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MEASUREMENTS OF THE OXYGEN ISOTOPIC RATIO OF ATMOSPHERIC CO_2 AT SYOWA STATION, ANTARCTICA

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Abstract: Ground-based and aircraft measurements of the $\delta^{18}O$ of atmospheric CO_2 were made at Syowa Station, Antarctica during 1989 and 1990. To minimize the possibility of exchange of oxygen atoms between CO_2 and water during storage in sampling flasks, CO_2 was extracted from each air sample at the station within a few days of its collection. The maximum of the seasonal $\delta^{18}O$ cycle at the station appeared in summer and the minimum in winter. The peak-to-peak amplitude of the cycle was 0.27‰. $\delta^{18}O$ of CO_2 was slightly lower in the upper troposphere than in the middle and lower troposphere over the station during the measurement period. These results are in good agreement with those at Cape Grim, Tasmania and over Bass Strait. From a comparison of the CO_2 measurements at and over Syowa Station and $\delta^{18}O$ at other stations, it is suggested that seasonally dependent atmospheric transport to the Antarctic region is one of the important processes determining the seasonal cycle and the height variation of $\delta^{18}O$ of CO_2 at the station.

1. Introduction

Oxygen atoms in CO_2 are exchanged with water through a hydrolysis reaction with liquid water. The ratio of oxygen isotopes (^{18}O and ^{16}O) in atmospheric CO_2 should follow the $^{18}O/^{16}O$ ratio of the liquid water reservoirs with which the atmosphere makes most frequent and prolonged contact. Among these reservoirs, the oceans were previously believed to be the primary reservoir for the exchange of oxygen atoms in atmospheric CO_2, because the data observed until then were close to the oxygen isotopic values expected from local equilibrium between CO_2 and surface sea water (*cf.*, BOTTINGA and CRAIG, 1969). However, more precise measurements have been made subsequently, and it has been found that the spatial and temporal distributions of the ratio $^{18}O/^{16}O$ ($\delta^{18}O$) of atmospheric CO_2 do not agree with those expected from the variations of sea surface temperature (MOOK *et al.*, 1983; TANS *et al.*, 1986; FRANCEY and TANS, 1987; FRIEDLI *et al.*, 1987; FRANCEY *et al.*, 1990). The observed $\delta^{18}O$ data show a strong, asymmetric meridional gradient, and show much lower values, especially at northern high latitudes, compared to the $\delta^{18}O$ values expected from oceanic exchange (FRANCEY and TANS, 1987; FRANCEY *et al.*, 1990).

To explain such latitudinal distribution, the exchange of oxygen atoms between CO_2 and leaf water was proposed (FRANCEY and TANS, 1987). They speculated that a large fraction of the CO_2 that enters leaf cells was leaking back out to the atmosphere after exchanging oxygen atoms with leaf water. This process of exchanging oxygen atoms occurs very quickly due to the existence of the enzyme carbonic anhydrase. Thus, $\delta^{18}O$ of atmospheric CO_2 has recently become a valuable tracer for better understanding of not only hydrological cycles, but also carbon cycles, especially between the atmosphere and land biosphere. FARQUHAR *et al.* (1993) have attempted to simulate the behavior of $\delta^{18}O$ using a model which includes the processes proposed by FRANCEY and TANS (1987). However, this model necessarily requires keeping track of a large number of variables, and some assumptions which are not yet fully understood are necessary to obtain these variables.

Systematic isotopic measurements of atmospheric CO_2 are still scarce. In particular, the reported results for $\delta^{18}O$ are fewer than those for $\delta^{13}C$ (MOOK *et al.*, 1983; KEELING *et al.*, 1984; FRANCEY and TANS, 1987; FRIEDLI *et al.*, 1987; FRANCEY *et al.*, 1990; NAKAZAWA *et al.*, 1993), though the carbon and oxygen isotopic ratios of a CO_2 sample can be measured simultaneously using a mass spectrometer. This is attributed mainly to the fact that $\delta^{18}O$ tends to be affected by exchange of oxygen between CO_2 and water during storage in the sample flasks, and that interpretation of the results obtained from $\delta^{18}O$ measurements is more difficult compared to $\delta^{13}C$. Especially, the reported results of $\delta^{18}O$ measurements in the Antarctic region have been very few (MOOK *et al.*, 1983; FRANCEY *et al.*, 1990).

We have made ground-based and aircraft CO_2 measurements at Syowa Station (69°00'S, 39°35'E) since 1983 (TANAKA *et al.*, 1987; NAKAZAWA *et al.*, 1991a; MURAYAMA *et al.*, 1995). From these measurements, it is suggested that the CO_2 concentration in the Antarctic region is affected by seasonally dependent atmospheric transport processes from other latitudes, which reflects the fact that Antarctica is largely free from local and regional effects due to vegetated lands and industrial area. For a better understanding of CO_2 variations in the Antarctic region in terms of atmospheric transport processes, we initiated $\delta^{18}O$ and $\delta^{13}C$ measurements of atmospheric CO_2 using an extraction system installed at Syowa Station in 1989.

In this paper, we report the results of the measurements of $\delta^{18}O$ of atmospheric CO_2 at and over Syowa Station. Comparison with the results of the measurements of the CO_2 concentration at and over Syowa Station and $\delta^{18}O$ at other stations is also made. From these, the seasonal cycle and height-dependent distribution of $\delta^{18}O$ observed at Syowa are discussed.

2. Measurements

In this study, air samples at Syowa Station were taken from the top of the mast used for continuous measurements of CO_2 and CH_4 concentrations (NAKAZAWA *et al.*, 1991a; AOKI *et al.*, 1992). Detailed descriptions of the sampling site have been given elsewhere (NAKAZAWA *et al.*, 1991a). Each 1500 m*l* evacuated Pyrex glass flask was filled with air sample by a diaphragm pump. NAKAZAWA *et al.* (1991a) have shown that the CO_2 concentration variations resulting from station activities are barely observable at Syowa

Station but such contaminated data sometimes appear under weak wind conditions. In order to avoid contamination by station activities, samples were collected during times when the output of a CO_2 analyzer used for the continuous measurements indicated "background" conditions. Sample collections at the station were made twice a week on average.

Aircraft measurements of $\delta^{18}O$ were made over Syowa Station once or twice a month from May 1989 to January 1990, except during the polar night months of June and July. Detailed descriptions of the aircraft measurements have been presented elsewhere (MURAYAMA *et al.*, 1995). As discussed in their paper, air sample collections for the CO_2 analysis were made about every kilometer up to approximately 7 km height. On the other hand, air samples for $\delta^{18}O$ analysis were collected by 550 m*l* Pyrex glass flasks about every 2 km.

It has been indicated that the exchange of oxygen atoms between CO_2 and water in the flasks during the storage influences $\delta^{18}O$ of CO_2 in the sample air (FRANCEY and TANS, 1987; FRIEDLI *et al.*, 1987; MOOK *et al.*, 1983). Therefore, in this study, CO_2 was extracted cryogenically from the air samples at the station within at most 4 days (mostly within 1 day) after their collection. Our extraction procedures were the same as those described by NAKAZAWA *et al.* (1993).

The mass spectrometer used in this study was a Finnigan MAT-δE installed at the National Institute of Polar Research, Tokyo. Detailed descriptions of the isotopic analyses using this mass spectrometer have been presented in NAKAZAWA *et al.* (1993). The N_2O correction for $\delta^{18}O$ data, based on the measured relative ionization efficiencies of CO_2 and N_2O in the mass spectrometer and the N_2O concentration at the South Pole (FERGUSON, 1992), was made by adding +0.31‰. The precision of the analysis for oxygen isotope was estimated to be within 0.07‰ by measuring 70 CO_2 samples extracted from a CO_2-in-air standard gas. In this analysis, the values of $\delta^{13}C$ were also determined. The results for $\delta^{13}C$ will be presented elsewhere.

The air samples were also collected at and over Syowa Station for CO_2, CH_4 and CO concentrations and $\delta^{13}C$ measurements. These were returned to Tohoku University and analyzed using a non-dispersive infrared analyzer and a gas chromatograph. Carbon dioxide extraction and the isotopic analysis were made on the leftover air samples. These samples were stored in the flasks for more than 100 days (more than 400 days in some cases) before the extraction. The measured $\delta^{18}O$ values of these samples were much more scattered than those for the samples extracted within a few days at Syowa Station. From comparisons of $\delta^{18}O$ values between the samples collected simultaneously at the station, it was apparent that $\delta^{18}O$ of the sample stored for >100 days is lower than that of the sample extracted within a few days by more than 1‰. From these results, it is concluded that CO_2 extraction at Syowa Station within a short time after the collection is very important for the measurement of $\delta^{18}O$ at the station.

In this paper, the $^{18}O/^{16}O$ ratios of CO_2 are expressed as deviations from the ratio of the carbonate standard PDB, as follows;

$$\delta^{18}O = [\ (^{18}O/^{16}O)_{sa} / (^{18}O/^{16}O)_{PDB} - 1\ ] \times 1000\ \ (‰), \qquad (1)$$

where the subscripts sa and PDB denote the sample and the PDB standard, respectively. Details of the system and the preparation of standards for our isotopic measurements have been described in NAKAZAWA *et al.* (1993). In this study, the primary standard was CO_2 gas produced by reacting NBS-18 ($\delta^{13}C$ = –5‰ and $\delta^{18}O$ = –23‰ with respect to PDB) with 100% phosphoric acid at 25°C.

FRANCEY *et al.* (1994) have reported on the possibility of systematic difference of the isotopic data between laboratories. Therefore, the intercalibration is likely to be useful for precise comparisons of $\delta^{18}O$ data shown in this paper with those obtained at other laboratories.

3. Results and Discussion

Figure 1 shows the temporal variation of $\delta^{18}O$ of atmospheric CO_2 at Syowa Station during the period of observation. The fitted curve to the measured data is obtained using the technique of NAKAZAWA *et al.* (1991b, 1992). $\delta^{18}O$ shows a prominent seasonal cycle with maximum in summer and minimum in winter. The peak-to-peak amplitude of the cycle is 0.27‰. The seasonal cycle of $\delta^{18}O$ observed at Cape Grim, Tasmania (FRANCEY *et al.*, 1995) is similar to that at Syowa Station; the maximum of the average seasonal cycle at Cape Grim during 1982–1991 calculated from the results of FRANCEY *et al.* (1995) occurs early in January and the minimum late in June with an amplitude of 0.22‰. The annual mean $\delta^{18}O$ value at Syowa Station is about 1.15‰. Annual mean $\delta^{18}O$ values at Cape Grim in 1989 and 1990 calculated from the results of FRANCEY *et al.* (1995) are 0.95 and 1.14‰, respectively. These values are comparable to the mean value at Syowa

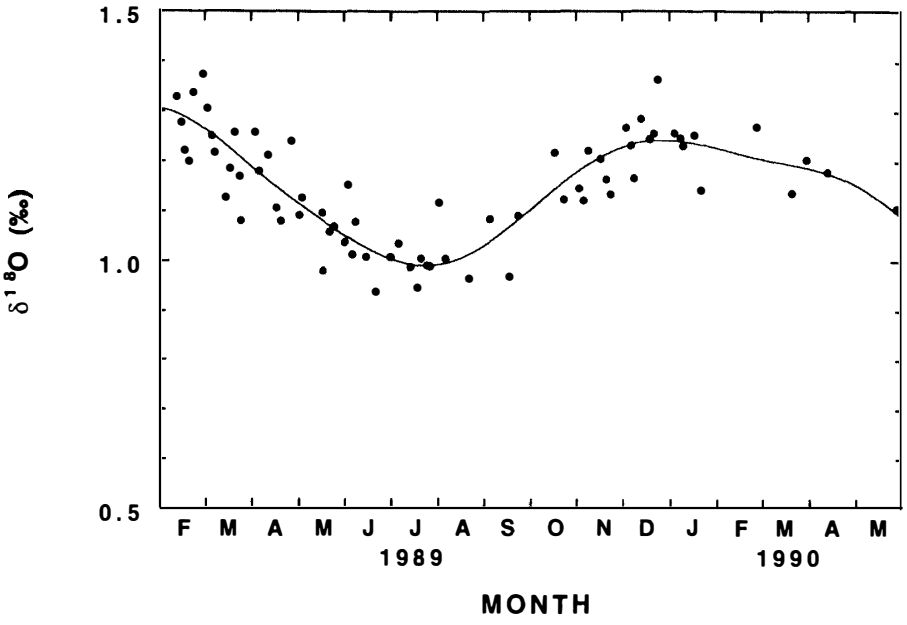

Fig. 1. Temporal variation of $\delta^{18}O$ of atmospheric CO_2 at Syowa Station. Solid circles represent observed $\delta^{18}O$ values and the solid line the best fit curve to the data.

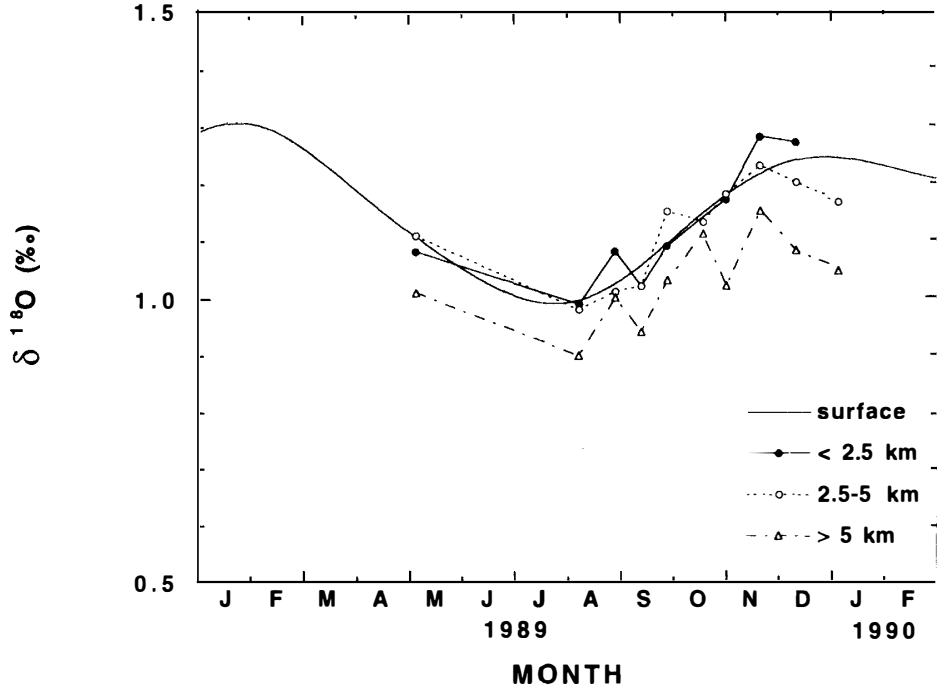

Fig. 2. Temporal variations of $\delta^{18}O$ of atmospheric CO_2 at selected height intervals over Syowa Station.

Station.

The tropospheric $\delta^{18}O$ variations obtained from 10 flights over Syowa Station are shown in Fig. 2, together with the fitted curve for $\delta^{18}O$ at the station. It is seen from this figure that $\delta^{18}O$ shows low values in winter and high values from late spring to early summer throughout the troposphere. The values in the lower and middle troposphere are close to those at the ground surface, but $\delta^{18}O$ in the upper troposphere is slightly lower than those in the lower levels throughout the observed period. The surface value measured on the same day is subtracted from each of the 10 aircraft measurements made at a particular level shown in Fig. 2. These differences are then averaged for each level and plotted in Fig. 3 as a vertical variation in $\delta^{18}O$ relative to the surface. The difference between the surface and the upper troposphere is 0.10‰. A similar vertical profile is also observed over Bass Strait (FRANCEY *et al.*, 1990).

In Figs. 4 and 5, average seasonal cycles and an annual mean vertical profile of the CO_2 concentration over Syowa Station obtained by MURAYAMA *et al.* (1995) are shown respectively. Compared with $\delta^{18}O$ variations (Fig. 2), high CO_2 concentration of the seasonal cycle generally appears during a low $\delta^{18}O$ period, and *vice versa*. However, the maximum and the minimum of the seasonal cycle of the CO_2 concentration appear later by about 2 months than the minimum and the maximum of $\delta^{18}O$, respectively. Such a phase difference is also observed at other sites such as Cape Grim and Mauna Loa (TANS *et al.*, 1986; FRANCEY *et al.*, 1990). Mean vertical profiles of $\delta^{18}O$ and CO_2 (Figs. 3 and 5, respectively) seem to mirror each other. In particular, it is interesting that the vertical gradient is steeper in the upper troposphere than in the lower levels in both profiles.

To interpret the CO_2 variations over Syowa, MURAYAMA *et al.* (1995) have performed

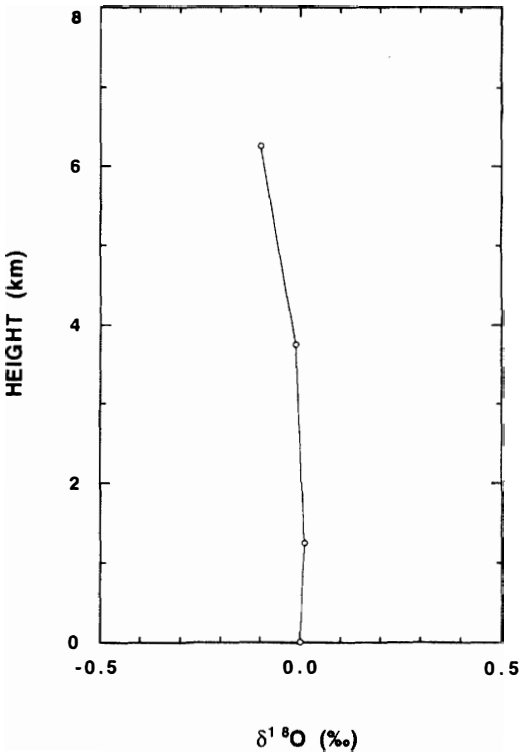

Fig. 3. Average vertical difference of $d^{18}O$ of atmospheric CO_2 from the values at the ground surface over Syowa Station.

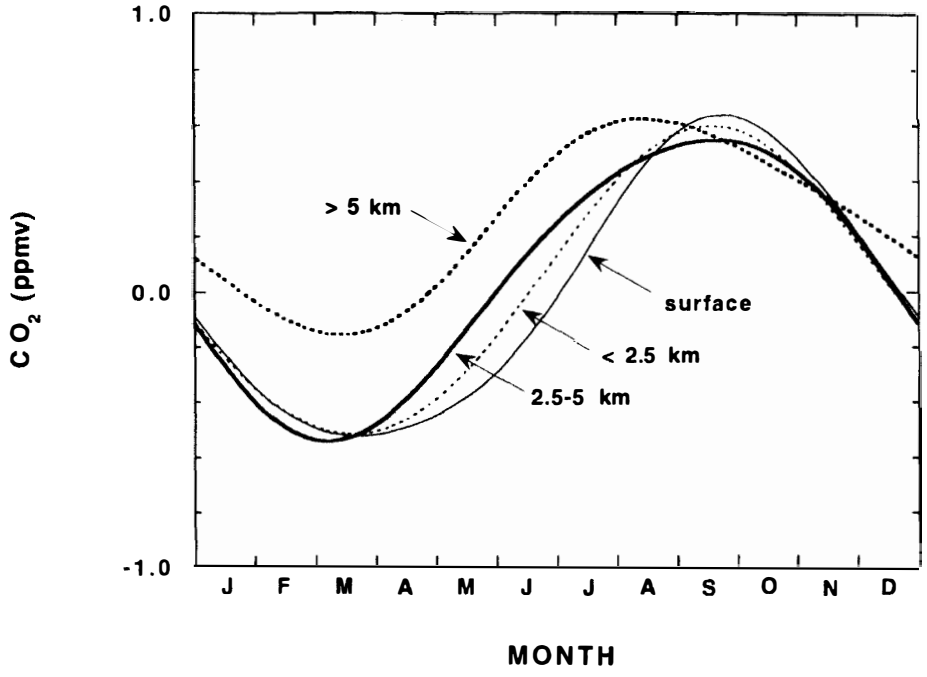

Fig. 4. Average seasonal cycles of the CO_2 concentration at selected height intervals over Syowa Station. Respective values are shown as deviations from the mean value at the surface during January 1983–January 1992.

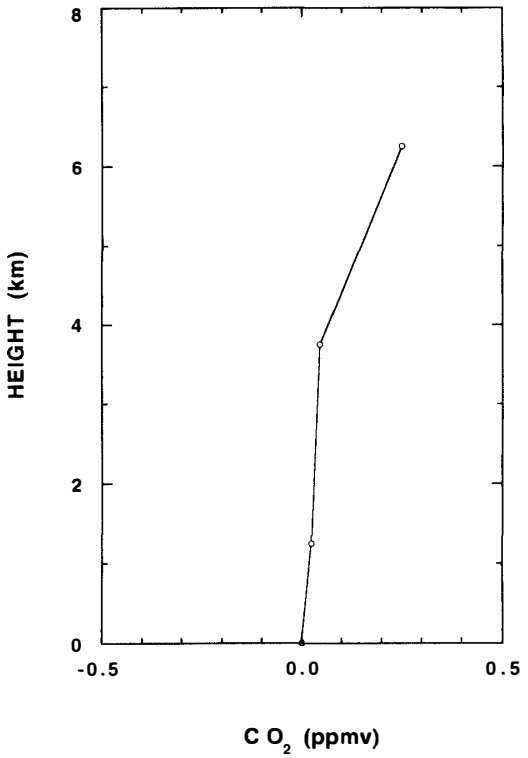

Fig. 5. Annual mean vertical profile of the CO_2 concentration over Syowa Station. Respective values are shown as deviations from the mean value at the surface during January 1983–January 1992.

a 3-D trajectory analysis. From that analysis, it was postulated that northern hemispheric air with relatively high CO_2 concentration is transported to the Antarctic region through the upper troposphere from late fall to winter, while low CO_2 air is transported from the southern hemisphere middle latitudes into the Antarctic region through the lower troposphere in the remaining seasons. FRANCEY *et al.* (1990) reported a latitudinal distribution of $\delta^{18}O$ of CO_2; the mean values of $\delta^{18}O$ at the South Pole (90°S), Cape Grim (41°S), Samoa (14°S), Mauna Loa (20°N) and Barrow (71°N) for 1985–1988 were 0.87, 0.93, 0.62, 0.24 and –1.09 ‰, respectively. These data show that the $\delta^{18}O$ values are relatively constant between southern high and middle latitudes, and decrease northward from southern middle latitudes. In their paper, $\delta^{18}O$ values at respective sites also exhibit strong inter-annual variations. A secular increase or decrease of $\delta^{18}O$ is not observable except at Barrow, where a decreasing trend was observed during 1984–1988.

From the comparison of our data observed at Syowa with these results of FRANCEY *et al.* (1990), the following seasonally dependent processes of atmospheric transport may be thought to be one of the important factors contributing to the variations of $\delta^{18}O$ at Syowa Station. From fall to winter, air with a high CO_2 concentration and low $\delta^{18}O$ is transported from southern low latitudes and/or the northern hemisphere to the Antarctic region through the upper troposphere. As a result, the CO_2 concentration is increased and the $\delta^{18}O$ is decreased at Syowa Station during the period. As described above, the

results by FRANCEY *et al.* (1990) show that the $\delta^{18}O$ value in the Antarctic region is relatively lower than that at southern middle latitudes. Though our $\delta^{18}O$ value at Syowa is close to that of Cape Grim, in fact, the $\delta^{18}O$ at Syowa may also be lower than that at Cape Grim because our data have not yet been intercalibrated with those of FRANCEY *et al.* If this is the case, it may be suggested that air with relatively low CO_2 and high $\delta^{18}O$ is transported from southern middle latitudes to the Antarctic region through the lower troposphere from spring to summer. Consequently, the CO_2 concentration decreases and $\delta^{18}O$ increases at Syowa during these seasons. These processes are also consistent with the vertical distribution of $\delta^{18}O$ over Syowa Station. In this regard, FRANCEY *et al.* (1990) have also suggested that atmospheric transport from the tropics or the northern hemisphere has an influence on the vertical gradient of $\delta^{18}O$ at southern middle latitudes. Compared with $\delta^{18}O$ values of atmospheric CO_2 in local equilibrium with ocean water which depends mainly on sea surface temperature (BOTTINGA and CRAIG, 1969; FARQUHAR *et al.*, 1993), the $\delta^{18}O$ values observed at southern middle and high latitudes are lower. Especially at southern high latitudes, the observed values are depleted by more than 3‰ relative to the equilibrium values. Therefore, these facts are also likely to reflect the transport of air with low $\delta^{18}O$ from southern low latitudes and/or the northern hemisphere to these latitudes.

However, such seasonally dependent atmospheric transport processes cannot explain the phase difference in the seasonal cycle between the CO_2 concentration and $\delta^{18}O$ of CO_2 at Syowa. In this connection, TANS *et al.* (1986) have sought an explanation for the seasonal variation and mean level of $\delta^{18}O$ in the Cape Grim CO_2. In their paper, they have concluded that the exchange with leaf water seems to offer the best prospect of explaining the Cape Grim $\delta^{18}O$ and suggested that some ocean exchange may be related to the phase difference observed at Cape Grim. In case of Syowa, a local effect due to land biosphere is unlikely, since the surface around the station is covered with snow and ice except in summer, when naked rocks with some lichen growth are exposed. Also, since the amplitude of the seasonal cycle of sea surface temperature is smaller around Antarctica than at southern middle latitudes, the effect of ocean exchange on the $\delta^{18}O$ variation is thought to be very small. Therefore, it is suggested that $\delta^{18}O$ variations at Syowa may be caused mainly by atmospheric transport processes from different latitudes and may also be affected by other factors such as exchange with leaf and soil water at southern lower latitudes and exchange of oxygen between ocean water and CO_2 at the sea surface of the Southern Ocean. There might be other factors which we have not yet considered.

For a better understanding of $\delta^{18}O$ of CO_2 in the Antarctic region, further systematic and more precise measurements of $\delta^{18}O$ in the southern hemisphere are required. Intercalibration between laboratories is also very important. The knowledge gained will contribute to an increased understanding of the global carbon and hydrological cycles, as well as of the atmospheric transport processes.

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