

Salt Origin in the Wright Valley, Antarctica

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南極ライト谷における塩類の起源

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要旨: 南極, 上部ライト谷における塩類の分布および硫黄の安定同位体組成を調べた。

ドンファン湖盆における塩収支の計算から, ドンファン池の塩化カルシウム型の塩類の起源は, 主として塩化カルシウム型の地下水であり, また, 硫酸イオンとカルシウムイオンの起源については, 地下水と融氷水以外の供給源, たとえば風送塩として運ばれてくる硫酸塩などを考慮に入れなくてはならないことが明らかとなった。この塩収支の計算から, ドンファン池の年齢は, 9 000-37 000 年より新しいことが示唆された。

硫黄の同位体比からは, ラビリンス地域とドンファン湖盆の硫酸イオンの起源を明らかにすることはできなかった。バンダ湖盆の硫酸イオンについては, かつて谷がフィヨルドとなった時に残された海水起源の硫酸塩が寄与していると推察された。

Abstract: Salt distribution and stable isotope composition of sulfur have been investigated in the upper Wright Valley area, Antarctica. From the discussion on the mass balance of chemical constituents in the Don Juan basin, it is clear that calcium chloride-rich groundwater is an important source for the pond water, and another sources of sulfate and calcium ions other than groundwater and stream water should be considered. The age of Don Juan Pond was calculated to be younger than 9 000–37 000 years.

The isotope composition of sulfur does not indicate the origin of sulfate ions at the Labyrinth area and the Don Juan basin. For the Vanda basin, it is supposed that the sulfate origin is sulfate minerals which were formed from seawater on the slope of the valley during the valley was fjord.

1. Introduction

There are many saline lakes in the Dry Valleys area of south Victoria Land, Antarctica. The chemical compositions of the lake waters and their deposits are so characteristic that the origin of salts and the formation process of the lakes have been studied very actively. The analytical data of lake waters in the Dry Valleys area were reported by TORII *et al.* (1975). Lakes in the Wright Valley in the area are one of the most interesting ones because of the unique chemical composition of calcium chloride.

Figure 1 shows the major chemical compositions of typical saline waters such as seawater and the waters in the Dry Valleys area, Great Salt Lake in Utah and Dead Sea in Israel. This figure indicates that the chemical compositions of Lakes Bonney and Fryxell in the Taylor Valley of the Dry Valleys area are similar to that of seawater. With increasing concentration of seawater under normal temperature, first calcium

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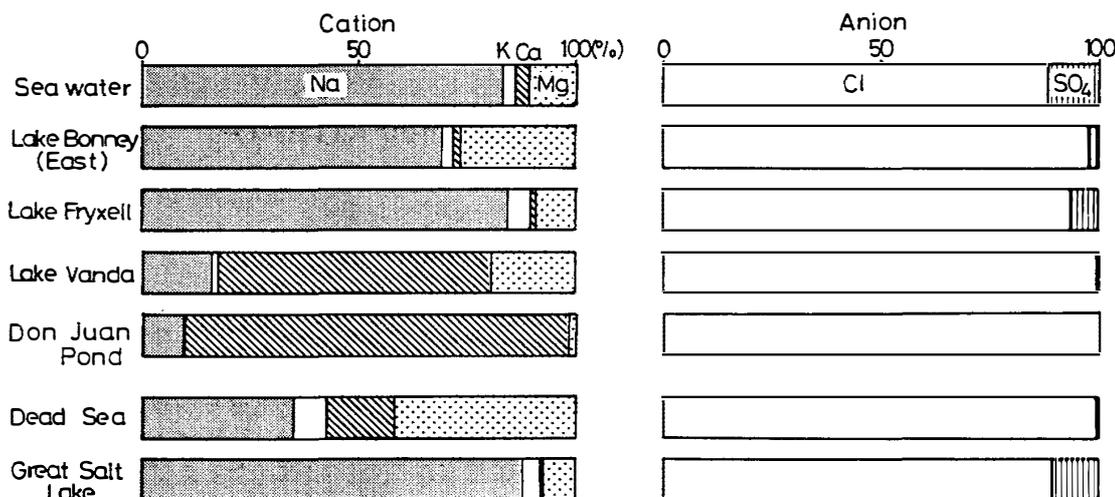


Fig. 1. Chemical composition of seawater and lake waters in the Dry Valleys area, Dead Sea and Great Salt Lake.

carbonate and calcium sulfate are precipitated, next sodium chloride is precipitated and magnesium concentration in the residual solution increases. The chemical composition of Lake Bonney water seems to be explained by the concentration process of seawater (BOSWELL *et al.*, 1967). The chemical compositions of Great Salt Lake and Dead Sea waters are also formed by the concentration process of seawater. However, the chemical compositions of Lake Vanda and Don Juan Pond waters in the Wright Valley are completely different from those in the other valleys.

The formation process of the calcium chloride waters in the Wright Valley has been discussed by many researchers, and the following salt origins and the formation processes have been proposed; (1) concentration of seawater under frigid conditions, (2) chemical weathering of rocks, (3) sea spray, (4) aerosol, (5) groundwater and (6) hydrothermal water. In order to clarify the salt origin, the present authors already estimated the amounts of salts existing in the Vanda basin and the Don Juan basin from the analytical results of their sediments (TOMIYAMA and KITANO, 1982; TOMIYAMA *et al.*, 1983). They proposed that the calcium chloride-rich groundwater is an important salt source in the Wright Valley. However, the concentration of sulfate ions in the groundwater is too small to satisfy a large amount of sulfate in the Vanda basin and the Don Juan basin. Therefore, another source for sulfate ions in the Wright Valley must be considered.

This paper will describe the distribution of salts at the Labyrinth area of the upper Wright Valley, and will discuss on the mass balance of salts in the Don Juan basin and the salt origin in the Wright Valley.

2. Samples and Analyses

Samples: Salt deposits were collected around small saline ponds at the Labyrinth area in the 1983–1984 austral summer season. Figure 2 shows the locations of the saline ponds, from which the salt samples were taken, at the Labyrinth area. Water samples were also collected from the saline ponds and the chemical compositions of the

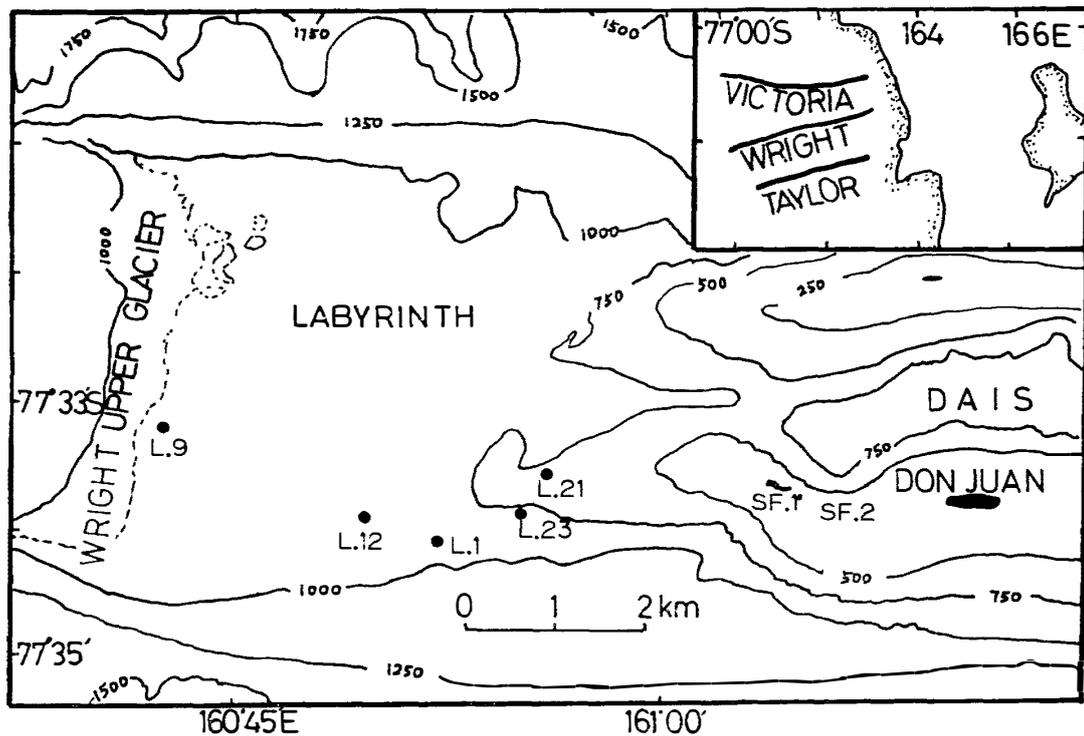


Fig. 2. Sampling location at the Labyrinth area.

water samples were analyzed by TORII *et al.* (private communication).

Analyses: Mineral species were identified by the powdered X-ray diffractometry. For the analyses of chemical composition of salt samples, the samples were dissolved with deionized water and 1 M acetic acid successively. Na^+ , K^+ , Ca^{2+} and Mg^{2+} in the leached solution were determined by using a Perkin Elmer 503 atomic absorption spectrometer. Sulfate and chloride ions were determined colorimetrically by using barium chromate-acid suspension and mercury thiocyanate, respectively. Stable isotope ratio of sulfur ($^{34}\text{S}/^{32}\text{S}$) was determined with a Varian Mat CH 7 mass spectrometer. This ratio is expressed in permil deviation ($\delta^{34}\text{S}$) from standard (troilite sulfur in the Canyon Diablo meteorite).

3. Results and Discussion

3.1. Salt distribution at the Labyrinth area

The Labyrinth area is located at an elevation of about 900 m on the upper Wright Valley, and shows a very complicated topographical feature. Small saline ponds are found in this area and the chemical compositions of those pond waters were reported by TORII *et al.* (private communication). It was reported that the salinities of the ponds vary from 15 mg/l to 100 g/l with varied chemical compositions.

Figures 3 and 4 show the weight proportions (%) of cations and anions of salts and pond waters at the Labyrinth area, respectively, where the order of samples in the figures is expressed geographically from west to east at the Labyrinth area. The proportions of cations and anions of the pond waters were calculated from the analytical data presented by TORII *et al.* (private communication). The cation composition of

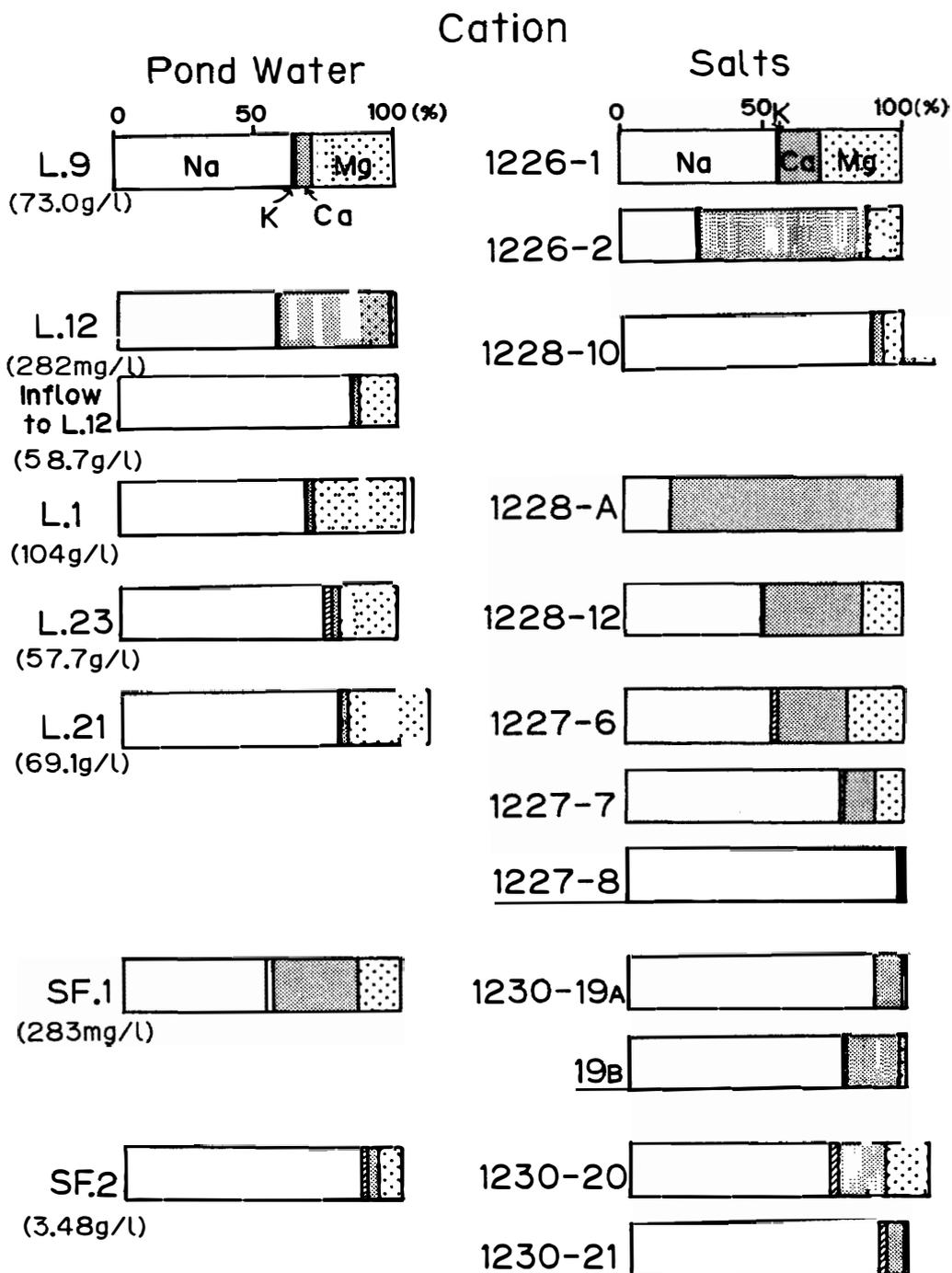


Fig. 3. Proportions of cations in pond waters and salts collected from the Labyrinth area (weight %). The salinities of pond waters are represented in parentheses.

the salt samples includes the water soluble fraction and also the acetic acid soluble fraction which is carbonate precipitated from the pond water. The salinity of pond waters is shown under the name of pond (Fig. 3).

L. 9 is located at 100 m east from the Wright Upper Glacier. Therefore, the pond water itself is supposed to be mainly glacial meltwater. The chemical compositions of the pond water and the water soluble and acetic acid soluble salts in L. 9 are quite

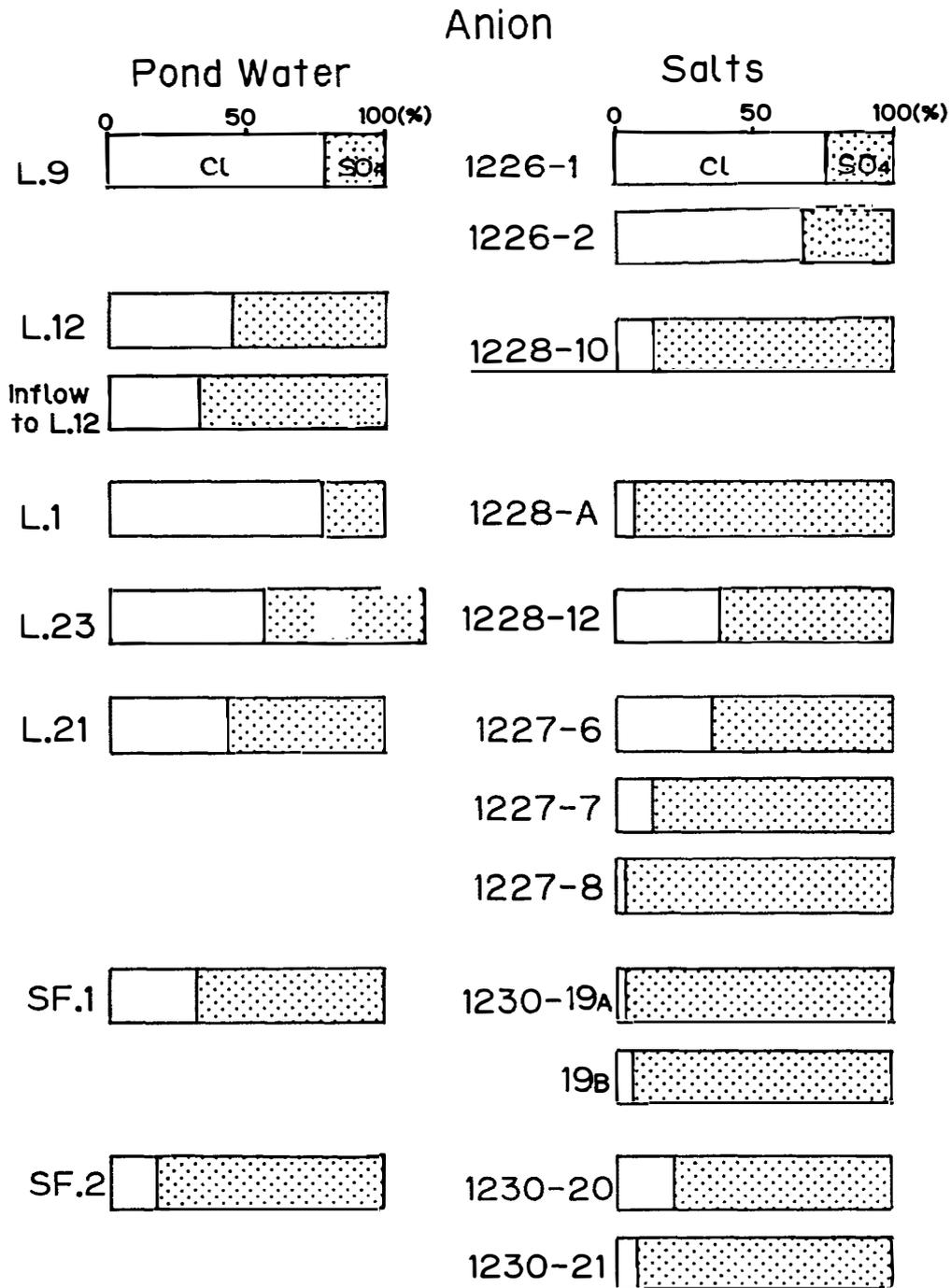


Fig. 4. Proportions of anions in pond waters and salts collected from the Labyrinth area (weight %).

similar to that of seawater. This result suggests that the salts contained in glacial ice are mostly marinogenic, although the chemical composition of glacial ice in this area is not available.

The composition of cations indicates that the proportion of calcium ions is large in fresh waters at L. 12 and SF. 1, whereas those in saline pond waters are very small because of the deposition of calcium salts (Fig. 3). The proportions of sodium ions

in fresh and saline pond waters and also their salt deposits seem to increase as the sampling location goes to seaward. Figure 4 indicates that the proportions of sulfate ions in anion composition in fresh and saline waters and their salts also increase as the sampling location goes to east. It is supposed that sulfate and sodium ions are supplied by glacial meltwater. The origin of sulfate ions will be discussed later.

3.2. Mass balance of salts in the Don Juan basin

The total amounts of individual chemical constituents in the Don Juan basin were estimated previously from the observation on the distribution of salts in the area (TOMIYAMA *et al.*, 1983). The estimated total amounts indicated that large amounts of calcium and chloride ions exist in the basin. It was supposed that the calcium chloride-rich groundwater which had been reported by HARRIS and CARTWRIGHT (1981) is very important as a salt source in the basin. They observed a stream flow in summer season from rock glacier located at the western end of the basin. The inflow continued for 25 days in January 1975 and only 16 days in January 1976. The stream inflow supplies chemical elements to the basin every summer. Table 1 shows the chemical composition of the stream water.

Here, the amounts of salts supplied by groundwater and stream water and calculated in the same way as reported in the previous paper (TOMIYAMA and KITANO, 1984):

$$\begin{cases} a_1X + b_1Y = K_1 \\ a_2X + b_2Y = K_2 \end{cases}$$

where a_1 and a_2 are the concentrations of chloride and magnesium ions in the groundwater respectively, b_1 and b_2 are the concentrations of chloride and magnesium ions in the stream water respectively. X is the amount of the groundwater, Y is the amount of the stream water, and K_1 and K_2 are the total amounts of chlorine and magnesium respectively in the Don Juan basin. The data used for this calculation are shown in Table 1.

The results of the calculation are as follows:

$$X = 2.0 \times 10^6 \text{ t}, \quad \text{and} \quad Y = 3.7 \times 10^7 \text{ t}$$

By using these values, the amounts of individual chemical constituents supplied through

Table 1. Calculated values of total amounts of individual chemical constituents existing in Don Juan basin and chemical compositions of groundwater and stream water in the basin.

	Total amounts of chemical constituent in the basin (t)	Groundwater* (g/kg)	Stream water* (mg/kg)
Ca ²⁺	2.70×10^5	65.6	28
Na ⁺	1.60×10^4	6.35	42
Mg ²⁺	2.69×10^3	0.93	22
Cl ⁻	2.36×10^5	114	140
SO ₄ ²⁻	3.12×10^5	0.085	60

* Data from HARRIS and CARTWRIGHT (1981).

Table 2. Comparison of total amounts of individual chemical constituents in the Don Juan basin with the calculated values by using X and Y values.

	Total amount of chemical constituent (t)	Calculated value		Total (t)
		from groundwater ($X=2.0 \times 10^6$) (t)	from stream water ($Y=3.7 \times 10^7$) (t)	
Ca ²⁺	2.70×10^5	1.3×10^5	1.0×10^3	$1.3_1 \times 10^5$
Na ⁺	1.60×10^4	1.3×10^4	1.6×10^3	$1.4_4 \times 10^4$
Mg ²⁺	2.69×10^3	1.8×10^3	8.1×10^2	$2.6_0 \times 10^3$
Cl ⁻	2.36×10^5	2.3×10^5	7.0×10^3	$2.3_7 \times 10^5$
SO ₄ ²⁻	3.12×10^5	1.7×10^2	3.0×10^3	$3.1_7 \times 10^3$

groundwater and stream water are calculated, as shown in Table 2. The composition of calculated values with observed values indicates that the calculated amount of sulfate ions is too small to explain the observed value. Therefore, another sulfate source other than groundwater and stream water must be considered.

HARRIS and CARTWRIGHT (1981) reported that the discharge of the groundwater to the Don Juan Pond is 30 m³/day throughout a year. Using this discharge rate and the X value described above, the discharge of groundwater to the basin is assumed to have started about 180 years ago. The age of Don Juan Pond as a calcium chloride pond is assumed to be the order of 10² years.

On the other hand, only two estimated values are available for the inflow flux of stream water. HARRIS and CARTWRIGHT (1981) reported that the inflow of stream water was estimated to be 4000 m³ in 1975 and 1200 m³ in 1976 from the hydrological view. If the present annual stream inflow is 1000 to 4000 m³, the history of the stream flow to the Don Juan basin is calculated to be 9 000 to 37 000 years by using the Y value described above. This age does not give the age of Don Juan Pond with a chemical composition of calcium chloride. The present water depth of Don Juan Pond is only about 10–20 cm, but the old shoreline is found at about 20 m high in the slope of the basin. Thus, it is assumed that the past stream inflow was greatly larger than the present as a result of climatic change. Therefore, the age of the Don Juan Pond as a glacial pond is supposed to be greatly younger than 9 000–37 000 years. At least, it is assumed that the chemical composition of water in the early stage of the Don Juan Pond was not calcium chloride type and the supply of the calcium chloride-rich groundwater has started only a few hundreds years ago.

3.3. Source of sulfate ions

For the mass balance of salts in both Lake Vanda and the Don Juan basin, the origin for a large amount of sulfate ions must be considered seriously (TOMIYAMA and KITANO, 1984). The stable isotope ratio of sulfur in sulfate salt may give a useful information for the sulfate origin. NAKAI *et al.* (1975) reported that the $\delta^{34}\text{S}$ values of gypsum deposits from Lake Vanda and the lake area are +18.9 to +20.9‰. Thus, they insisted that the origin of sulfate ions in Lake Vanda is seawater. The $\delta^{34}\text{S}$ values of sulfate from the Don Juan basin were measured by NAKAI (private communication) to be +31.8 to +37.5‰. Such high values of $\delta^{34}\text{S}$ were considered to be attributed to sulfate reduction by bacteria.

Table 3. $\delta^{34}\text{S}$ values of sulfate in the upper Wright Valley.

Sample no.	Sampling place	Evaporite minerals	$\delta^{34}\text{S}$
1226-1	L. 9	Hal., Cal.	9.6
1226-2	L. 9	Hal., Gyp.	9.3
1226-5	L. 9	Gyp.	12.6
1227-6	L. 21	The.	12.9
1227-7	L. 21	The., Cal.	13.4
1228-10	L. 12	Hal., The.	12.3
1228-inflow	L. 12		13.0
1228-12	L. 23	Gyp., Cal.,	12.5
1230-19A	SF. 1	The., Gyp., Hal.	13.0
1230-20	SF. 2	Hal., Gyp.	12.8
1231-26	Don Juan Pond	The.	30.5
1231-28	East of Don Juan	Hal., Cal., Gyp.	13.5
1230-30	SF. 3	Gyp., Cal.	14.8
1231-32	South of L. Vanda	Gyp.	17.2
Onyx	Onyx Riv.	Gyp.	19.2

Cal.: calcite, Hal.: halite, Gyp.: gypsum, The.: thenardite

Table 3 shows the stable isotope ratios of sulfur in sulfate minerals from the eastern end of the Wright Upper Glacier to Lake Vanda. The $\delta^{34}\text{S}$ values of sulfate minerals taken from station L. 9, near the end of the glacier, are +9.3 to +9.6‰. The $\delta^{34}\text{S}$ values of sulfate minerals at the Labyrinth area except for the L. 9 area are relatively uniform, +12.6 to +13.4‰. And that in the Don Juan basin is +30.5‰. The $\delta^{34}\text{S}$ values of sulfate minerals from the east part of the basin are +13.5 to +14.8‰, and those from the Lake Vanda area are +17.2 and +19.2‰.

The low values in L. 9 are supposed to be derived from the atmospheric sulfate existing in glacial ice. As described in the previous section, sulfate ions are assumed to be supplied to glacial meltwater during meltwater flows through the Labyrinth area. From $\delta^{34}\text{S}$ values, it is considered that $\delta^{34}\text{S}$ value in sulfate ions supplied to the meltwater having $\delta^{34}\text{S}$ values of +9.3 to +9.6‰ may be a little larger than +12.6 to +13.4‰.

Here, two sulfate sources at the Labyrinth area are expected: One is the supply through chemical weathering of parent rocks. Ferrar dolerite is a main basement rock in this area. The sulfur content of basalt is reported to be 70–9400 ppm (SCHNEIDER, 1975), although that of Ferrar dolerite has not been available. Therefore, the sulfur content of the parent rock seems to be enough for the source of sulfate ions. However, since the $\delta^{34}\text{S}$ values of basalt are low in the range of –10 to +10‰ (SCHNEIDER, 1975), the sulfur in basalt cannot be considered as the source of sulfate ions.

Another sulfate source is the supply of sulfate salts transported by wind. NISHIYAMA (1977) reported that calcium sulfate, sodium sulfate and calcium carbonate exist in the slope of the Taylor Valley at an elevation of about 1000 m. Such salts found in the high altitude are considered to be transported by wind from the basin and/or the Ross Sea. WELLMAN and WILSON (1963) observed a large amount of salts on Ross Sea ice, the major component of which is sodium sulfate. The salt, which is

precipitated from seawater through the concentration process under frigid conditions, is mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$). The salts found on sea ice are supposed to be derived from seawater. The existence of thick mirabilite layers was reported in Hobbs Glacier, Miers Valley (DORT and DORT, 1970) and Cape Barne (NAKAI and MIZUTANI, 1977). That is, a large amount of sulfate minerals is found around the McMurdo Sound region. The transportation of these sulfate salts by wind from the Ross Sea to the Wright Valley is quite conceivable. However, $\delta^{34}\text{S}$ value of sulfate transported from the Ross Sea is assumed to be similar to that of the present seawater, $+20\text{‰}$. If the dry fallout derived from seawater is one of the sulfate sources at the Labyrinth area, the $\delta^{34}\text{S}$ values of the salts observed in the area should be more scattered, owing to different mixing ratios. But, the observation in Table 3 shows that the $\delta^{34}\text{S}$ values at the Labyrinth area are constant. Moreover, if sodium sulfate as the dry fallout is the significant source of sulfate ions, the proportion of calcium and sodium contents shown in Table 2 is difficult to be explained. From these discussion, it should be considered that the origin of sulfate ions at the Labyrinth area is still in question.

It is considered that sulfate-rich water should be sometimes supplied to the Don Juan basin. The process for the high $\delta^{34}\text{S}$ values in sulfate of the Don Juan basin is also a problem to be solved. If it was due to the fractionation by bacterial reduction of sulfate as proposed by NAKAI *et al.* (1975), a large amount of hydrogen sulfide with low $\delta^{34}\text{S}$ value should generate and escape from the pond water, and iron sulfides should be found in the sediment. But, analytical results do not show any evidence of the existence of iron sulfides in the Don Juan basin. The $\delta^{34}\text{S}$ values of sulfate ions in calcium chloride-rich groundwater were reported to be very high, $+39.1\text{‰}$ to $+48.8\text{‰}$, in Lake Vanda. Since the origin of the groundwater in the Don Juan basin is considered to be the same as that in Lake Vanda, the $\delta^{34}\text{S}$ values of sulfate ions of the groundwater in the Don Juan basin are assumed to be high. However, the amount of sulfate ions supplied through the groundwater is no more than 0.5% of the total amount of sulfate ions in the basin (Table 2). Therefore, the supply of heavy sulfate ions through the groundwater is not quantitatively significant for the explanation of the sulfur isotope composition. The quantitative discussion about the mechanisms for high $\delta^{34}\text{S}$ values in the Don Juan basin cannot be shown in this paper, though two possibilities for the formation processes of the heavy sulfate ions are suggested; (a) heavy sulfate ion formed through the fractionation by bacterial reduction of sulfate and (b) heavy sulfate ions supplied through groundwater discharge.

The $\delta^{34}\text{S}$ values in sulfate from the sediment of Lake Vanda were the same value as that of seawater (NAKAI *et al.*, 1975). The Wright Valley was once covered with seawater to be fjord after the main valley had been formed through glaciation (VUCETICH and TOPPING, 1972; BRADY, 1974). But the mass balance of salts from the observed data does not support the idea that the salts in Lake Vanda was originated from seawater (TOMIYAMA and KITANO, 1984). From these consideration, it is assumed that sulfate minerals were deposited from seawater on the slope of the valley during the valley was fjord and later the sulfate minerals were transported to the Vanda basin through the glacial meltwater in summer season.

3.4. Salt origin in the Wright Valley

From the distribution of salts reported in the present and previous papers (TOMIYAMA and KITANO, 1982, 1984; TOMIYAMA *et al.*, 1983), the salt deposits in the Wright Valley area are classified into three types from chemical composition.

(1) $[\text{CaSO}_4 \gg \text{CaCl}_2 > \text{NaCl} > \text{MgCl}_2, \text{KCl}]$ type: This type of deposit is found in the surface bottom sediment of Lake Vanda and in the sediment of the Don Juan basin. Calcium chloride-rich groundwater and glacial meltwater are major sources for the salts. The calcium chloride-rich groundwater is assumed to be discharged from the deep part in the basin. Also glacial meltwater flows into the basin with some chemical elements dissolved from rocks in summer season. And sometimes, a large amount of water might have transported sulfate ions to the basin from the upper part of the Wright Valley and the slope of the valley. Calcium sulfate should be precipitated from the lake water.

(2) $[\text{CaCl}_2 \gg \text{NaCl} > \text{MgCl}_2, \text{CaSO}_4]$ type: This type of deposit is found in the deep sediments of Lake Vanda and the Don Juan basin, and in the sediment from the western part of Lake Vanda. For the formation of this type of deposit, calcium chloride-rich groundwater seems to be essentially important.

(3) $[\text{Na}_2\text{SO}_4 \gg \text{CaSO}_4, \text{NaCl}, \text{MgCl}_2]$ type: This type of deposit is found in the surface salt sediment of the Labyrinth area. The salts are formed through the evaporation of glacial meltwater, to which water soluble components of surrounding rocks and salts are discharged. The origins of the salts are aerosol, sea spray, dry fallout such as sodium sulfate and calcium sulfate, and chemical constituents from chemical weathering of rocks. The quantitative contributions of those sources are not clear at the Labyrinth area.

The characteristic salt deposits in the Wright Valley is calcium chloride formed from calcium chloride-rich water. Calcium chloride-rich water has been found only in very special and limited area such as deep groundwaters (deeper than 1000 m), and oil field waters (FRITZ and FRAPE, 1982; GRAF, 1978; SKINNER *et al.*, 1967). Calcium chloride type groundwater is important in the Dry Valleys area for the formation of calcium chloride type pond water. Calcium chloride rich water is believed to have developed through the water-rock interaction under the ground. The existence of this type water in the Dry Valleys area is expected to give important informations on environmental situations under the ground in the area.

4. Summary

Salt distribution has been investigated at the Labyrinth area of the upper Wright Valley. The chemical compositions of salts are varied corresponding to the salt origins. The proportions of sodium ions in cations and of sulfate ions in anions increase as the sampling station goes to seaward. This trend is attributed to that the origins of salts at the Labyrinth area are dry fallout and chemical constituents from chemical weathering of rocks.

In the mass balance of chemical constituents in the Don Juan basin, the another source of sulfate ions and calcium ions and calcium ions other than groundwater and stream water should be considered. The isotope composition of sulfur does not

indicate the origin of sulfate ions in the Don Juan basin. It is supposed that a large amount of sulfate ions was supplied to the basin in the past. From the isotope data of sulfur in the Vanda basin, the sulfate origin seems to be sulfate minerals which were formed from seawater on the slope of the valley during the valley was fjord.

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