

# Snow isotopic content change by sublimation

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**ABSTRACT.** We present results from cold-laboratory observations of changes in isotopic ( $\delta^{18}\text{O}$  and  $\delta\text{D}$ ) content by sublimation in snow and ice samples under nearly isothermal conditions. The results show large increases in observed  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in snow samples within several centimeters of the surface. They contradict the assumption of a non-changing isotopic content due to layer-by-layer transport mechanisms driven by sublimation/desublimation processes. The data also do not support the idea that isotopic changes of snow and firn are limited by the possibility that the ice matrix incorporates the atmospheric water vapor and that forced water-vapor diffusion in the pore space (wind pumping) is a requirement for isotopic content change. The observations show that sublimation from ice samples results in much lower increases in heavy-isotope content in the first several millimetres near the sublimating surface over the same time period, despite sublimation intensities similar to those of the snow samples. The results suggest that continuous phase transitions inside snow (recrystallization) are the process responsible for the isotopic content change because they are the primary mass-exchange mechanism between the snow mass and the surrounding environment. Modeling the isotopic content of the ice matrix therefore requires inclusion of a two-stage process: fractionation at the ice-matrix surface due to repetitive phase transitions, and fractionation due to preferable diffusion of light water isotopes in the pore space. For interpretation of the observed natural isotopic profiles in snow, the first process can be linked to the time a snow layer undergoes recrystallization, while the second process is related to the total ice/snow mass gain/loss determined by the external environmental conditions.

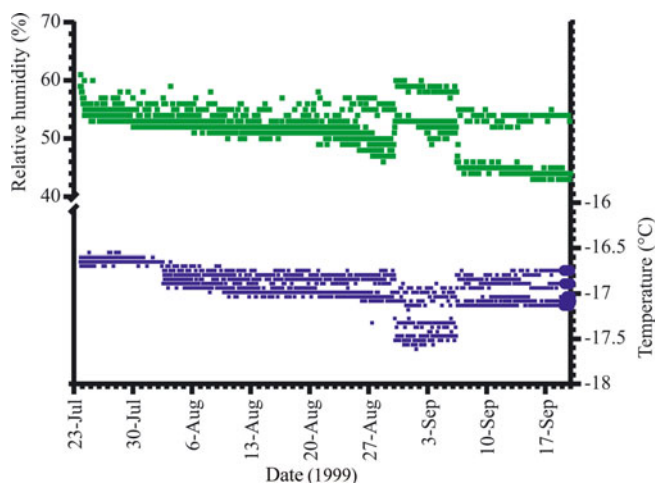
## INTRODUCTION

A fundamental problem in snow science is to quantify mass and energy exchanges at the surface and within the pore space of the snow cover. This information is particularly important for climate-change models since these exchanges influence the global water and energy cycle (Strasser and others, 2008). At present, snow-cover models are based on the assumption that water-vapor sublimation is mainly governed by external forcing (water-vapor flux regulated by temperature gradients within snow cover and mass loss/gain at the top surface of snow cover regulated by relative humidity, wind speed and radiation). In this paper, we describe the results of experiments devised to find the connection between the mass balance of snow and ice bodies and the evolution of their isotopic content. Previous studies on the isotopic content change of snow (Moser and Stichler, 1975; Sommerfeld and others, 1987, 1991; Friedman and others, 1991; Hachikubo and others, 1997, 2000; Satake and Kawada, 1997) show that the released water vapor has to be lighter in its isotopic content than the remaining bulk ice or liquid water. Thus, measurements of isotopic content are surrogate measures of mass-exchange rates. Therefore, measurements of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  in natural waters (Shanley and others, 1999; Oeschger, 2000) and snow in particular (Stichler, 1987) can have a wide range of environmental applications.

Isotopic fractionation is associated with the phase transitions at different stages of the water cycle. The water cycle connects evaporation of oceanic water, formation of clouds, cloud transfer by wind far from an initial source of the water vapor, precipitation, and construction of the isotopic content of the run-off back to the ocean (Gat, 1996; Kendall and

Caldwell, 1998; Araguás-Araguás and others, 2000; Taylor and others, 2001). Despite the reported uncertainties in relation to atmospheric circulation patterns, the measured isotopic composition of solid precipitation and variations of the isotopic content in the ground ice and snow are believed to represent climate variations (Moorman and others, 1996; Jouzel and others, 1997; Holdsworth, 2008). The simplest scheme describing these processes was presented by Dansgaard and others (1973, fig. 1). Such schemes are still in use in recent more complex climate reconstructions involving general circulation models (GCMs) (Delaygue and others, 2000; Hoffmann and others, 2000). One of the basic assumptions of such models is that the 'evaporation from solids causes no isotopic fractionation' (Dansgaard and others, 1973, p.10). In more recent ice-core paleoclimate research this is expressed as: 'the isotopic content of snow ... is linearly related to the temperature above the inversion level, ... where precipitation forms, and also to the surface temperature of the precipitation site...', with the two temperatures linearly related as well (Petit and others, 1999, p. 431). In other words, the isotopic content of buried snow represents temperature at the time of deposition.

However, isotopic fractionation does occur in a dry snowpack after deposition, though it is difficult to quantify (Cooper, 1998). Approximate values have been estimated in laboratory experiments (Moser and Stichler, 1975; Sommerfeld and others, 1987, 1991; Hachikubo and others, 2000) and have been reported for natural firn and snow (Friedman and others, 1991; Hachikubo and others, 1997; Satake and Kawada, 1997). Fractionation inside the snow cover is related to the non-equilibrium processes associated with water-vapor diffusion. These processes are considered to explain the isotopic profile homogenization in firn (Johnsen



**Fig. 1.** Measured temperature and relative humidity change during the Shinjo experimental run.

and others, 2000) and lead to the spatial difference in the observed isotopic content of snow and firn caused by mass exchanges between the ice matrix and the atmosphere when enhanced by airflow (Neumann and others, 2005). The fractionation factors involved are taken from experimental observations of the fractionation at an ice surface under controlled equilibrium conditions (e.g. Majoube, 1971).

The results of isotopic content observations in natural snow suggest that isotopic content change also depends on mass exchange with the atmosphere due to sublimation at the snow surface (Hachikubo and others, 1997; Stichler and others, 2001; Konishchev and others, 2003). However, the fact that the observed isotopic content change cannot be explained by mass loss alone requires additional processes to be considered, such as the existence of a liquid layer between the solid ice and the water vapor in the pore space of snow (Sommerfeld and others, 1991).

The question that arises from the current state of knowledge is how the layer-by-layer sublimation, which cannot change the isotopic content of remaining bulk significantly, can coexist with the observed enrichment of sublimating snow cover by heavy stable isotopes. The experiments described below were designed to gather data to answer this question.

## EXPERIMENTAL SET-UP

Two sets of independent experiments were performed in our investigations. The first set was carried out in the cold laboratory of the Shinjo Branch of Snow and Ice Studies, National Institute of Earth Science and Disaster Prevention, Shinjo, Japan, with the artificial snow vapor produced by the Cryospheric Environment Simulator (CES) ([http://www.bosai.go.jp/seppyo/jikkentou/jikkentou\\_syoukai\\_e.html](http://www.bosai.go.jp/seppyo/jikkentou/jikkentou_syoukai_e.html); Higashiura and others, 1997). The second set was carried out from 11 April 2001 to 22 May 2001 in the cold laboratory of the Laboratory of Snow Avalanches and Debris Flows, Faculty of Geography, Moscow State University, Russia, with natural snow collected in February 2001.

In both cases, in order to minimize disturbance of the environment in the cold room, human activity inside the room was limited to measurements related to these experiments.

## Shinjo experiments

Ten metal Petri dishes (diameter 0.149–0.152 m, height 0.027–0.029 m) and two plastic containers (bottom diameter 0.145 m, top diameter 0.15 m, height 0.087 m) were used in the experiments. The snow settled for a week in all containers, and before the start of the experiment the excess snow was cut off the top to produce 'fresh' snow at a smooth sublimating surface. Simultaneously, the settled snow produced by the CES was put into plastic bottles which were closed hermetically. One Petri dish was filled with water, the water was frozen, and the ice surface was polished to be at the level of the top edge of the dish.

The samples were positioned on a netted shelf, situated horizontally in the center of a large (1 m × 1 m × 1 m) plywood box that could be opened from the top. The box was placed in a cold room. All sides of the box were thermo-insulated with hydrophobic foam plastic. A lamp and a fan were installed in one of the bottom corners of the box. An additional fan was placed in a 0.05 m diameter hole in the wall opposite the lamp. The temperature in the box was regulated by two loggers connected to platinum resistances, one inside and one outside the box. The temperature in the box was set to  $-15^{\circ}\text{C}$  while the temperature in the cold room was set to  $-20^{\circ}\text{C}$ . The accuracy of temperature regulation in the cold room was not better than  $\pm 5^{\circ}\text{C}$ . The lamp inside the box was automatically switched on or off (with the additional fan in the hole switched off or on) when the temperature inside the box was below or above  $-15^{\circ}\text{C}$  respectively. The fan near the lamp was 'on' in either case. All regulation could automatically be switched off if the temperature in the cold room exceeded  $-15^{\circ}\text{C}$  (possible during short periods of daily defrosting of the cold room). Such regulation maintained the box at a temperature varying by not more than  $\pm 0.3^{\circ}\text{C}$ . The temperature difference between the cold room and the inside of the box could cause unplanned condensation of the atmospheric water vapor on the foam plastic walls on the inside of the box, thus drying the incoming air relative to conditions in the cold room. Hourly temperature and humidity inside the box during the experiment were recorded using micro memory loggers.

Every few days the sublimating samples were weighed to an accuracy of  $\pm 0.1$  g. One closed bottle and one metal cup with snow were removed from the box for isotopic analysis during each weighing. At the final weighing, the snow in the 0.087 m high containers was taken for isotopic analysis in 0.01–0.015 m horizontal layers.

During the experiment the amount of snow in the Petri dishes decreased due to sublimation. As a result, the initially smooth sublimating surfaces deformed and, at the end of the experiment, channels formed from the bottom to the top surface in one Petri dish. Snow thickness variations (0.035–0.06 m) were observed in the 0.087 m height cups at the end of the experiment.

Unavoidable temperature variations during the experiment corresponded to the observed variations of air humidity (Fig. 1). A period of notable difference from this general pattern, both in temperature and humidity, was caused by a defective lamp which was exchanged as soon as it was discovered. The decrease in temperature inside the box, with 'normal' temperature conditions outside the box, resulted in an increase in measured relative humidity values. However, we do not rely on the high accuracy of the humidity values reported by the conventional micro memory loggers, partly due to the known problems with

humidity measurements in cold environments (Makkonen and Laakso, 2005) and partly due to uncertainty in the explanation of the overall relatively steep trend of the measured relative humidity values for a small decrease in air temperature during the described experimental run.

### Moscow experiments

Ice samples were prepared from three types of frozen water: mineralized water, distilled water and melted filtered natural snow. The same snow was used for the sublimation of the 'snow sample'. Glass Petri dishes containing the samples were thermally insulated with thick foam plastic from all sides except the tops, and surrounded by 20 cm paper walls. The cold-laboratory temperature regulation was controlled by a temperature sensor positioned between the dishes.

Similar to the Shinjo experiments, sublimation resulted in a decrease in sample (especially snow-sample) thickness. Temperature and humidity above the sublimating samples were measured by thermograph and hygograph and were  $-4.3 \pm 1.5^\circ\text{C}$  and  $78 \pm 10\%$  respectively.

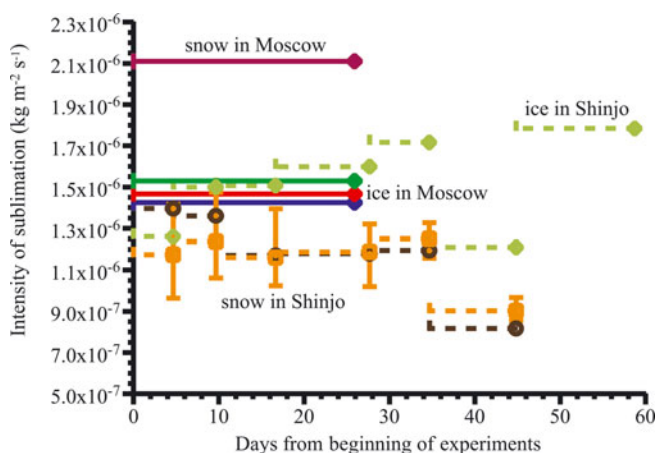
Isotopic sampling for the analysis was made (1) from initial material and (2) by 'shaving' from the top down in 0.5–1 mm thick sections (three layers from each sample) at the end of the experiment.

The principal difference in the experimental set-ups was between specifically suppressed air circulation in the Moscow experiments and freely circulating air (though resulting in much higher accuracy in maintaining the temperature regime) in the Shinjo experiments.

## EXPERIMENTAL RESULTS AND DISCUSSION

Intensity of sublimation can be considered as an integrated result of the effect of varying temperature, humidity and airflow velocity on the process of sublimation of snow. The mean intensity of sublimation of the snow and ice samples differed significantly between mass-loss estimations (weightings) during the experimental runs and in the different environmental conditions of the two laboratories (Fig. 2). The ice samples sublimated more intensively than the snow samples in the Shinjo experiments, while the opposite behavior was observed in the Moscow experiments. This difference can be explained by the air circulation conditions in the different cold rooms. Surface roughness can increase the area subjected to sublimation, as shown in the results by the difference in sublimation intensity in the Moscow experiments with stagnant air, and by the increase with time of the ice sublimation intensity in the Shinjo experiments. The latter is likely related to the sublimating erosion of an ice surface, reported by Kuroiwa (1969). Wind velocity is lower over rough snow surfaces than over smooth ice surfaces (Bintanja, 2001). Thus, for the snow samples sublimation intensity should be less affected by wind velocity. The decrease in sublimation intensity of the snow samples may be partly related to wind and sublimation smothering of the snow-sample surfaces relative to their initial state.

The isotopic content of ice samples could only be measured in the Moscow experiments. The fractionation due to solid-to-vapor water phase transition under the conditions and for the time period of the experimental run made the isotopic composition of the remaining bulk heavier by 1–2‰ for  $\delta^{18}\text{O}$  and by 4–8‰ for  $\delta\text{D}$ . This change was pronounced in 2–3 mm of ice near a sublimating surface, and



**Fig. 2.** Intensity of sublimation during both experimental runs. Colors of the solid horizontal bars correspond to the experimental runs as shown in Figure 3 for the experiments in Moscow (three different types of ice samples in Moscow). Dashed bars show the Shinjo results. The length of the horizontal bars indicates the time interval for the mean intensity determination. Vertical bars show the difference between samples where available.

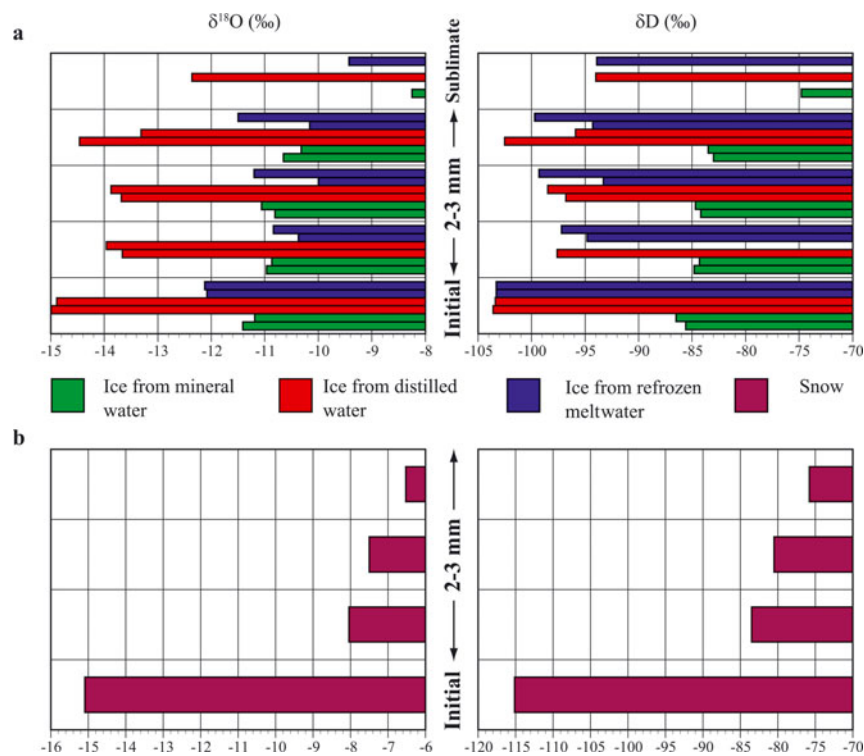
no gradient in heavy-isotope content was clearly identified over this distance (Fig. 3a).

A failure of the refrigerating equipment occurred during the experimental run in Moscow. The samples, hermetically covered by plastic tape, were therefore removed to a colder ( $-8^\circ\text{C}$ ) refrigerator for 2 days. After refrigeration repair, they were returned to the  $-4^\circ\text{C}$  cold room where it was found that the internal surface of the plastic tape was covered by sublimate. For some of the ice samples the sublimate was analyzed for its isotopic content. The results show differences in heavy-isotope content between the sublimate and the bulk samples. These are similar to the differences between the initial and final isotopic content of the ice over the whole experimental run (top bars in Fig. 3a). They are attributed to more active condensation of heavy isotopes at the tape from the water vapor produced by sublimation from warmer ice after temporary positioning in the refrigerator.

The isotopic content change in snow samples at the end of the experimental run was much greater than that observed in ice samples, about 8‰ for  $\delta^{18}\text{O}$  and about 40‰ for  $\delta\text{D}$ . There was a pronounced gradient in heavy-isotope content near the sublimating snow-sample surface (Fig. 3b).

In the Shinjo experiments the 'initial' isotopic composition of the snow samples was measured in snow kept in sealed plastic bottles during the experimental run. Observation of eight such samples yields  $\delta^{18}\text{O} = -7.3 \pm 0.1\text{‰}$ , which confirms the uniformity of the initial isotopic content of the sublimating snow. The isotopic content of the tap water was not measured, but was expected to be  $\delta^{18}\text{O} \approx -9\text{‰}$  and  $\delta\text{D} \approx -60\text{‰}$ . The initial  $\delta$  values of snow were higher as a result of fractionation during snow-grain formation ( $\delta^{18}\text{O} = -7.3\text{‰}$ ,  $\delta\text{D} = -46\text{‰}$ ), and close to the previously measured values of the snow produced in the CES ( $\delta^{18}\text{O}/\delta\text{D}$  were  $-7.3/-42$  and  $-7.6/-43$  respectively).

However, in the Moscow experiments, continuous change in the isotopic content of the sublimating snow was observed (Fig. 4a). The change shows nearly linear dependence on mass loss, which, except for the range of  $\delta$  values, in general corresponds to previously reported results (Moser and Stichler, 1975, 1980), and presents heavy-isotope enrichment



**Fig. 3.** Isotopic content measured in (a) ice and (b) snow samples in Moscow experiments. The layers from top down correspond to sublimate (for ice only), three 'shaved' layers (2–3 mm thick in total) and the original bulk material.

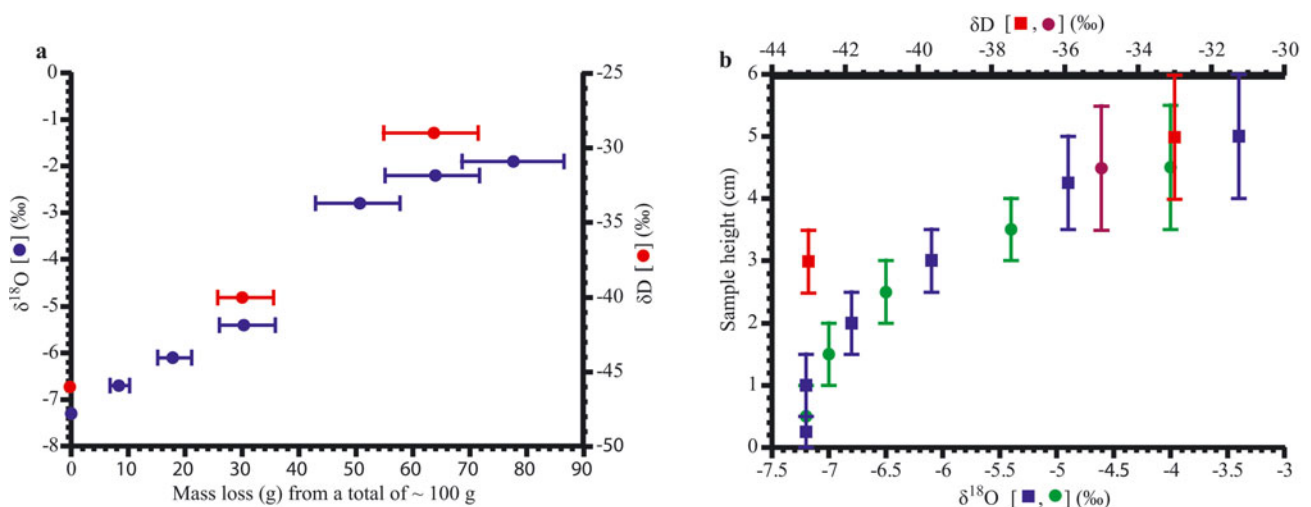
of 5‰ for  $\delta^{18}\text{O}$  and  $\sim 20\%$  for  $\delta\text{D}$ . For the final stage, vertical profiles of  $\delta^{18}\text{O}$  and  $\delta\text{D}$  over the remaining snow-sample thickness in the 0.087 m high containers were obtained (Fig. 4b). Extrapolation of the observed  $\delta^{18}\text{O}$  profile suggested the isotopic content in the immediate vicinity of the sublimating surface was similar to the final measurements in the time series with the Petri dishes (Fig. 4a). The same probably applies to the  $\delta\text{D}$  values; unfortunately, however, few  $\delta\text{D}$  measurements were possible. The available values constructing the  $\delta^{18}\text{O}/\delta\text{D}$  dependence are shown in Figure 5.

As a result of sublimation from the snow samples, the isotopic content of the remaining snow became noticeably

heavier and the departure from the global meteoric water-line (Fig. 5) corresponds to those explained by evaporation from an open surface (Clark and Fritz, 1997).

## CONCLUSIONS AND APPLICATIONS

Our experimental results are in agreement with previous data on isotopic content change in snow, suggesting that the lost material (water vapor) was lighter in isotopic content than the remaining bulk. This is due to the dependence of the diffusion coefficients of isotopic molecules of water on the molecular mass.



**Fig. 4.** Isotopic content of snow samples in Shinjo experiments: (a) changing with mass loss (near-linear with time) in samples from metal cups (horizontal bars show the possible error in mass loss determination); and (b) at the end of the experiment for 'thick' samples (vertical bars show the thickness of layers taken for analysis).

The presented results suggest the following interplay between the theoretically expected layer-by-layer sublimation/desublimation at the ice-matrix surface and the isotopic content evolution of snow cover due to mass exchange between the snow cover and the atmosphere:

The process of sublimation is continuous molecular exchange between the phases at the interface, with only part of the released water vapor transferred out of the system when conditions allow. The empirical equilibrium fractionation factors corresponding to this mass exchange are not directly linked to the overall evolution of the isotopic content of snow. The latter is affected at least by the specific surface area of snow exposed to mass exchange (Horita and others, 2008) and by the depth of the snow layer exposed to the mass exchange with the atmosphere (He and Smith, 1999). Several centimeters of snow are a source of sublimation from snow, partly because of interconnection of pores and partly due to observed sublimative cooling resulting in temperature gradient formation (Golubev and Sokratov, 1991). This is supported by our experimental results (Fig. 4b).

In principle, the process of isotopic content change by sublimation can be linked to the model approach of Feng and others (2002), which has been shown to describe the isotopic content change of snow due to repetitive melting/refreezing (Zhou and others, 2008). However, modeling of the fractionation due to sublimation requires an accurate enough description of the dependence of the activity of recrystallization of snow on environmental parameters (Sokratov, 2001; Bartelt and others, 2004). The data provided by computed micro-tomography (Schneebeli and Sokratov, 2004) show that the mass exchange between the neighboring ice-matrix structures (recrystallization of snow) can be orders of magnitude higher than the mass transferred in the direction of a temperature gradient due to water-vapor concentration variation (Sokratov and others, 2004).

For application to natural snow covers, the experimental results suggest that the isotopic content, in addition to the initial isotopic content of solid precipitations and activity of sublimation, is regulated by the time of exposure of deposited snow to the atmosphere (Moser and Stichler, 1975). The latter depends on the time between precipitation events and their amount of precipitation. A snow-cover depth dependence of evolution of the isotopic content has been reported for northern Eurasia (Konishchev and others, 2003).

Since the isotopic content of snow is determined by the 'openness' of the system, similar effects must be expected in glacier snow and firn down to considerable depths. Though the mass fluxes deep below the surface are small, the duration of the ice-matrix surface exposure to fractionation during gradual sinkage of each specific snow layer (due to accumulation at the surface) reaches tens or hundreds of years.

In addition to the dependence on temperature at the moment of deposition (linear, according to Petit and others (1999)), the isotopic content in a buried snow layer is determined by (1) relative mass loss during the layer presence near the day surface (the surface exposed to light), (2) dependence of snow recrystallization activity on the snow layer temperature (presumably exponential, when determined by the water-vapor content in the pore space of snow), varying seasonally and in the long term, and (3) possible changes in the mass transfer into and out of the layer, varying with the depth and age of the snow layer. Despite a considerable literature on the isotopic composition of snow, estimation of

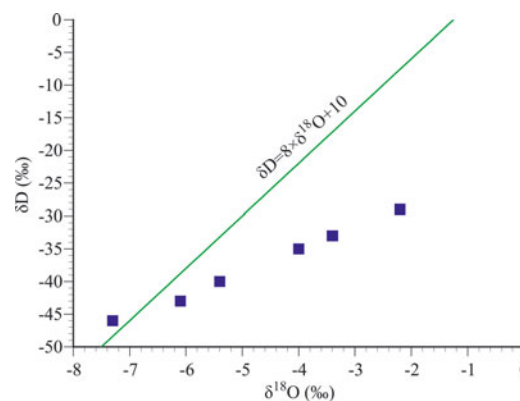


Fig. 5. The  $\delta^{18}\text{O}/\delta\text{D}$  ratio for snow samples in the experimental run at Shinjo.

the importance of the above listed processes in various applications still requires additional experimental data on simultaneously measured isotopic content change and environmental parameters responsible for fractionation due to sublimation.

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