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Isotope studies of inner snow layers in a temperate region

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Abstract:

Post-depositional change of isotope concentration was investigated in a temperate snow pack. Daily snow pit studies were carried out at Moshiri, Hokkaido, for about a week in April 1998. During this time the snow layers were at the melting point. Isotope analysis was conducted on snow particles and the liquid water in between the particles every 5 cm in depth, and on the precipitation and the runoff water (from the bottom of the snow pack) collected during the observation period. It was found that the oxygen isotope concentration in the liquid part was smaller by about 2‰ than the solid part of the wet snow. The difference between the isotope concentrations of the liquid and the solid was low in the surface snow layer, but increased at the bottom. The results suggested that isotope fractionation took place between the liquid and the solid, when liquid water (formed by melting at the snow surface layer) flowed groundwards. Copyright © 2002 John Wiley & Sons, Ltd.

KEY WORDS isotope fractionation; ice; pore water; temperate snow pack

INTRODUCTION

Ice core studies are one of the most promising methods of reconstructing the past climate and environment (e.g. Shoji and Langway, 1989). In particular, isotope concentrations in polar ice cores have been used as indicators of past temperatures (e.g. Robin, 1983).

Such studies have been made recently not only with polar ice cores but also with ice cores from glaciers at low and middle latitudes (Thompson *et al.*, 1988; Aizen *et al.*, 1996). Snow and ice cores from temperate regions, however, have possibly been subjected to a melt and/or freeze after the snow deposition at the surface, due to the intensity of solar radiation. Isotope concentrations would undergo post-depositional changes at those sites. In attempting to reconstruct the past climate, it is therefore important to have a qualitative understanding of the post-depositional changes in the isotope concentration of wet snow.

Several studies have investigated the isotope concentration of snow packs during melt seasons. For instance, Arnason *et al.* (1973) used deuterium and tritium as tracers to monitor the movement of water in snow packs and temperate glaciers. Krouse *et al.* (1977) reported that there was no significant change over time of the isotopic vertical profile in snow packs in the dry snow zone or superimposed ice zone, but that the profile was homogenized in wet snow packs.

It was reported by Suzuki (1993) that the isotope concentration of the total snow pack and of the discharge water increased as the melting season progressed. Nakawo *et al.* (1999), moreover, indicated that the isotope concentration increased with time in shallow layers, but decreased in deep layers of a snow pack. These investigations, however, did not measure the isotope concentrations in the solid (snow particles) and the liquid (pore water) components of a snow pack. The present paper investigates the fractionation process by examining the temporal change of isotope concentrations in the two phases.

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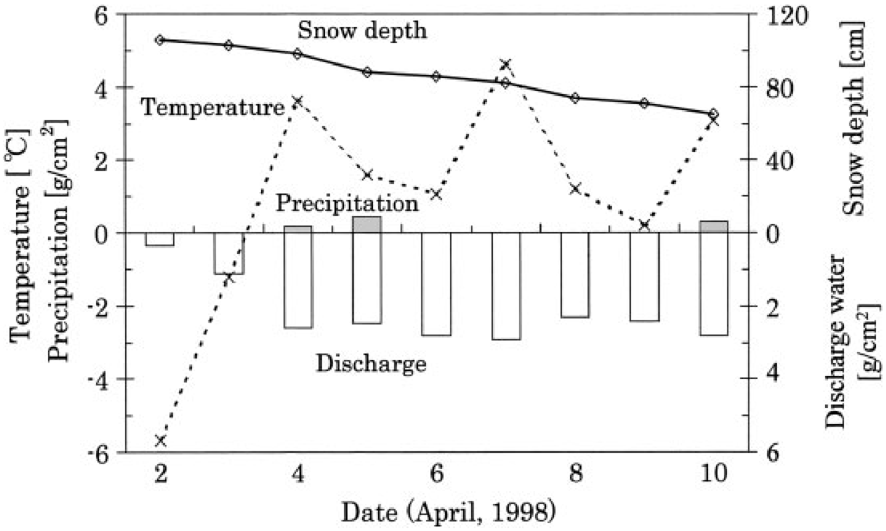

Figure 1. Air temperature, snow depth, daily precipitation and daily discharge during 2 April to 10 April 1998

OBSERVATIONS

Field observations were carried out at Moshiri, northern Hokkaido, Japan, between February and April 1998. The data used in the present analysis were obtained between 29 March and 10 April, when snow temperature was 0 °C throughout the snow pack. Snow pit studies, which included stratigraphic observation and measurement of snow density, snow temperature and water content, were carried out daily at 10 a.m. Each day, 5 cm depth snow samples were collected from the snow surface down to ground level at 5 cm intervals. The solid (snow particles) and liquid (water-filled pores) components of each snow sample were separated using centrifugation. Separated snow samples, however, contained a small amount of liquid water, and this water content was measured using the calorimetric method (Akitaya, 1978). Snow particles from each sample were photographed for later analysis of the particle size distribution.

Precipitation was collected twice a day, morning and evening, using a pan of 30 cm diameter and approximately 10 cm depth. The only rainfall occurred between 21:00 on 4 April and 7:00 on 5 April, and after 18:00 on 10 April. No snowfall was observed during the analysis period. A lysimeter located at the snow–ground interface sampled discharge water from the snow pack twice daily, morning and evening.

These samples were subjected to isotopic analysis for δ^{18}O and δD. The analytical errors for δ^{18}O and δD were ±0·1‰ and ±1·0‰ respectively.

Figure 1 shows the variation of daily mean air temperature, snow depth, precipitation and discharge between 2 April and 10 April, 1998. Snow depth (solid line) decreased by about 40 cm during the period, which was approximately 200 mm water equivalent (w.e.). After 3 April, air temperature (broken line) rose above 0 °C and daily discharge (white bar) increased to an average 20 or 30 mm w.e.

RESULTS

Isotope concentration of snow particles and pore water

Snow samples were separated into liquid water and snow particles, although there was a small amount of liquid water that could not be entirely removed from the snow portion. The isotope concentration in solid snow particles without any liquid water δ'_g was hence calculated by

$$\delta'_g = (\delta_g - c\delta_l)/(1 - c) \quad (1)$$

where c is the water content in the separated snow sample, and δ_g and δ_l represent the isotope concentrations in the separated snow sample and water samples respectively.

Figure 2 shows vertical profiles of the isotope concentration in the solid δ'_g and the liquid δ_l. In the lower layer, the isotope concentration of the liquid water (dotted line) is about 2‰ lower than the concentration of the solid phase (solid line). The difference in isotope concentration between the liquid and the solid is small near the surface, but increases towards the bottom.

Isotope concentration of precipitation, discharge water and the total snow pack

The isotope concentration of the total snow pack δ_s (including both the solid and the liquid phases) can be expressed by

$$\delta_s = \sum(W_i \delta_i)/W \qquad (2)$$

where δ_i is isotope concentration in each snow layer, and W and W_i are the masses of the total snow pack and of each corresponding snow layer respectively. The suffix i indicates the respective layer number, numbered upwards from the ground to the surface. The temporal change of δ_s is plotted in Figure 3. It increased over time, however it was not due to precipitation from the night of 4 April to the morning of 5 April. The precipitation amount was 6·4 mm and the average oxygen isotope concentration was −6·61‰.

The oxygen isotope concentration in the discharge also increased with time, as did δ_s (the isotope concentration in snow), although the former was less pronounced than the latter (Figure 3). Suzuki (1993) also reported that both oxygen isotope concentrations increase over time. The difference in isotope concentration between the snow and the discharge water decreased over time. During the period 2 April to 10 April, the snow pack lost 18·5 g cm^{-2} of mass and the discharge water collected by the lysimeter was 17·4 g cm^{-2}. Thus the collection rate was 94%, and this difference is statistically insignificant.

A plot of δD against δ^{18}O is called aδD–δ^{18}O diagram, and is usually used for examining fractionation processes. Figure 4 shows the δD–δ^{18}O diagram for the precipitation between February and April 1998 (Figure 4a) and for the total snow from 2 April to 10 April (Figure 4b). The regression line in Figure 4a represents the relationship

$$\delta D = 8{\cdot}0\,\delta^{18}O + 20{\cdot}9 \qquad (3)$$

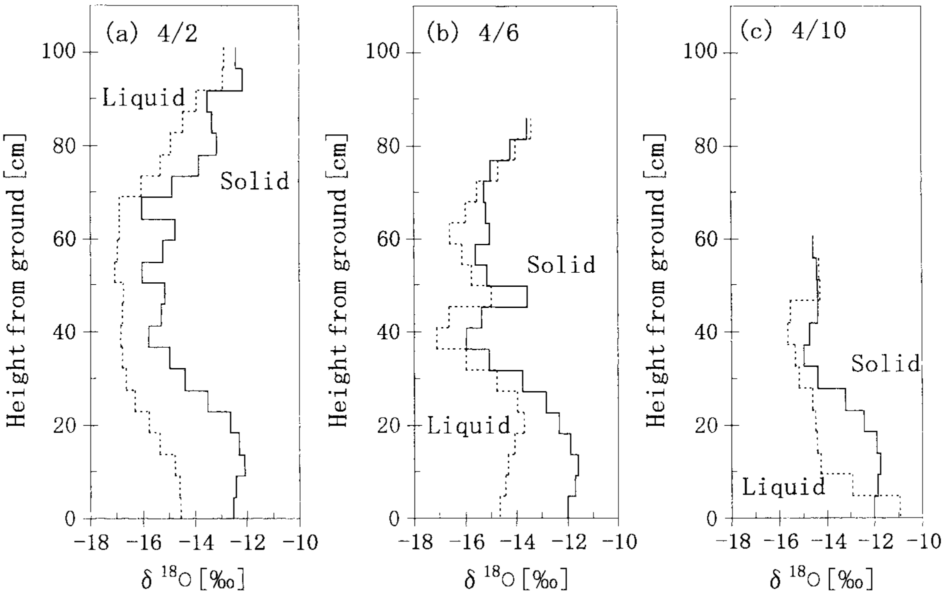

Figure 2. The vertical profiles of δ^{18}O in the solid (solid line) and the liquid (broken line) on (a) 2 April, (b) 6 April, and (c) 10 April

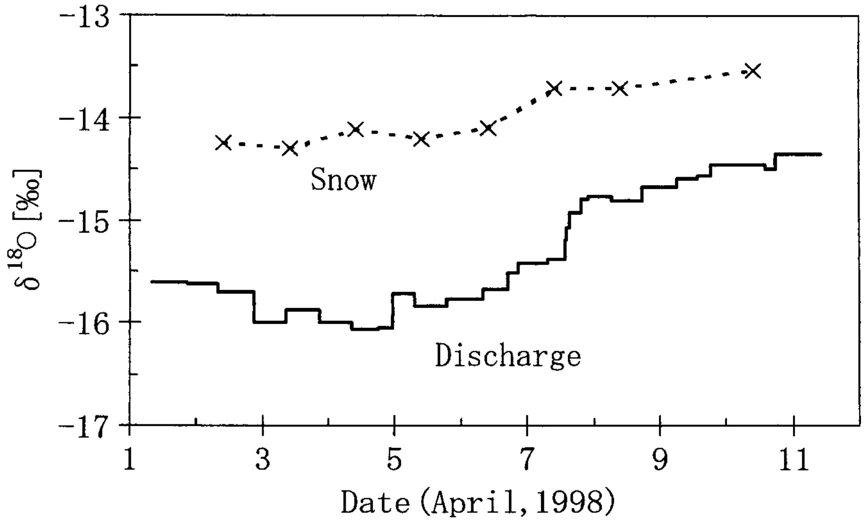

Figure 3. Temporal change of $\delta^{18}O$ in total snow pack and discharge water

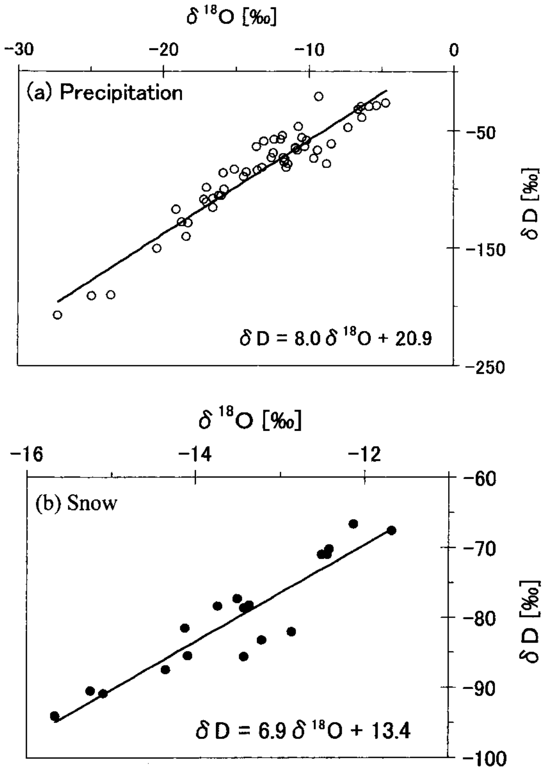

Figure 4. Relationship between δD and $\delta^{18}O$ in (a) precipitation and (b) total snow pack

According to Craig (1961), the relationship between δD and δ^{18}O is linear in the majority of meteoric waters, obeying the equation

$$\delta D = 8\,\delta^{18}O + 10 \tag{4}$$

which is the so-called 'meteoric water line'. The quantity called deuterium excess d

$$d = 8\,\delta^{18}O - \delta D \tag{5}$$

has been used to determine the moisture source for precipitation (e.g. Johnson *et al.*, 1989). For meteoric water, the deuterium excess value is generally close to 10‰. On the coast of the Sea of Japan, however, the deuterium excess of isotope concentration in the winter precipitation was slightly larger than 20‰. This is because the cold and dry Siberian air mass would contain significant amounts of vapour that had rapidly evaporated from the Sea of Japan (Waseda and Nakai, 1983). The deuterium excess for precipitation collected in Moshiri was consistent with these findings.

The relationship between δD and δ^{18}O for the total snow pack was approximated by

$$\delta D = 6{\cdot}9\,\delta^{18}O + 13{\cdot}4 \tag{6}$$

as shown in Figure 4b. The gradient of the regression line in Figure 4b was smaller for the snow pack than for the precipitation (Figure 4a), indicating that the isotope concentration changed after the snow deposition.

Temporal isotope change in individual layers

When snow layers are at melting point, snow particles tend to become granular, and under isothermal conditions the boundaries between neighbouring snow layers become less identifiable. Assuming that the water flowing down to the ground is at 0 °C, the solid portion of the inner snow layers will not change in mass. Hence, the snow pack was divided, following convention, into four layers (I–IV), as shown in Figure 5, based on the conservation law for individual layers. Geothermal heat, however, would cause a certain amount of melt at ground level. On 4 February, a marker was placed at a height of 22 cm from ground level. The mass of the solids in the basal layer (layer I) decreased with time due to melting caused by the geothermal

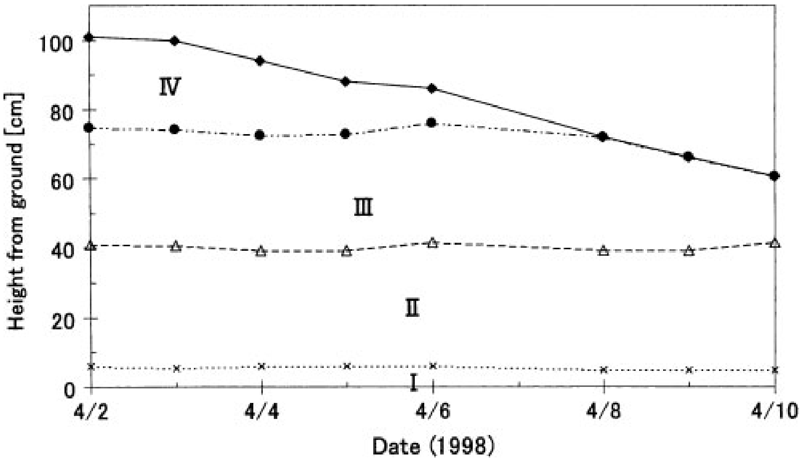

Figure 5. Conventional layering for the snow pack. The layers were defined to have the solid mass of 15 g cm^{-2} for the inner layers II and III, except after 8 April for layer III

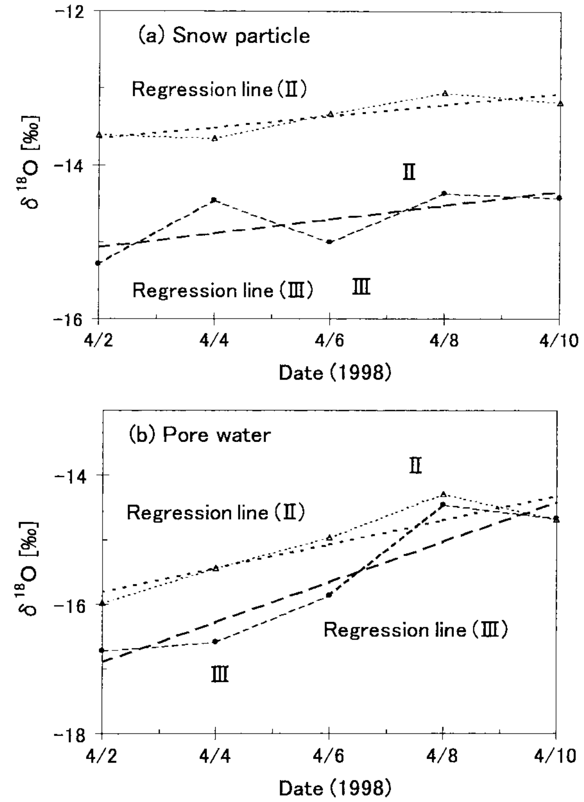

Figure 6. Temporal change of δ^{18}O in (a) the solid and (b) the liquid for layers II and III. The bold dashed and dotted lines indicate the respective regression lines

heat. Layers II and III are the subsequent two inner layers above the basal layer, defined as having 15 g cm^{-2} of solid snow mass in each layer. Layer IV is the surface layer remaining until 8 April.

Figure 6 shows the temporal isotope change for snow particles and pore water in layers II and III, where the mass of the solid was assumed to be constant. In these two inner layers, the isotope concentration tended to increase over time.

δD–$\delta^{18}O$ diagram for each layer

The δD–δ^{18}O diagram for snow particles in each layer is shown in Figure 7. The slopes of the regression lines are 4·7, 7·0, 7·3, and 4·2 for layers I, II, III, and IV respectively. These slopes are all smaller than 8, which is the slope of the meteoric water line.

Temporal change of average snow particle size

The average snow particle size for each layer is shown in Figure 8. The snow particle size tends to increase with time except in the surface layer I.

DISCUSSION

Near the surface, the difference between the isotope concentrations of the solid and the liquid was small. This difference, however, increased with depth (Figure 2). This suggests that isotopic fractionation between the

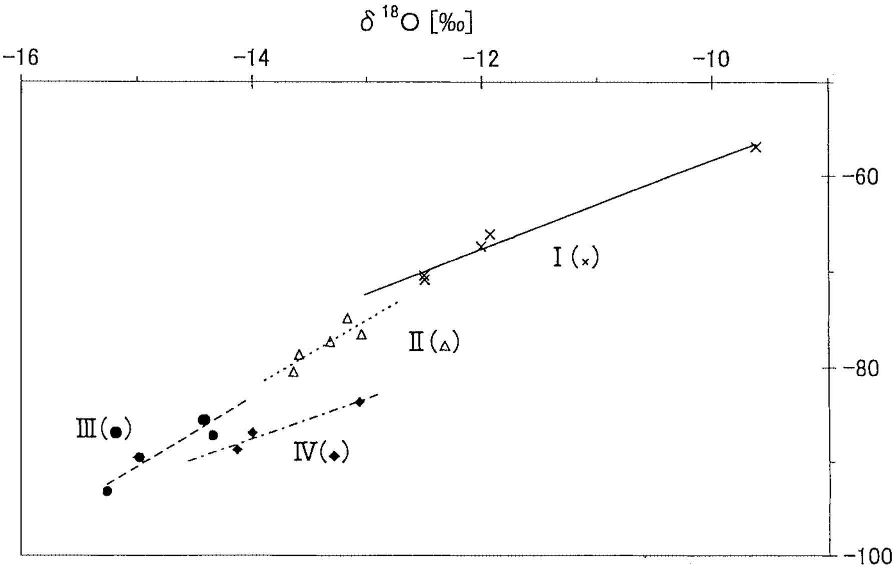

Figure 7. Relationship between δD and δ^{18}O in the solid for each layer. The slopes of the regression lines are 4·7, 7·0, 7·3, and 4·2 for layers I, II, III, and IV respectively

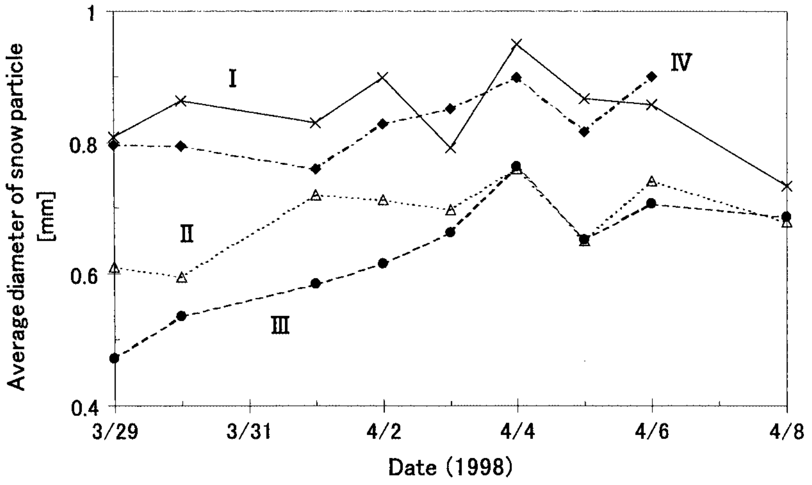

Figure 8. Temporal change of average snow particle size for each layer

liquid and the solid takes place as liquid water formed by melting at the surface flows downward. The results shown in Figure 3 suggest that the average isotope concentration for the total snow pack increased with time, because discharge water with relatively smaller isotope concentration than the total snow pack flowed out.

In the δD–δ^{18}O diagram (Figure 7), the slope of the regression line for both layer II and layer III, where the mass of the snow particles in each layer should be constant, is less than 8.

In a wet snow pack at 0 °C, due to the curvature effect, it is assumed that small particles shrink by melting and grow by freezing, and that both these processes take place simultaneously (Raymond and Tusima, 1979). However, isotope fractionation during snow melting can be ignored because the self-diffusion coefficient is very small in ice matrices (e.g. Jouzel and Souchez, 1982; Nakawo *et al.*, 1993). Thus, the freezing process is considered to be solely responsible for the change in isotope concentration in the wet snow. In this paper, the isotope fractionation during freezing is called the 'interaction' between snow particles and pore water.

Under isothermal conditions at 0 °C, the amount of relatively small particles melting is identical to the amount of water freezing to become large particles. This amount is henceforth called the 'exchange mass'.

Suppose water with the value of $\delta_a D$ and $\delta_a{}^{18}O$ is frozen (the subscript 'a' denotes 'initial liquid'). According to Souchez and Juzel (1984), the slope of the δD–$\delta^{18}O$ diagram of this freezing process is given by

$$S = \frac{\alpha_D \left[(\alpha_D - 1) \left(1 + \dfrac{\delta_a D}{1000} \right) \right]}{\alpha_{^{18}O} \left[(\alpha_{^{18}O} - 1) \left(1 + \dfrac{\delta_a{}^{18}O}{1000} \right) \right]} \tag{7}$$

where $\alpha_{^{18}O}$ and α_D are the isotope fractionation factors for freezing (for oxygen and hydrogen respectively) at the liquid–solid interface. They have values of 1·002 91 ($\alpha_{^{18}O}$) and 1·0212 (α_D) (Lehmann and Siegenthaler, 1991). Inserting the isotope concentrations of pore water (−15·97‰ for $\delta_a^{18}O$ and −95·87‰ for $\delta_a D$ in layer II and −16·72‰ for $\delta_a^{18}O$ and −103·31‰ for $\delta_a D$ in layer III on 4 April) into Equation (1), S was calculated to be 6·8 for both layers. The calculated results are in close agreement with the observed result of 6·9 (Equation (6)). Hence, it was suggested that the isotope concentration of the snow pack is modified by the freezing process that takes place in the snow pack.

When the 'isotope amount' ($W\delta$) is defined as the product of the water mass W and the isotope concentration δ, the isotope budget for the whole snow pack is expressed by

$$(W_0\delta_0) + (W_p\delta_p) = (W_1\delta_1) + (W_r\delta_r) \tag{8}$$

where the subscripts '0' and '1' denote an observation day and the next observation day respectively. In addition, the subscripts 'p' and 'r' refer to precipitation and discharge water between the observation days respectively.

The isotope amount budget of the solid in the inner layers, II and III, can be expressed as

$$\left(\frac{W_{g0}}{h} \delta'_{g0} \right) + (f\delta_f) = \left(\frac{W_{g1}}{h} \delta'_{g1} \right) + (f\delta_m) \tag{9}$$

where f is the exchange mass. The subscripts g0 and g1 denote snow particles per unit area on the observation day and on the next observation day respectively. The values of W_{g1} and W_{g0} are equal because the water mass of snow particles is constant in an inner layer. δ_f and δ_m refer to the isotope concentrations of frozen ice and of melted water respectively during two observation days, and h is the thickness of each layer.

When the isotope concentration of pore water is δ_{w0}, the isotope concentration of the frozen ice portion δ_f is expressed by

$$\delta_f/1000 = \alpha(1 + \delta_{w0}/1000) - 1 \tag{10}$$

where α is the isotope fractionation factor for freezing at the liquid–solid interface. δ_m is equal to δ'_{g0}, because melting occurs without the isotope fractionation (e.g. Jouzel and Souchez, 1982; Nakawo *et al.*, 1993). In addition, the following assumptions were made when calculating the exchange mass:

(1) the isotope concentration of snow particles changes linearly with time, as shown by regression lines in Figure 6a;
(2) the δ'_{g0} and δ_{w0} are constant over the two observation days;
(3) pore water flows downward from the snow surface to the ground, but the effect of this water flow is negligible compared with the effect of isotope change.

From Equations (2) and (3), 'exchange mass' in a day can be expressed by

$$f = \frac{W_{g0}}{h}\frac{(\delta'_{g1} - \delta'_{g0})}{(\delta_f - \delta_m)} = \frac{W_{g0}}{h}\frac{(\delta'_{g1} - \delta'_{g0})}{1000\left[\alpha\left(1 + \dfrac{\delta_{w0}}{1000}\right) - 1\right] - \delta'_{g0}} \tag{11}$$

Using this formula, the values for 'exchange mass' in this study were calculated as $5{\cdot}87 \times 10^{-2}$ g cm^{-3} in layer III and $7{\cdot}33 \times 10^{-2}$ g cm^{-3} in layer II.

The exchange mass value can also be calculated from the snow particle size distribution data. Raymond and Tusima (1979) proposed the following equation to describe the normalized cumulative frequency Ψ^* of the size distribution during the grain coarsening of wet snow:

$$\Psi^*\left(\frac{V}{V_m}\right) = \left(1 - \frac{aV}{bV_m}\right)^{1/a} \tag{12}$$

where V is the volume of a particle, V_m is the median volume, and a and b are parameters with constant values. Results using Equation (5) are consistent with the measured data, as shown in Figure 9. As can be

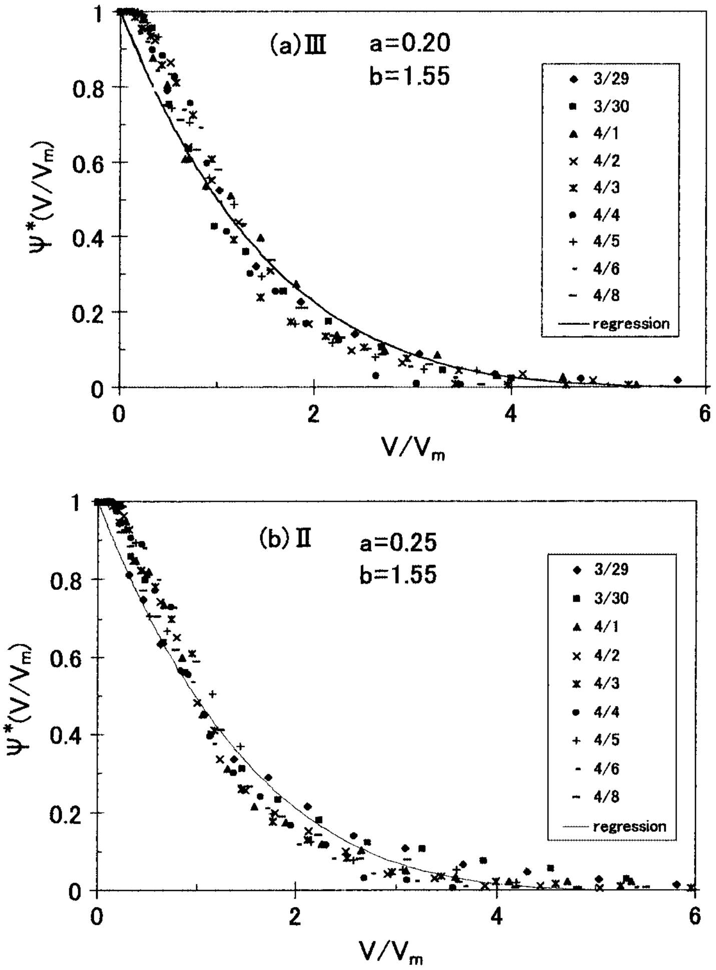

Figure 9. An example of cumulative distribution of particle volume of dry snow plotted as a function of V/V_m for layers III (a) and II (b)

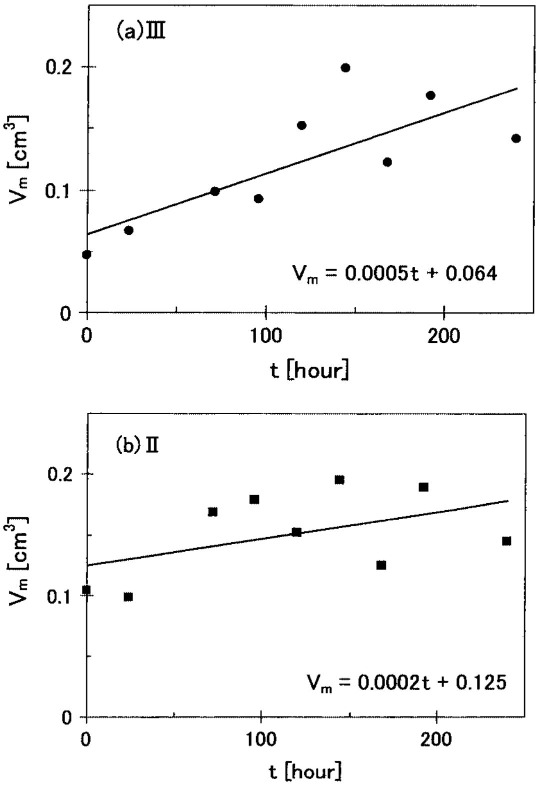

Figure 10. Median volume versus time for snow layers III (a) and II (b)

seen in Figure 10, V_m was assumed to increase linearly with time for simplicity.

$$V_m = V_0 + \gamma t \tag{13}$$

where V_0 is median volume at time zero (29 March, 10:00), dV_m/dt has a constant value, and γ is derived experimentally (Raymond and Tusima, 1979). The exchange mass f can then be calculated by

$$f = \beta \ln\left(1 + \frac{\gamma}{V_0}t\right) \tag{14}$$

where

$$\beta = \left(\frac{1}{1+a}\right)^{1/a} \tag{15}$$

as indicated by Raymond and Tusima (1979). Using values of 0·2 for a, 1·55 for b, 0·0005 cm^3 h^{-1} for γ, and 0·0638 cm^3 for V_0, a value of 5·07 × 10^{-2} g cm^{-3} for exchange mass f was obtained for layer III. Similarly, using the values of 0·25 for a, 1·55 for b, 0·0002 cm^3 h^{-1} for γ, and 0·1247 cm^3 for V_0, a value of 1·4 × 10^{-2} g cm^{-3} for exchange mass f was obtained for layer II. Thus, the values obtained for f differ significantly from the values estimated by the concentration of isotope amounts with fractionation.

This discrepancy could indicate that the isotope fractionation occurs during melting, although the diffusion coefficients of heavy isotopes for ice at 0 °C (the order of 10^{-14} m^2 s^{-1} (Hobbs, 1974)) are much smaller than those for water near 0 °C (on the order of 10^{-9} m^2 s^{-1} (Eisenberg and Kauzmann, 1969)). As indicated by Moser and Stichler (1980), however, the isotope fractionation during melting could occur if the solid phase is in the form of porous snow or firn. This may be due to the great surface area and the small surface layer thickness of the grains, which would allow isotope equilibrium by convection within the pore volume between neighbouring grains. If melting occurs as a result of isotope fractionation, δ_m would no longer be equal to δ'_{g0} in Equation (2). The water produced by melting δ_m can be calculated by

$$\delta_m/1000 = (1/\alpha)(1 + \delta'_{g0}/1000) - 1 \tag{16}$$

where α is the isotope fractionation factor for freezing at the liquid–solid interface, and δ'_{g0} is the isotope concentration of the snow particles on the observation day. As a result, the exchange mass in a day (including the isotope fractionation during melting) is calculated to be $2{\cdot}0 \times 10^{-2}$ g cm^{-3} in layer III and $1{\cdot}8 \times 10^{-2}$ g cm^{-3} in layer II. Although the latter value is consistent with that derived by the change in particle size, the former is not. The disparity between the calculations from isotope data and those from the change in particle size is considered to be caused not by the isotope fractionation during melting, but by the isotope change of pore water during ‘interaction’ (inflowing and outflowing). Therefore, the exchange between snow particles and pore water while flowing downward should be taken into account when examining isotope data from a wet snow pack. In order to clarify the change in isotope concentration with time (including effects of the downward flow of pore water), further studies, to look at such aspects in more detail, would be required.

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