDA (1983), YAMAZAKI (1992) and TAGUCHI (1994) have also found the character of the stratospheric-tropospheric and/or inter-hemispheric transport processes using statistical distribution of air parcels obtained from their long-term trajectory analysis for more than 30 days. Therefore, in this study many trajectories are also calculated, their distributions are statistically analyzed, and not the details of individual trajectories but qualitatively characteristic features of the atmospheric transport are discussed.

4. Results

4.1. *Altitude dependent ozone variations*

Figure 1 shows average seasonal variations of the O_3 mixing ratio by volume at selected pressure levels over SYO. Monthly mean values at each height in the respective years were calculated from the observed data, and then the values for each month were simply averaged during 1988–1990. Here, to calculate the monthly mean values in the respective years, the daily mean values were used for the surface, while the observed data in a month (the number of data is about 3 on average) were used for the 350 and 500 hPa levels. In this figure, year-to-year variations of the monthly mean value for each month of the year are also represented as the standard deviations (1σ) from the average monthly values for 1988–1990 obtained above. In this study, the monthly mean values of the respective years were calculated from the observed data in each month, whose numbers are much smaller at 350 and 500 hPa than at the ground surface, as mentioned above. Therefore, the calculated monthly mean values depend on the observation date, and the large standard deviations at 350 hPa discussed below reflect mainly the large O_3 mixing ratio variation in a month rather than its year-to-year variation.

The O_3 mixing ratio increases with height in all seasons. Surface O_3 shows a prominent seasonal variation with a maximum in winter and a minimum in summer, which is similar to those observed at other southern hemisphere stations: the South Pole,

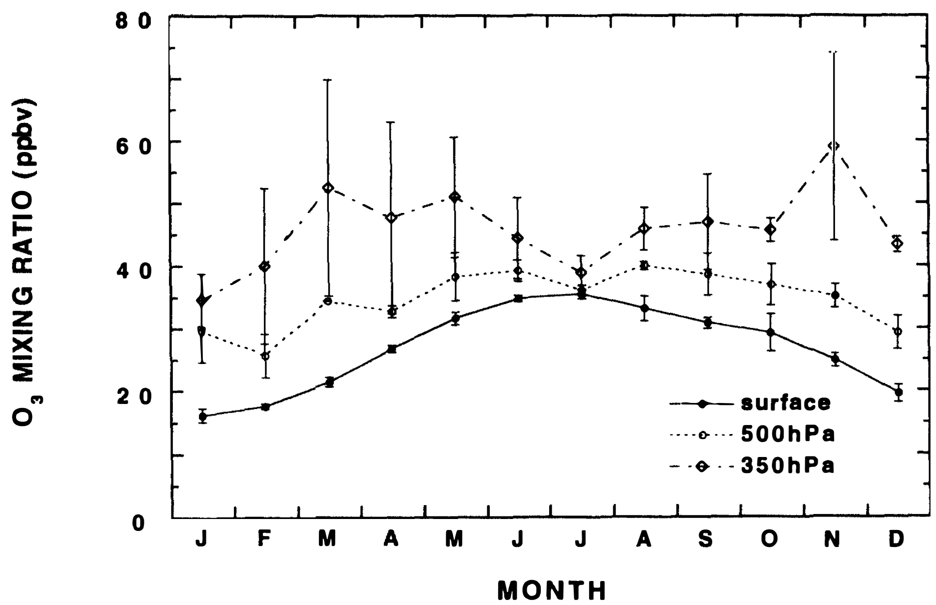

Fig. 1. Average seasonal variations of the tropospheric O_3 mixing ratio at 350 (~7.4 km), 500 hPa (~5.0 km) levels and the ground surface at Syowa Station, Antarctica for 1988–1990. The standard deviations (1σ) from the average monthly values during the period are also shown.

Cape Grim and Samoa (OLTMANS and KOMHYR, 1986; DOUGLAS *et al.*, 1985). The seasonal variation of middle tropospheric O_3 (500 hPa) is similar to that at the surface. However, since the O_3 mixing ratio at this pressure level does not decrease as steeply as at the ground surface from spring to summer, the peak-to-peak amplitude of the seasonal variation decreases with height. On the other hand, the seasonal variation of the O_3 mixing ratio in the upper troposphere (350 hPa) is not so clear; the mixing ratio shows high values during the period of late summer to autumn (February to May), decreases from autumn to winter (May to July), increases gradually from late winter to spring (August to October) and then shows a high value in late spring (November). As a result, the height-dependent difference of the O_3 mixing ratio is large from late spring (around November) to early autumn (around March); a minimum mixing ratio gradient in O_3 is observed in winter. The standard deviations at the 350 hPa level are much greater than those at the ground surface and the 500 hPa level, and the observed data at 350 hPa showed larger variability of the mixing ratio between each observed date in a month compared to lower height levels. This is likely to be related to the intrusion of stratospheric air with rich O_3 into heights above 6 km which is sometimes observed over SYO (Fig. 4 in MURAYAMA *et al.*, 1992).

4.2. Atmospheric transport

To examine the relationship between these seasonal O_3 variations and the atmospheric transport, we first summarized the results of the trajectory analysis in terms of the vertical air mixing.

Using the distributions of air parcels obtained from the trajectory analysis, we counted the numbers of air parcels located in each height layer with 100 hPa intervals (layer 1: 0–

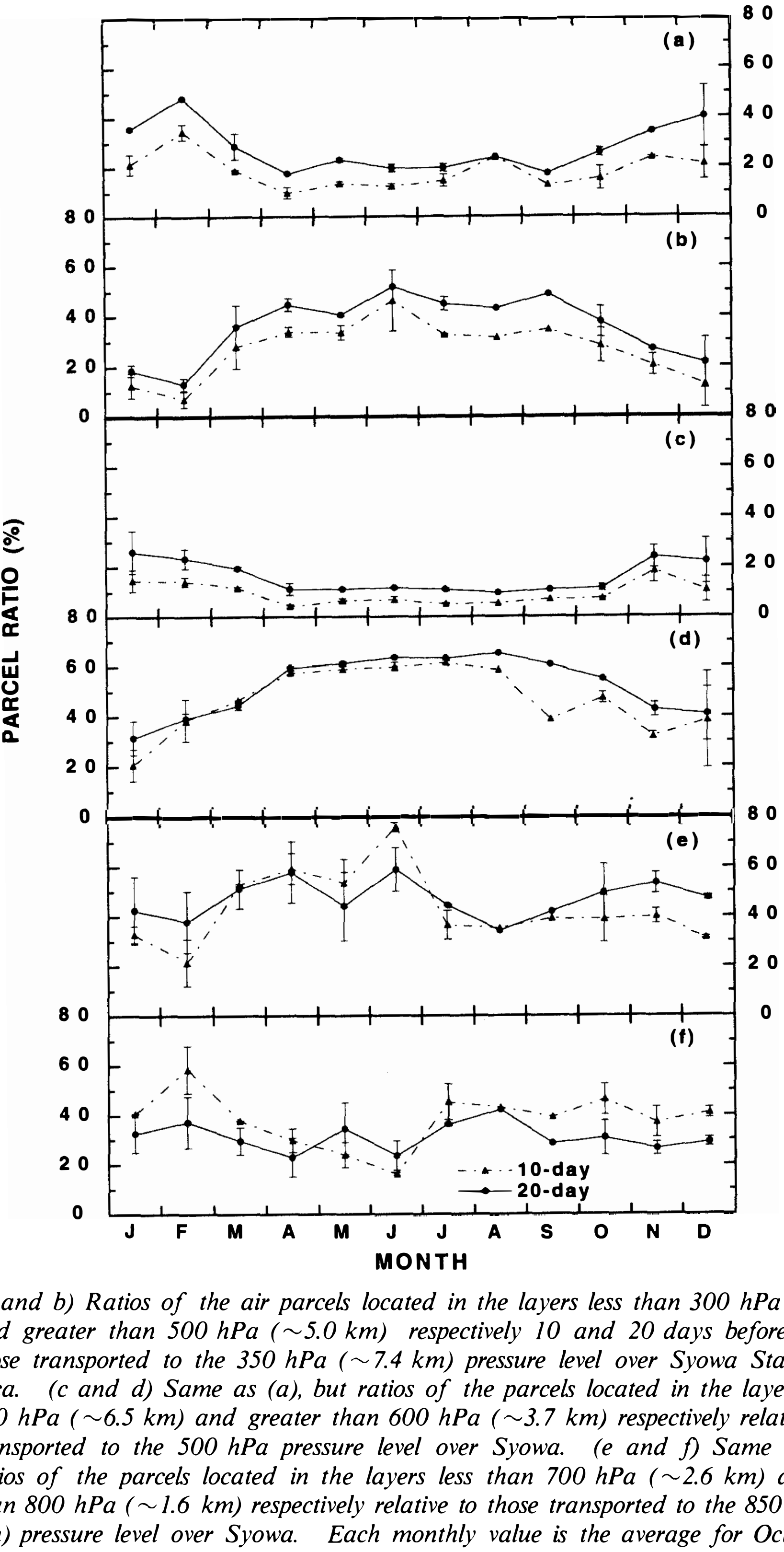

Fig. 2. *(a and b) Ratios of the air parcels located in the layers less than 300 hPa (~8.4 km) and greater than 500 hPa (~5.0 km) respectively 10 and 20 days before relative to those transported to the 350 hPa (~7.4 km) pressure level over Syowa Station, Antarctica. (c and d) Same as (a), but ratios of the parcels located in the layers less than 400 hPa (~6.5 km) and greater than 600 hPa (~3.7 km) respectively relative to those transported to the 500 hPa pressure level over Syowa. (e and f) Same as (a), but ratios of the parcels located in the layers less than 700 hPa (~2.6 km) and greater than 800 hPa (~1.6 km) respectively relative to those transported to the 850 hPa (~1.1 km) pressure level over Syowa. Each monthly value is the average for October 1988–July 1990. The standard deviations (1σ) from the average monthly values during the period are also shown.*

100 hPa, layer 2: 100–200 hPa, ……, layer 10: 900–1000 hPa, layer 11: >1000 hPa) 10 and 20 days prior to the arrival at a particular level (p) over SYO in a particular month. After the counting, we calculated the ratios of the number of air parcels ($r^{p}_{k1,k2}$) located in a particular layer (layer $k1$ to $k2$) 10 and 20 days prior to arrival at a particular pressure level (p) over SYO in a particular month, to the total number of air parcels arriving at the same level (p) over SYO from all layers (*i.e.*, layer 1 to 11) in the same month, for each month from October 1988 to July 1990, as follows:

$$r^{p}_{k1,k2}=\sum_{l=k1}^{k2} n_{p,l}\Big/\sum_{l=1}^{11} n_{p,l}, \qquad (1)$$

where $n_{p,l}$ is the number of air parcels in the layer l 10 or 20 days prior to the arrival at a particular pressure level (p) over SYO in a particular month. For example, the number of air parcels located in layers of less than 300 hPa pressure level (<300 hPa; heights greater than 300 hPa height level; layers 1–3) 10 or 20 days prior to a particular arrival time at 350 hPa level over SYO is divided by the total number of air parcels arriving from all layers at the 350 hPa level over SYO at the same time ($r^{350}_{1,3}$). Similar calculations were carried out to obtain the relative numbers of parcels transported from layers greater than 500 hPa pressure level (>500 hPa; heights less than 500 hPa height level; layers 6–11) to 350 hPa ($r^{350}_{6,11}$) over SYO, that from <400 hPa layers (layers 1–4) to 500 hPa over SYO ($r^{500}_{1,4}$), that from >600 hPa layers (layers 7–11) to 500 hPa over SYO ($r^{500}_{7,11}$), that from <700 hPa layers (layers 1–7) to 850 hPa over SYO ($r^{850}_{1,7}$) and that from >800 hPa layers (layers 9–11) to 850 hPa over SYO ($r^{850}_{9,11}$). These ratios from individual months were then used to calculate 12 average monthly ratios. The results thus obtained are shown in Fig. 2. In Fig. 2, year-to-year variations of the monthly values are also represented as standard deviations (1σ) from average monthly values during the period except in August and September in which the monthly values were calculated only for 1989. From these results, we can qualitatively discuss seasonally-dependent atmospheric transport processes to the respective levels in the troposphere over SYO.

It is seen from Fig. 2 that the atmospheric transport from the layer of <300 hPa at other locations to the 350 hPa level over SYO is enhanced from summer to early autumn (roughly November to March) and transport from lower height levels is largely suppressed; this situation is reversed in cold seasons. The results of both 10-day and 20-day trajectory analyses are generally similar to each other. The difference between 10-day and 20-day trajectory results is attributed mainly to divergence of backward trajectories from the arrival pressure level to the upper and lower levels occurring with time. On the other hand, the results of the 10-day trajectory analysis for the 850 hPa level over SYO show that the vertical transports to 850 hPa level over SYO from the layers of <700 and >800 hPa are enhanced and suppressed, respectively, from autumn to early winter (March to June) and the situation is reversed in the remaining seasons of the year, though the seasonal variation of the atmospheric transport estimated by 20-day trajectory analysis for the same pressure level over the station is somewhat obscure. From comparison of the results for 850 hPa of the 10-day analysis with those of the 20-day analysis, it is seen that the ratios of air parcels located at upper levels are greater in the results for the 20-day analysis than in those for the 10-day analysis except in May and June. This may show that backward trajectories are dispersed from the 850 hPa level mainly to upper levels with time because parcels which have spread to lower levels can easily arrive at the ground surface. The opposite situation in May and June will be discussed later. It is therefore thought that vertical mixing of tropospheric air is enhanced from autumn to early winter (April to June) and reduced in the remaining seasons. With respect to the results of the analysis for the middle troposphere (500 hPa), the transports of air parcels from upper (<400 hPa) and lower height levels (>600 hPa) are enhanced in summer and in winter, respectively; this is similar to the result for the 350 hPa level. However, the transport of the parcels from lower levels dominates over that from upper levels throughout the year except in summer, as seen in Figs. 2 (c) and (d).

We also estimated the seasonality of the advection from higher and lower latitudes to different heights over SYO by a similar method to that used to obtain Fig. 2. In this case, we counted the numbers of air parcels located in each latitudinal band with 10° intervals (band 1: 90–80°S, band 2: 80–70°S, ······, band 17: 70–80°N, band 18: 80–90°N) 10 and 20 days prior to a particular level over SYO in a particular month. Similar to eq. (1), the relative number of air parcels ($R^p_{j1,j2}$) located in a particular latitudinal band (band $j1$ to $j2$) 10 and 20 days prior to the arrival at a particular pressure level (p) over SYO in a particular month is represented as follows:

$$R^p_{j1,j2} = \sum_{L=j1}^{j2} N_{p,L} \Big/ \sum_{L=1}^{18} N_{p,L}, \qquad (2)$$

where $N_{p,L}$ is the number of air parcels in the latitudinal band L. Figure 3 shows the seasonal variations of the relative number of the air parcels transported from higher (>70°S) and lower (<60°S) latitudes to the 350 ($R^{350}_{1,2}$, $R^{350}_{4,18}$), 500 ($R^{500}_{1,2}$, $R^{500}_{4,18}$) and 850 hPa ($R^{850}_{1,2}$, $R^{350}_{4,18}$) levels over SYO.

The results of the 10-day trajectory analysis for the 850 hPa level show that the transport from higher latitudes (>70°S) is enhanced from autumn to early winter (March to June), suppressed from late winter to early spring (July to September) and slightly enhanced from late spring to early summer (October and November), though the result of the 20-day trajectory is not very clear. On the other hand, many air parcels are transported from lower latitudes (<60°S) from late winter to summer (July to February); number is decreased from autumn to early winter (March to June). By comparing the results in Fig. 3 with those in Fig. 2, it is found that the seasonal variations of the atmospheric transport from upper (<700 hPa) and lower layers (>800 hPa) to 850 hPa level over SYO are similar to those from higher (>70°S) and lower latitudes (<60°S), respectively. Accordingly, the transport of air from higher latitudes and upper levels to the 850 hPa level over SYO is predominant from autumn to early winter (March to June) and that from lower latitudes and lower levels is enhanced in the remaining seasons. On the other hand, the seasonal variations of the relative number of the air parcels transported from higher and lower latitudes are hardly seen in the results at 350 and 500 hPa levels. The number of parcels transported from lower latitudes (<60°S) is larger than that from higher latitudes (>70°S) throughout the year. The comparison of the result of the 10-day backward trajectory analysis with that of the 20-day analysis shows that more air parcels are located outside the Antarctic region 20 days before arrival to respective heights over SYO than 10 days before. This may reflect divergence of backward trajectories occurring with time and predominant poleward transport from lower latitudes at upper levels.

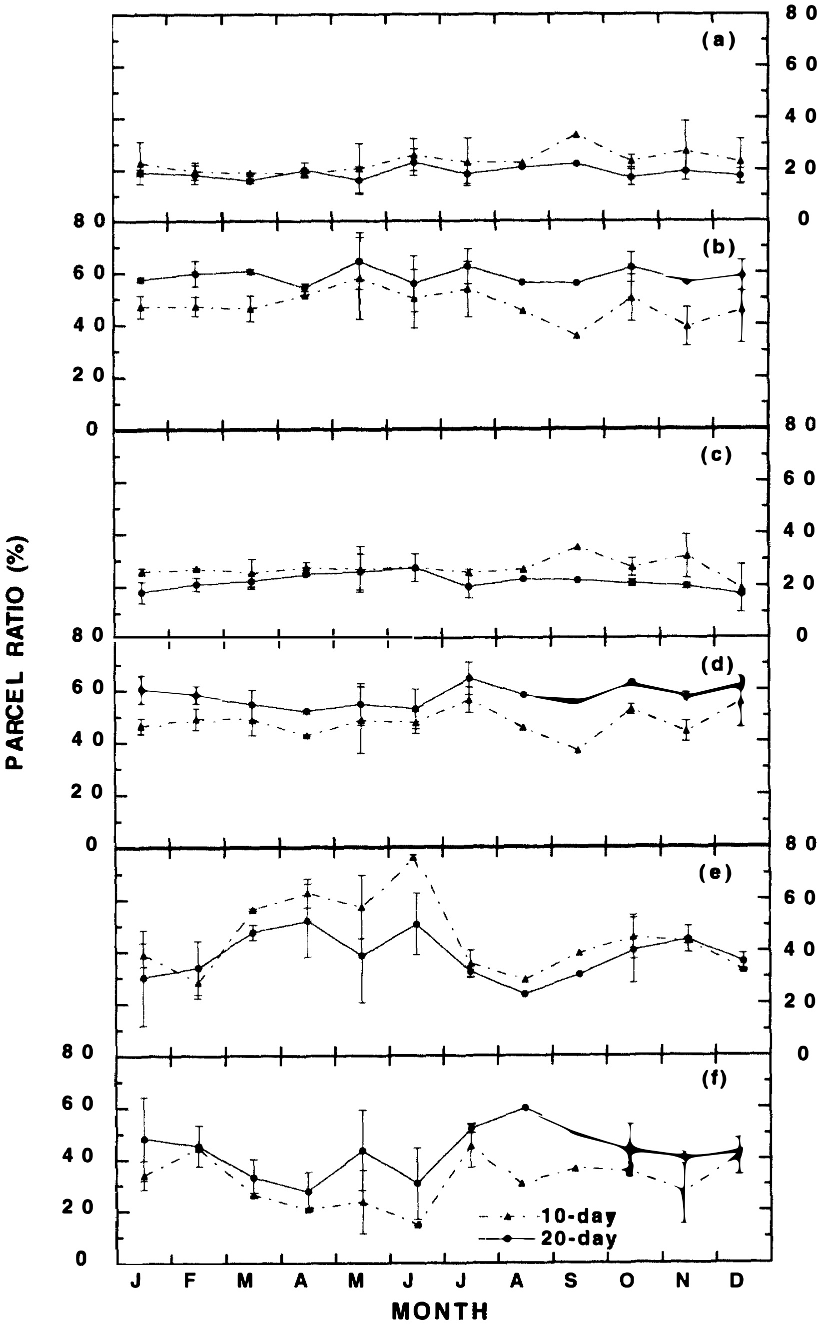

Fig. 3. Same as in Fig. 2, but ratios of the air parcels located at latitudes of <60°S and >70°S 10 and 20 days before, respectively, relative to those transported to (a and b) 350 (~7.4 km), (c and d) 500 (~5.0 km) and (e and f) 850 hPa (~1.1 km) pressure levels over Syowa Station, Antarctica.

5. Discussion

We suggested above from the results of our trajectory analysis that the atmospheric vertical mixing is more enhanced in the troposphere over SYO in cold seasons than in warm seasons. With respect to this, the seasonal variation of the Richardson number (Ri) which represents atmospheric stability in the selected layer was examined. Ri is defined as

$$Ri=\left(\frac{g}{\theta_0}\right)\cdot\frac{\partial\theta}{\partial z}\Big/\left(\frac{\partial u}{\partial z}\right)^2, \tag{3}$$

where g is the gravity acceleration, θ is the potential temperature, θ_0 is the representative potential temperature of the selected layer (in this study, we used the average of the potential temperatures at 300 and 850 hPa), u is the horizontal wind speed and z is height.

Figure 4 shows the seasonal variations of zonal mean Ri in the layer 300–850 hPa over latitudes 60, 70 and 75°S in 1989 calculated using the U.S. NCEP data and that of Ri in the same layer over SYO in 1989 calculated using the radiosonde data at SYO. It is seen from this figure that zonal means of Ri at each latitude show high values in summer and low values in the remaining seasons, and that Ri is slightly smaller at 60°S than at 70 and 75°S throughout the year, which may suggest that vertical air mixing is more enhanced in the circumpolar trough zone than in the Antarctic region. On the other hand, high Ri values are seen over SYO from winter to early spring as well as in summer. With respect to this, the monthly mean 300 hPa geopotential height maps showed that centers of low pressure were located over the Ross Sea and near SYO from winter to early spring in 1989. As a result, the circumpolar trough moved northward away from SYO and wind speed at 300 hPa was lower over SYO than that of the zonal mean in the same latitude band. Since the wind is generally weaker at 850 hPa than that at 300 hPa, vertical wind shear (du/dz) in the layer 300–850 hPa over SYO was reduced during this period, and then Ri was increased. Taking account of the fact that air is zonally mixed immediately, however,

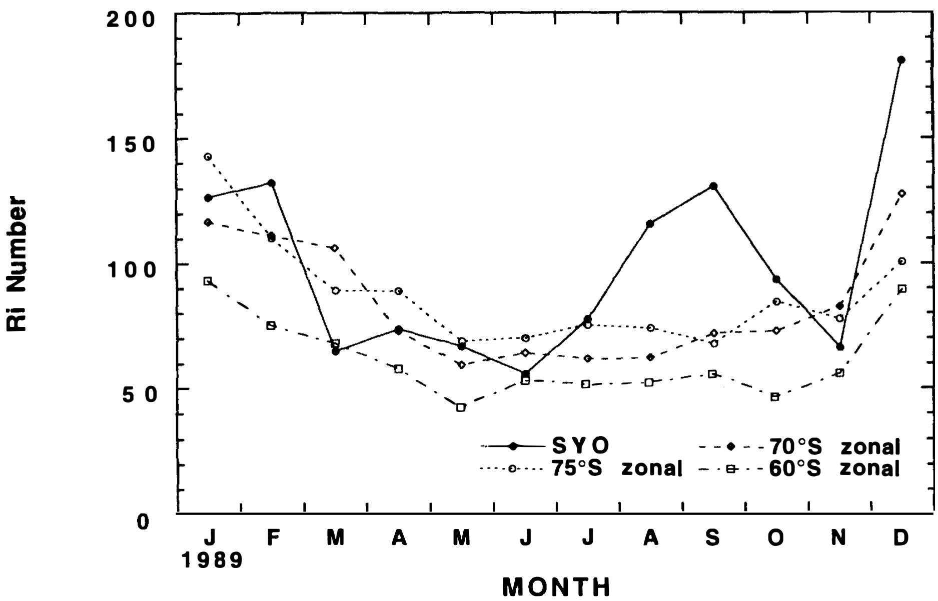

Fig. 4. Zonal and monthly average values of the Richardson number in the layer 300–850 hPa at latitudes of 60, 70 and 75°S in 1989 calculated from the U. S. NCEP meteorological data and the monthly values at Syowa Station (SYO) in 1989 from aerological data at the station.

it is thought that the zonal mean of *Ri* is more useful for comparison with results of the trajectory analysis than *Ri* at SYO. The zonal mean *Ri* suggests that the vertical atmospheric stability is greater at latitudes near SYO in summer than in cold seasons, and that the vertical atmospheric mixing is enhanced in cold seasons. This result is consistent with the results of the trajectory analysis.

By using the results of the trajectory analysis the seasonal variations of the O_3 mixing ratio at different levels may be explained to a certain extent in terms of vertical transport and advective processes as follows.

In the upper troposphere, the downward transport of air rich in O_3 from upper levels is enhanced and stratospheric-tropospheric air exchange is likely to occur in summer. In this regard, YAMAZAKI *et al.* (1989) have suggested that aerosol particles from the Mt. El Chichon eruption and accumulated in the upper troposphere over SYO in January 1983 were transported to the Antarctic through the stratosphere and then descended, and indicated that this route is of primary importance for transport to the Antarctic upper troposphere in summer. In addition, the transport of air poor in O_3 from lower levels to the upper troposphere is suppressed during this period. Therefore, the O_3 mixing ratio shows high values in the upper troposphere from late spring to early autumn. During the period from late autumn to winter, the transport from lower levels takes precedence over that from upper levels, and the upper tropospheric O_3 mixing ratio decreases. Downward and upward transports of the air are respectively enhanced and suppressed again in spring, by which upper tropospheric O_3 is increased. Large standard deviations of the mixing ratio appeared at 350 hPa in months when the monthly mean O_3 showed high values. This may be attributed to the fact that the air is more often and strongly influenced by the stratospheric-tropospheric exchange at this height level in these months than in the remaining months of the year, which is in general consistent with the results of the trajectory analysis.

For the seasonal variation of the lower tropospheric O_3 mixing ratio, not only the photochemical destruction of O_3 but also the following transport processes may be very important factors. Taking account of the latitudinal distribution of the surface O_3 in the southern hemisphere (MURAYAMA *et al.*, 1992), where the mixing ratio increases with increasing latitudes, the horizontal transport of air with poor O_3 from low height levels at lower latitudes is suppressed and the downward transport of air with rich O_3 from upper levels over the Antarctic is enhanced from autumn to early winter. By these processes, the lower tropospheric O_3 mixing ratio is increased. In the remaining seasons of the year, the mixing ratio is decreased due to the suppression of vertical transport of air rich in O_3 from upper levels and enhancement of the horizontal transport of air poor in O_3 from the lower troposphere over lower latitudes. SCHNELL *et al.* (1991) also suggested that this intrusion of air with low O_3 mixing ratios into the Antarctic is one of the causes of the summertime decrease of surface O_3 at the South Pole.

In the middle troposphere, upward transport of the lower tropospheric air takes precedence over downward air transport from upper levels throughout the year, except in summer. Therefore, similar seasonal variations of the O_3 mixing ratio appear in the middle and lower troposphere. Especially, in cold seasons when the transport from lower levels is enhanced, the middle tropospheric O_3 mixing ratio is close to that in the lower troposphere. However, since the downward and upward transports of the air to the middle troposphere are respectively enhanced and suppressed in summer, the decrease in O_3 is reduced and is not as much as in the lower troposphere. As a result, the peak-to-peak amplitude of the seasonal O_3 variation is smaller in the middle troposphere than in the lower troposphere.

The steep O_3 vertical gradient observed from summer to early autumn is thought to be produced by high O_3 mixing ratios in the upper troposphere due to the downward transport of air rich in O_3, as well as by low O_3 mixing ratios in the lower troposphere due to the southward transport of lower tropospheric air poor in O_3. This vertical difference of the O_3 mixing ratio is reduced by vertical mixing of air (enhancement of atmospheric instability) from late autumn to winter, in association with the seasonality of the activities of the circumpolar trough and the katabatic circulation. A similar seasonally-dependent vertical mixing ratio difference has been observed by MURAYAMA *et al.* (1995) for tropospheric CO_2 over SYO, whose cause is also ascribed to the air transport processes.

A considerable amount of air is transported to the middle and upper troposphere over SYO from lower latitudes. Therefore, it is suggested that the O_3 variations at these height levels over SYO are affected by those at lower latitudes, in addition to those in the Antarctic.

Though day-to-day O_3 variations in the upper troposphere and stratosphere are much larger than those in the lower troposphere, the frequency of O_3 observations is much smaller at the upper levels than at the surface. The fact that enhancement of downward air transport from the stratosphere to the upper troposphere estimated in this study is not fully consistent with occurrence of high monthly mean mixing ratios in the upper troposphere (Figs. 1 and 2) may be due partly to this low frequency of observations at upper levels.

We now discuss the results of our trajectory analysis in terms of meteorological processes at southern high latitudes. In Fig. 5, zonally and monthly averaged distributions of potential temperature for January and July in 1989 calculated from the U.S. NCEP data are shown. It is found from this figure that each isentropic surface slopes downward from high to low latitudes, and that its vertical gradient is steeper over the Antarctic region in January than in July. The differences of potential temperature between 300 and 800 hPa at 70°S are 39 and 30 K in January and July, respectively. It is also suggested from this that vertical mixing of air in the troposphere can be more enhanced in cold seasons than in warm seasons, though consistency with the seasonality of *Ri* is not surprising because *Ri* is dependent on the vertical gradient of potential temperature. The assumption of adiabatic motion of air parcels is not bad as a first-approximation in the extra-tropical troposphere for short-term period (DANIELSEN, 1961). From a comparison of Fig. 5 with Figs. 2 and 3, the upward and poleward transport to 350 and 500 hPa over SYO is thought to be related to movement on constant potential temperature surfaces. The height-latitude gradient of isentropic surfaces in the middle and upper troposphere is steeper in winter than in summer. Especially in the upper troposphere, the gradient is very gentle south of 60° S in summer. This might suppress upward air transport from lower levels to the Antarctic middle and upper troposphere more in warm seasons than in cold seasons, which is consistent with the results of the trajectory analysis. Since the transport to 850 hPa over SYO from higher latitudes and upper levels from autumn to early winter and that from lower latitudes and lower levels in the remaining seasons seem to be due to motion on

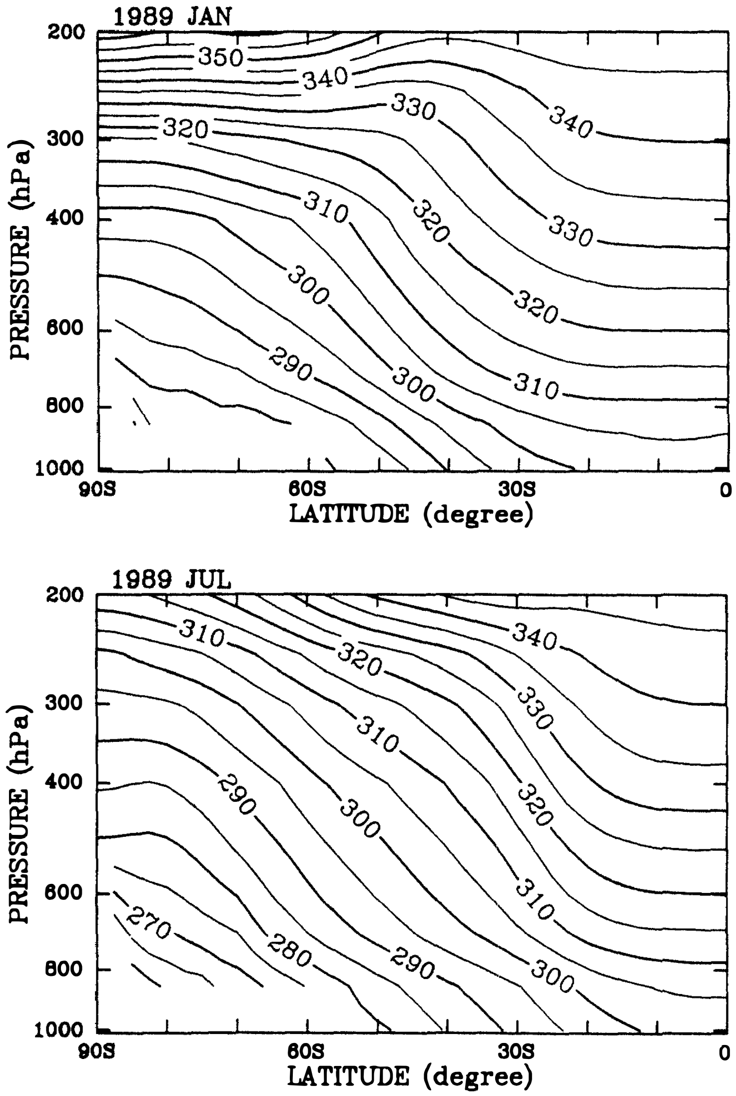

Fig. 5. Zonally and monthly averaged distributions of potential temperature for January and July of 1989 calculated from the U.S. NCEP meteorological data.

isentropic surfaces sloping downward from high to low latitudes, they may be also partly related to the adiabatic motion.

The activities of cyclones in the circumpolar trough zone and the katabatic circulation over the Antarctic continent are enhanced in cold seasons and are largely suppressed in summer. Since such seasonal variations are consistent with those of vertical mixing of the tropospheric air estimated from the trajectory analysis and *Ri* analysis, the seasonality of these activities may be responsible for the tropospheric vertical mixing. In Figs. 2(e) and 3(e), a small peak is also seen in spring, which shows that the air transport from the high interior of the continent to low levels over SYO is somewhat enhanced in this season. This might be related to semi-annual oscillations of the intensity and the position of the circumpolar trough reported by VAN LOON (1967); its intensity is increased and its center moves most southward in spring and autumn. The atmospheric transport from lower levels at lower latitudes to the lower troposphere over SYO is also expected to be enhanced with enhancement of activity of the circumpolar trough. However, the transport from lower levels at lower latitudes to 850 hPa over SYO seems to be generally enhanced in seasons when its activity is relatively low, though this transport is also enhanced late in winter when the circumpolar trough is active (Figs. 2(f) and 3(f)). In the case of the lower troposphere over SYO, downward transport from higher latitudes associated with the katabatic meridional circulation largely reflects the activity of the circumpolar trough, while it is speculated that the air transport from lower levels at lower latitudes may be attributed not only to activity of the circumpolar trough but also to synoptic-scale air transport caused by invasion of cyclones from lower latitudes into the region near the Antarctic coast. In this connection, STRETEN and TROUP (1973) showed that cyclones which developed at southern midlatitudes often move poleward and then decay near the Antarctic coast. By this process, the air originating at lower latitudes is transported to the lower troposphere over SYO through lower levels. Its effect may become relatively prominent in seasons when the katabatic circulation is relatively reduced.

The air circulation over SYO estimated from the trajectory analysis from autumn to early winter seems to represent the katabatic circulation clearly. In these seasons, downward and northward transport of air parcels from higher latitudes and upper levels over the continent to the lower troposphere (850 hPa) over SYO is enhanced, and upward and poleward transport from lower latitudes and lower levels to the middle and upper troposphere (500 and 350 hPa) over SYO is predominant. These air flows are thought to correspond to the divergent drainage flow in the lower layer and to the convergent flow in the upper layer in the katabatic circulation, respectively, which is in good agreement with the schematic view of the mass circulation in the Antarctic region suggested by JAMES (1989) and PARISH and BROMWICH (1991).

During May and June, the relative number of parcels located at $>70^{\circ}$S from 10 days before to 20 days before arrival at 850 hPa over SYO is reduced compared to the remaining months (Fig. 3(e)), which corresponds to the decrease of the relative number of parcels located in layers higher than 700 hPa in height between 10 days before and 20 days before arrival at 850 hPa in the same months (Fig. 2(e)). This might suggest the following processes in terms of the katabatic circulation: (1) many air parcels transported to 850 hPa over SYO are located at upper levels over the Antarctic continent, where the subsiding motion is enhanced, 10 days before, reflecting the enhancement of the drainage flow during this period, (2) some of these parcels are located in lower levels in the circumpolar region, where the convective motion is enhanced, 20 days before. Thus, this convective motion partly compensates for the subsiding motion over Antarctica, which is consistent with the meridional air circulation in this region simulated using general circulation models (PARISH *et al*., 1994; TZENG *et al*., 1993, 1994).

6. Conclusions

To interpret the seasonal variations of the tropospheric O_3 mixing ratio observed at different heights over SYO, atmospheric transport processes were examined using a 3-dimensional trajectory analysis with U.S. NCEP data. Results from the analysis suggest that a combination of the following processes is an important factor governing the seasonal tropospheric O_3 variations over SYO: (1) in the upper troposphere over SYO, the downward transport of air rich in O_3 from the stratosphere is enhanced, while the transport of air poor in O_3 from lower levels is suppressed from summer to early autumn, (2) in the lower troposphere over SYO, lower tropospheric air poor in O_3 is transported horizontally from lower latitudes and the transport of air with high O_3 mixing ratios from upper levels is reduced from late spring to early autumn, (3) vertical air mixing is enhanced in the Antarctic troposphere in cold seasons, and (4) in the middle troposphere, the upward transport of lower tropospheric air takes precedece over air transport from upper levels throughout the year except in summer. These transport processes are partly related to meteorological features at southern high latitudes such as the circumpolar trough and katabatic circulation.

In this paper, we have discussed the seasonal variations of the tropospheric O_3 mixing ratio over SYO in terms of atmospheric transport processes. However, other factors should also be considered in a more complete interpretation of the O_3 mixing ratio variations over SYO. For example, photochemical destruction of O_3 also plays an important role in the seasonal variation of tropospheric O_3 in the Antarctic region, especially in the lower troposphere. In fact, the observed mixing ratios of lower tropospheric O_3 showed a prominent seasonal variation, while the seasonal variation of the atmospheric transport processes in the lower troposphere is not so clear from the present trajectory analysis. Therefore, it seems difficult to explain the O_3 variations in the lower troposphere over SYO only in terms of the atmospheric transport processes.

For a more definitive and quantitative understanding of the seasonal variation of tropospheric O_3 over Antarctica, further analyses, including photochemical processes and atmospheric diffusive effect, are necessary. Systematic measurements of tropospheric and stratospheric O_3 are also required, especially at southern middle and high latitudes. Though day-to-day O_3 variations in the upper troposphere and stratosphere are much larger than those in the lower troposphere, the frequency of observations is also smaller at upper levels than at the surface. To grasp more accurate features of the seasonal O_3 variations at upper levels, more frequent observations are necessary. It is also interesting to examine the relationship of inter-annual variability between the tropospheric O_3 and the atmospheric transport. The comparisons of O_3 variations with those of other trace gases such as CO_2 and CH_4, as well as with that of atmospheric radio nuclides such as ^7Be and ^{14}C, originating in the upper troposphere and/or the stratosphere over the Antarctic region, would be very useful to study transport processes of tropospheric O_3 in more detail.

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