# Experimental observation of transient $\delta^{18}O$ interaction between snow and advective airflow under various temperature gradient conditions

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## Abstract

Stable water isotopes ($\delta^{18}O$) obtained from snow and ice samples of polar regions are used to reconstruct past climate variability, but heat and mass transport processes can affect the isotopic composition. Here we present an experimental study on the effect of airflow on the snow isotopic composition through a snow pack in controlled laboratory conditions. The influence of isothermal and controlled temperature gradient conditions on the $\delta^{18}O$ content in the snow and interstitial water vapor is elucidated. The observed disequilibrium between snow and vapor isotopes led to the exchange of isotopes between snow and vapor under non-equilibrium processes, significantly changing the $\delta^{18}O$ content of the snow. The type of metamorphism of the snow had a significant influence on this process. These findings are pertinent to the interpretation of the records of stable isotopes of water from ice cores. These laboratory measurements

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suggest that a highly resolved climate history is relevant for the interpretation of the snow isotopic composition in the field.

*Keywords*: snow, isotope, isothermal, metamorphism, advection, tomography, post-depositional process

## 1. Introduction

Water stable isotopes in polar snow and ice have been used for several decades as proxies for global and local temperatures (e.g. Dansgaard, 1964; Lorius et al., 1979; Grootes et al., 1994; Petit et al., 1999; Johnsen et al., 2001; EPICA Members, 2004). However, the processes that influence the isotopic composition of precipitation in high-latitude are complex, making direct inference of paleo temperatures from the isotopic record difficult (Cuffey et al., 1994; Jouzel et al., 1997, 2003; Hendricks et al., 2000). Several factors affect the vapor and snow isotopic composition, which give rise to ice core isotopic composition, starting from the process of evaporation in the source region, transportation of the air mass to the top of the ice sheet, and post-depositional processes (Craig and Gordon, 1964; Merlivat and Jouzel, 1979; Johnsen et al. 2001; Ciais and Jouzel, 1994; Jouzel and Merlivat, 1984; Jouzel et al., 2003; Helsen et al., 2005, 2006, 2007; Cuffey and Steig, 1998; Krinner and Werner, 2003). Mechanical processes such as mixing, seasonal scouring, or spatial redistribution of snow can alter seasonal and annual records (Fisher et al., 1983; Hoshina et al., 2014). Post-depositional processes associated with wind scouring and snow redistribution are known to introduce a “post-depositional noise” in the surface snow. Comparisons of isotopic records obtained from closely located shallow ice cores have allowed for estimation of a signal-to-noise ratio and a common climate signal (Fisher and Koerner, 1988, 1994; White et al., 1997; Steen-Larsen et al., 2011; Sjolte et al., 2011; Masson-Delmotte et al., 2015). After deposition, interstitial diffusion in the firn and ice affects the water-isotopic signal but back-diffusion or deconvolution techniques have been used to establish the original isotope signal (Johnsen, 1977; Johnsen et al., 2000).

Snow is a bi-continuous material consisting of fully connected ice crystals and pore space (air) (Löwe et al., 2011). Because of the proximity to the melting point, the high vapor pressure causes a continuous recrystallization of the snow microstructure known as snow metamorphism, even under moderate temperature gradients (Pinzer et al., 2012). The whole ice matrix is continuously recrystallizing by sublimation and deposition, with vapor diffusion as the dominant transport process. Pinzer et al. (2012) showed that a typical half-life of the ice matrix is a few days. The intensity of the recrystallization is dictated by the temperature gradient and this can occur under mid-latitude or polar conditions. Temperature, and geometrical factors (porosity and specific surface area) also play a significant role (Pinzer and Schneebeli, 2009; Pinzer et al., 2012).

The interpretation of ice core data and the comparison with atmospheric model results implicitly rely on the assumption that the snowfall precipitation signal is preserved in the snow-ice matrix (Werner et al., 2011). Classically, ice-core stable-isotope records are interpreted as reflecting precipitation-weighted signals, and compared to observations and atmospheric model results for precipitation, ignoring snow-vapor exchanges between surface snow and atmospheric water vapor (e.g. Persson et al., 2011). However, recent studies carried out on top of the Greenland and Antarctic ice sheets combining continuous atmospheric water vapor isotope observations with daily snow surface sampling document a clear day-to-day variation of isotopic composition of surface snow between precipitation events as well as diurnal change in the snow isotopes (Steen-Larsen et al., 2014a; Ritter et al., 2016; Casado et al., 2016). This effect was interpreted as being caused by the uptake of the synoptic-driven atmospheric water-vapor isotope signal by individual snow crystals undergoing snow metamorphism (Steen-Larsen et al., 2014a) and the diurnal variation in moisture flux (Ritter et al., 2016). However, the impact of this process on the isotope-temperature reconstruction is not yet sufficiently understood, but crucial to constrain. This process, compared to interstitial diffusion (Johnsen, 1977; Johnsen et al., 2000), will alter the isotope mean value. The field observations challenge the previous assumption that sublimation occurred molecular layer-by-layer with no resulting isotopic fractionation (Dansgaard, 1964; Friedman et al., 1991; Town et al., 2008; Neumann and Waddington, 2004). It is assumed that the solid undergoing sublimation would not be unduly enriched in the heavier isotope species due to the preferential loss of lighter isotopic species to the vapor (Dansgaard, 1964; Friedman et al., 1991). Because self-diffusion in the ice is about three orders of magnitude slower than molecular diffusion in the vapor, the amount of isotopic separation in snow is assumed to be negligible.

Snow has a high permeability (Calonne et al., 2012; Zermatten et al., 2014), which facilitates diffusion of gases and, under appropriate conditions, airflow (Gjessing, 1977; Colbeck, 1989; Sturm and Johnson, 1991; Waddington et al., 1996). In a typical Antarctic and Greenland snow profile, strong interactions between the atmosphere and snow occurs, especially in the first 2 m (Neumann and Waddington, 2004; Town et al., 2008), called the convective zone. In the convective zone, air can move relatively freely and therefore exchange between snow and the atmospheric air occurs. Air flowing into the snow reaches saturation vapor pressure nearly instantly through sublimation (Neumann et al., 2008; Ebner et al., 2015a). Models of the influence of the so-called ‘wind pumping’-effect (Fisher et al., 1983; Neumann and Waddington, 2004), in which the interstitial water vapor is replaced by atmospheric air pushed through the upper meters of the snow pack by small-scale high and low pressure areas caused by irregular grooves or ridges formed on the snow surface (dunes and sastrugi), have assumed that the snow grains would equilibrate with the interstitial water vapor on timescales governed by ice self-diffusion. However, no experimental data are available to support this assumption. With this in mind the experimental study presented here is specifically developed to investigate the effect of ventilation inside the snow pack on the isotopic composition. Only conditions deeper than 1 cm inside a snowpack are considered. Previous work showed that (1) under isothermal conditions, the Kelvin effect leads to a saturation of the pore space in the snow but does not affect the structural change (Ebner et al., 2015a); (2) applying a negative temperature gradient along the flow direction leads to a change in the microstructure due to deposition of water molecules on the ice matrix (Ebner et al., 2015b); and (3) a positive temperature gradient along the flow had a negligible total mass change of the ice but a strong reposition effect of water molecules on the ice grains (Ebner et al., 2016). Here, we continuously measured the isotopic composition of an airflow containing water vapor through a snow sample under both isothermal and temperature gradient conditions. Micro computed-tomography (μCT) was applied to obtain the 3D microstructure and morphological properties of snow.

## 2. Experimental setup

Isothermal and temperature gradient experiments with fully saturated airflow and defined isotopic composition were performed in a cold laboratory at around $T_{\mathrm{lab}} \approx -15$ °C with small fluctuations of ± 0.8 °C (Ebner et al., 2014). Snow produced from de-ionized tap water in a cold laboratory (water temperature: 30 ºC; air temperature: -20 ºC) was used for the snow sample preparation (Schleef et al., 2014). The snow was sieved with a mesh size of 1.4 mm into a box, and isothermally sintered for 27 days at -5 ºC to increase the strength, in order to prevent destruction of the snow sample due to the airflow, and to evaluate the effect of metamorphism of snow. The morphological properties of the snow are listed in Table 1. The sample holder (diameter 53 mm, height 30 mm, 0.066 liter) was filled by a cylinder cut out from the sintered snow. To prevent air flow between snow sample and the sample holder walls, the undisturbed snow disk was filled in at a higher temperature (about -5 °C) and sintering was allowed for about 1 h before cooling down and start of the experiment. The setup of Ebner et al. (2014) was modified by additionally inserting a water vapor isotope analyzer (Model: L1102-I Picarro, Inc., Santa Clara, CA, USA) to measure the isotopic ratio $\delta^{18}O$ of the water vapor contained in the airflow at the inlet and outlet of the sample holder. The experimental setup consisted of three main components (humidifier, sample holder, and the Picarro analyzer) connected with insulated copper tubing and Swagelok fitting (Fig. 1). The tubes to the Picarro analyzer were heated to prevent deposition of water vapor and thereby fractionation. The temperature was monitored with thermistors inside the humidifier and at the inlet and outlet of the snow sample. A dry air pressure tank controlled by a mass flow controller (EL-Flow, Bronkhorst) generated the airflow. A humidifier, consisting of a tube (diameter 60 mm, height 150 mm, 0.424 liter volume) filled with crushed ice particles (snow from Antarctica with low $\delta^{18}O$ composition), was used to saturate the dry air entering the humidifier with water vapor at an almost constant isotopic composition. The air temperature in the humidifier and at the inlet of the snow sample was maintained at the same value (accuracy ± 0.2 K) to limit the influence of variability in absolute vapor pressure and isotopic composition. We measured the $\delta^{18}O$ of the water vapor produced by the humidifier before and after each experimental run ($\delta^{18}O_{hum}$). The outlet flow ($\delta^{18}O_a$) of the sample holder was continuously measured during the experiment to analyze the temporal evolution of the isotopic signal. All data from the Picarro analyzer were corrected to the humidity reference level using the established instrument humidity-isotope response (Steen-Larsen et al. 2013; 2014b). In addition, VSMOW-SLAP correction and drift correction were performed. We followed the calibration protocol and used the calibration system described in detail by Steen-Larsen et al. (2013; 2014b).

The sample holder described by Ebner et al. (2014) was used to analyze the snow by μCT. Tomography measurements were performed with a modified μ-CT80 (Scanco Medical). The equipment incorporated a microfocus X-ray source, operated at 70 kV acceleration voltage with a nominal resolution of 18 μm. The samples were scanned with 1000 projections per 180° in high-resolution setting, with typical adjustable integration time of 200 ms per projection. The field of view of the scan area was 36.9 mm of the total 53 mm diameter, and subsamples with a dimension of 7.2 × 7.2 × 7.2 $mm^3$ were extracted for further processing. The reconstructed μCT images were filtered using a 3 × 3 × 3 median filter followed by a Gaussian filter ($\sigma = 1.4$, support = 3). The Otsu method (Otsu, 1979) was used to automatically perform clustering-based image thresholding to segment the grey-level images into ice and void phase. Morphological properties in the two-phase system were determined based on the exact geometry obtained by the μCT. Tetrahedrons corresponding to the enclosed volume of the triangulated ice matrix surface were applied on the segmented data to determine porosity ($\varepsilon$) and specific surface area (SSA). Opening size distribution operation was applied in the segmented μCT data to extract the mean pore size ($d_{mean}$). The opening size distribution can be imagined as virtual sieving with different mesh size (Haussener et al., 2012).

Three experiments with saturated advective airflow through the snow sample were performed to record the following parameters and analyze their effects: (1) isothermal conditions to analyze the influence of curvature effects (Kaempfer al et., 2007); (2) positive temperature gradient applied to the snow sample where cold air entering the sample is heated while flowing through the sample in order to analyze the influence of sublimation; (3) negative temperature gradient applied to the snow sample where warm air entering the sample is cooled while flowing across the sample, to analyze the influence of net deposition. During the temperature gradient experiments, a temperature difference of 1.4 ºC and 1.8 ºC was imposed resulting in a gradient of +47 K $m^{-1}$ and -60 K $m^{-1}$, respectively. The runs were performed at atmospheric pressure and with a volume flow rate of 3.0 liter $min^{-1}$ corresponding to an average flow speed in the pores of ≈ 30 mm $s^{-1}$. In wind pumping theory, an airflow velocity of $u_D \approx 10$ mm $s^{-1}$ (corresponding Reynolds number Re ≈ 0.65) was estimated inside the surface snow layers ($d_{mean} \approx 1$ mm) for a high wind speed above the snow surface (≈ 10 m $s^{-1}$) (Neumann, 2003). We performed experiments with airflow velocities inside the snow sample of around 30 mm $s^{-1}$ (corresponding Reynolds number Re = 0.7), which was a factor three higher than in natural conditions. But looking at the Reynolds number our experiments were in the feasible flow regime (laminar flow) of a natural snow pack. In experiment (2) the outlet temperature and in experiment (3) the inlet and also the humidifier temperature were actively controlled using thermo-electric elements. Variations in temperature of up to ± 0.8 ºC were due to temperature fluctuations inside the cold laboratory, leading to slightly variable temperature gradients and mean temperature in experiment (2) and (3). Table 1 presents a summary of the experimental conditions and the morphological properties of the snow samples. At the end of each experiment, the snow sample was cut into five layers of 6 mm height and the isotopic composition of each layer was analyzed to examine the spatial $\delta^{18}O$ gradient in the isotopic composition of the snow sample.

A slight increase with a maximum of 0.7 ‰ of $\delta^{18}O$ in the water vapor produced by the humidifier was observed in experiment (1), with lower increases during experiments (2) and (3) (Table 2). This change of ~0.7‰ is not significant compared to the difference between the isotopic composition of the water vapor and the snow sample in the sample holder of ~53‰ and the temporal change of the water vapor isotopes on the back side of the snow sample.

In the first approximately 30 min, the isotopic composition of the measured outflow air $\delta^{18}O_a$ increased from a low $\delta^{18}O$ to a starting value of around -29‰ in each experiment. This was due to memory effect possible condensed water left in the tubes from a prior experiment.

# 3. Results

## 3.1 Isothermal condition

The experiment (1) was performed for 24 h at a mean temperature of $T_{mean}$ = -15.5 ºC. $\delta^{18}O_a$ decreased exponentially in the outlet flow observed throughout the experimental run as shown in Fig. 2. Initially, the $\delta^{18}O_a$ content in the flow was -27.7 ‰ and exponentially decreased to -47.6 ‰ after 24 h. The small fluctuations in the $\delta^{18}O_a$ signal at $t \approx 7$ h, 17 h and 23 h were due to small temperature changes in the cold laboratory.

We observed a strong interaction between the airflow and the snow as manifest by the isotopic composition of the snow. The $\delta^{18}O_s$ signal in the snow decreased by 4.75 – 7.78 ‰ and an isotopic gradient in the snow was observed after the experimental run, shown in Fig. 3. Initially, the snow had a homogeneous isotopic composition of $\delta^{18}O_s$ =

-10.97 ‰ but post-experiment sampling showed a decrease in the snow $\delta^{18}O$ at the inlet side to -17.75 ‰ and at the outlet side to -15.72 ‰. The spatial $\delta^{18}O_s$ gradient of the snow had an approximate slope of 0.68 ‰ mm$^{-1}$ at the end of the experimental run. Table 2 shows the $\delta^{18}O$ value in snow at the beginning ($t$ = 0) and end ($t$ = 24 h) of the experiment.

### 3.2 Air warming by a positive temperature gradient along the airflow

The experiment (2) was performed over a period of 24 h with an average temperature gradient of approximatively +47 K m$^{-1}$ (warmer temperatures at the outlet of the snow) and an average mean temperature of -14.7 ºC. As in the isothermal experiment (1), we observed a relaxing exponential decrease of $\delta^{18}O_a$ in the outlet flow throughout the measurement period as shown in Fig. 2, but the decrease was slower compared to the isothermal run. Initially, the $\delta^{18}O_a$ content in the flow coming through the snow disk was -29.8 ‰ and exponentially decreased to -41.9 ‰ after 24 h. The small fluctuations in the $\delta^{18}O_a$ signal at $t \approx 2.7$ h, and 12.7 h were due to small temperature changes in the cold laboratory.

The $\delta^{18}O_s$ signal in the snow decreased 4.66 – 7.66 ‰ and a gradient in the isotopic composition of the snow was observed after the experimental run, shown in Fig. 3. Initially, the snow had a homogeneous isotopic composition of $\delta^{18}O_s$ = -11.94 ‰, but post-experiment sampling showed a decrease at the inlet side to -19.6 ‰ and at the outlet side to -16.6 ‰. The spatial $\delta^{18}O_s$ gradient of the snow had an approximate slope of 1.0 ‰ mm$^{-1}$ at the end of the experimental run. Table 2 shows the $\delta^{18}O_s$ value in snow at the beginning ($t$ = 0) and end ($t$ = 24 h) of the experiment.

### 3.3 Air cooling by a negative temperature gradient along the air flow

The experiment (3) was performed for 84 h instead of 24 h to better estimate the trend in $\delta^{18}O_a$ in the outlet flow. An average temperature gradient of approximately -60 K $m^{-1}$ (colder temperatures at the outlet of the snow) and an average mean temperature of -13.2 ºC were observed during the experiment. As in the previous experiments, a relaxing exponential decrease of $\delta^{18}O_a$ in the outlet flow was observed throughout the experimental run as shown in Fig. 2, but the decrease was slower compared to the isothermal run and temperature gradient opposed to the airflow. Initially, the $\delta^{18}O_a$ content in the flow was -29.8 ‰ and exponentially decreased to -37.7 ‰ after 84 h. The small fluctuations in the $\delta^{18}O_a$ signal at $t \approx 7.3$ h, 21.3 h, 31.3 h, 45.3 h, 55.3 h, 69.3 h, and 79.3 h were due to small temperature changes in the cold laboratory.

The $\delta^{18}O_s$ signal in the snow decreased 4.46 – 15.09 ‰ and a gradient in the isotopic composition of the snow was observed after the experimental run, shown in Fig. 3. Initially, the snow had an isotopic composition of $\delta^{18}O_s$ = -10.44 ‰ but post-experiment sampling showed a decrease at the inlet side to -25.53 ‰ and at the outlet side to -15.00 ‰. The spatial $\delta^{18}O_s$ gradient of the snow had an approximate slope of 3.5 ‰ $mm^{-1}$ at the end of the experimental run. Table 2 shows the $\delta^{18}O_s$ value in snow at the beginning ($t = 0$) and end ($t = 84$ h) of the experiment.

## 4. Discussion

All experiments showed a strong exchange in $\delta^{18}O$ between the snow and water-vapor saturated air resulting in a significant change of the value of the stable isotopes in the snow. The advective conditions in the experiments were comparable with surface snow layers in Antarctica and Greenland, but at higher temperature, especially compared to interior Antarctica.

The results also showed strong interactions in $\delta^{18}O$ between snow and air depending on the different temperature gradient conditions. The experiments indicate that temperature variation and airflow above and through the snow structures (Sturm and Johnson, 1991; Colbeck, 1989; Albert and Hardy, 1995) seem to be dominant processes affecting water stable isotopes of surface snow. The results also support the statement that an interplay between theoretically expected layer-by-layer sublimation and deposition at the ice-matrix surface and the isotopic content evolution of snow cover due to mass exchange between the snow cover and the atmosphere occurs (Sokratov and Golubev, 2009). The specific surface area of snow exposed to mass exchange (Horita et al., 2008) and by the depth of the snow layer exposed to the mass exchange with the atmosphere (He and Smith, 1999) plays an important role. Our results support the interpretation that changes in surface snow isotopic composition are expected to be significant if large day-to-day surface changes in water vapor occur in between precipitation events, wind pumping is efficient and snow metamorphism is enhanced by temperature gradients in the upper first centimeters of the snow (Steen-Larsen et al., 2014a).

We expect that our findings will lead to improvement of the interpretation of the water stable isotope records from ice cores. Classically, ice core stable isotope records are interpreted as paleo-temperature reflecting precipitation-weighted signals. When comparing observations and atmospheric model results for precipitation with ice core records, such vapor-snow exchanges are normally ignored (e.g. Persson et al., 2011; Fujita and Abe, 2006). However, vapor-snow exchange enhanced by recrystallization rate seems to be an important factor for the high variation in the snow surface $\delta^{18}O$ signal as supported by our experiments. It was hypothesized that the changes in the snow-surface $\delta^{18}O$ reported by Steen-Larsen et al. (2014a) are caused by changes in large-scale wind and moisture advection of the atmospheric water vapor signal and snow metamorphism. The strong interaction between atmosphere and near-surface snow can modify the ice core water stable isotope records.

The rate-limiting step for isotopic exchange in the snow is isotopic equilibration between the pore-space vapor and surrounding ice grains. The relaxing exponential decrease of $\delta^{18}O$ in the outflow of our experiments predicted that full isotopic equilibrium between snow and atmospheric vapor will not be reached at any depth (Waddington et al., 2002; Neumann and Waddington, 2004) but changes towards equilibrium with the atmospheric state occurs (Steen-Larsen et al., 2013, 2014a).

As snow accumulates, the upper 2 m are advected through the ventilated zone (Neumann and Waddington, 2004; Town et al., 2008). In areas with high accumulation rate (e.g. South Greenland), snow is advected for a short time through the ventilated zone. The snow exposed a relatively short time to vapor snow exchange would result in higher spatial variability compared to long-time exposure. However, the effects of snow ventilation on isotopic composition may become more important as the accumulation rate of the snow decreases ($< 50$ mm $a^{-1}$), such that snow remains in the near-surface ventilated zone for many years (Waddington et al., 2002; Hoshina et al., 2014; Hoshina et al., 2016). As the snow remains longer in the near-surface ventilated zone, a larger $\delta^{18}O$ exchange between snow and atmospheric vapor will occur. Consequently, the isotopic content of layers at sites with high and low accumulation rates can evolve differently, even if the initial snow composition had been equal, and the sites had been subjected to the same histories of air-mass vapor.

Despite a relatively small change in the difference between the isotopic composition of the incoming vapor and the snow, large differences in the isotopic composition of the water vapor at the outlet flow exist for the three different experimental setups. Based on the difference in the outlet water vapor isotopic composition, we hypothesized that different processes are at play for the different experiments. It is obvious that there is a fast isotopic exchange with the surface of the ice crystals, and a much slower timescale on which the interior of the ice crystals is altered. Due to the low diffusivity of $H_2{}^{16}O$ and $H_2{}^{18}O$ in ice ($D_{H_2{}^{18}O} \approx D_{H_2{}^{16}O}$ = ~$10^{-15}$ $m^2$ $s^{-1}$ (Ramseier, 1967; Johnsen et al., 2000), we assumed that the interior of the ice crystals is not altered on the timescale of the experiment. This explained why the net isotopic change of the bulk sample is relatively small compared to the changes in the outlet water vapor isotopes. The effective 'ice-diffusion depth' of the isotopic exchange during the experiments is given as $L_D = \sqrt{D \cdot t}$, where $D$ is the diffusion coefficient of $H_2{}^{16}O$ and $H_2{}^{18}O$ in ice, respectively, and $t$ the experimental time. The calculated 'ice-diffusion depth' $L_D$, is ~ 9.3 µm for experiments (1) and (2), and ~17.4 µm for experiment (3), respectively, indicating an expected a minimal change of the interior of the ice crystal. However, snow has a large specific surface area and therefore a high exchange area. This has an effect on the $\delta^{18}O$ snow concentration. The fraction of the total volume $V_{tot}$ of ice that is close enough to the ice surface to be affected by diffusion in time $t$ is then $\rho_{ice} \cdot SSA \cdot L_D$, where SSA is the specific surface area (area per unit mass), and $L_D$ is the diffusion depth, defined above, for time $t$. For $t \approx 24$ hours, a large fraction (24 to 43 %) of the total volume $V_{tot}$ of the ice matrix can be accessed through diffusion. It is quite hard to see the total $\delta^{18}O$ snow difference between experiments (1) and (2) after the experiment compared to the $\delta^{18}O$ of the vapor in the air at the outlet. There is a small, but notable, difference in the total $\delta^{18}O$ of the snow between experiment (1) and (2). Due to the higher recrystallization rate of experiment (2) the spatial $\delta^{18}O_s$ gradient of the snow (1.0 ‰ $mm^{-1}$) is higher than for experiment (1) (0.68 ‰ $mm^{-1}$). Increasing the experimental time, the $\delta^{18}O$ change in the snow increases (experiment (3)). In general, the calculated 'ice-diffusion depth' is realistic under isothermal conditions where diffusion processes are the main factors

(Kaempfer and Schneebeli, 2007; Ebner et al., 2015). Applying a temperature gradient, the impact of diffusion is suppressed due to the high recrystallization rate by sublimation and deposition. Due to the low half-life of the ice matrix of a few days, the growth rates are typically on the order of 100 μm per day (Pinzer et al., 2012). Therefore, this redistribution of ice caused by temperature gradient counteracts the diffusion into the solid ice.

By comparing similarities and differences between the outcomes of the three experimental setups we will now discuss the physical processes influencing the interaction and exchange processes within the snowpack between the snow and the advected vapor. We first notice that the final snow isotopic profile of experiment (1) (isothermal) and (2) (positive temperature gradient along the direction of the flow) are comparable to each other. Despite this similarity, the evolution in the outlet water vapor of experiment (1) showed a significantly stronger depletion compared to experiment (2). For experiment (3) (negative temperature gradient along the direction of the flow) we observed the smallest change in outlet water vapor isotopes but the largest snow-pack isotope gradient after the experiment. However, this change was caused by 84 hours flow instead of 24 hours.

Curvature effects, temperature gradients and therefore the recrystallization rate influence the mass transfer of $H_2{}^{16}O$/ $H_2{}^{18}O$ molecules. The higher the recrystallization rate of the snow the slower the adaption of the outlet air concentration to the inlet air concentration (see in experiment (2) and (3)). Under isothermal conditions (experiment (1)) the only effect influencing the recrystallization rate is the curvature effect (Kaempfer and Schneebeli, 2007). However, based on the experimental observations (Kaempfer and Schneebeli, 2007) this effect decreases with decreasing temperature and increasing experimental time. Applying an additional temperature gradient on a snow sample causes complex interplays between local sublimation and deposition on surfaces and the interaction of water molecules in the air with the ice matrix due to changing saturation conditions of the airflow. Therefore, the recrystallization rate increases and causes the change in the $\delta^{18}O$ of the air. For experiment (2) there is a complex interplay between sublimation and deposition of water molecules into the interstitial flow (Ebner et al., 2015c) while for experiment (3) there is deposition of molecules carried by the interstitial flow onto the snow crystals (Ebner et al., 2015b). Furthermore, in the beginning of each experiment there is a tendency to sublimate from edges of the individual snow crystals due to the higher curvature. As the edges were sublimated and deposition occurred in the concavities, the individual snow crystals became more rounded, slowing down the transfer of water molecules into the interstitial airflow. We noticed for all three experiments that within the uncertainty of the isotopic composition of the snow, the initial isotopic composition of the vapor was the same and in isotopic equilibrium with the snow. The difference between experiment (1) and (2) lies in the fact that due to the temperature gradient in experiment (2) there is an increased transfer of water vapor with the isotopic composition of the snow into the airflow. Hence the depleted air from the humidifier advected through the snow disk is mixed with a relatively larger vapor flux from the snow crystals. Additionally, we also expected less deposition into the concavities in experiment (2) compared to experiment (1). However, it is interesting to note that the final isotopic profile of the snow disk is similar in experiment (1) and experiment (2). We interpreted this as being a result of two processes acting in opposite direction: although relatively isotope-depleted vapor from the humidifier was deposited on the ice matrix there was also a higher amount of sublimation of relatively isotope-enriched vapor from the snow disk in experiment (2). Experiment (3) separates itself from the other two experiments in the way that as the water vapor from the humidifier is advected through the snow disk there is a continuous deposition of very depleted air due the negative temperature gradient. As for the case of experiment (1) and (2) there was also in experiment (3) a constant sublimation of the convexities into the vapor stream. We notice that despite the fact that experiment (3) ran for 84 hours the snow at the outlet side of the snow-disk did not become more isotopically depleted compared to experiment (1) and (2). However, the snow on the inlet side became significantly more isotopically depleted. This observation, together with the fact that the vapor of the outlet of the snow-disk is less depleted compared to experiment (1) and (2), leads us to hypothesize that there is a relatively larger deposition of isotopically depleted vapor from the humidifier as the vapor is advected through the snow disk. This means that a relatively larger component of the isotopic composition of the vapor is originating by sublimation from the convexities of the snow disk and less from the isotopically depleted vapor from the humidifier.

Our results and conclusions indicate that there is a need for additional validation. Specifically, it would be crucial to know the mass balance of the snow disk more precisely, which could be done by reconstructing the entire snow disk following the change in density and morphological properties over the entire height. Ideally, the entire sample would be tomographically measured with a resolution of $4 \times 4 \times 4\ mm^3$, each cube corresponding to the representative volume. Insights would also be achieved with experiments using snow of the same isotopic composition, but different SSA, as more precise calculation of the different observed exchange rates would be allowed. Additionally, different and colder background temperatures should be tested to better understand inland Antarctic environment and the effect of the quasi-liquid layer, which is necessary for the development of a numerical model. Isotopically different combinations of vapor and snow should be performed. In the present manuscript, vapor with low $\delta^{18}O$ isotopic composition was transported through snow with relative high $\delta^{18}O$ isotopic composition. It would be interesting to reverse the combination and perform experiments with different combinations to provide more insights on mass and isotope exchanges between vapor and snow. Experiments with longer running time help to understand the change in the ice matrix better under low accumulation conditions.

## 4. Summary and conclusion

Laboratory experimental runs were performed where a transient $\delta^{18}O$ interaction between snow and air was observed. The airflow altered the isotopic composition of the snowpack and supports an improved climatic interpretation of ice core stable water isotope records. The water vapor saturated airflow with an isotopic difference of up to 55‰ changed within 24 h and 84 h the original $\delta^{18}O$ isotope signal in the snow by up to 7.64 ‰ and 15.06 ‰. The disequilibrium between snow and air isotopes led to the observed exchange of isotopes, the rate depending on the temperature gradient conditions. Concluding, increasing the recrystallization rate in the ice matrix causes the temporal change of the $\delta^{18}O$ concentration at the outflow to decrease (experiment (2) and (3)). Decreasing the recrystallization rate causes the temporal curve of the outlet concentration to become steeper reaching the $\delta^{18}O$ inlet concentration of the air faster (experiment (1)).

Additionally, the complex interplay of simultaneous diffusion, sublimation and deposition due to the geometrical complexity of snow has a strong effect on the $\delta^{18}O$ signal in the snow and cannot be neglected. A temporal signal can be superimposed on the precipitation signal, (a) if the snow remains near the surface for a long time, i.e. in a low-accumulation area, and (b) is exposed to a history of air masses carrying vapor with a significantly different isotopic signature than the precipitated snow.

These are novel measurements and will therefore be important as the basis for further research and experiments. Our results represent direct experimental observation of the interaction between the water isotopic composition of the snow, the water vapor in the air and recrystallization due to temperature gradients. Our results demonstrate that recrystallization and bulk mass exchange must be incorporated into future models of snow and firn evolution. Further studies are required on the influence of temperature and airflow as well as snow microstructure on the mass transfer phenomena for validating the implementation of stable water isotopes in snow models.


## Acknowledgements

The Swiss National Science Foundation granted financial support under project Nr. 200020-146540. H.C. Steen-Larsen was supported by the AXA Research Fund. The authors thank K. Fujita, E. D. Waddington and an anonymous reviewer for the suggestions and critical review. M. Jaggi, S. Grimm, A. Schlumpf, and S. Berben gave technical support. The data for this paper are available by contacting the corresponding author.


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**Table 1:** Morphological properties and flow characteristics of the experimental runs: snow density ($\rho$), porosity ($\varepsilon$), specific surface area per unit mass (SSA), mean pore space diameter ($d_{mean}$), superficial velocity in snow ($u_D$), corresponding Reynolds number (Re = $d_{mean} \cdot u_D/\nu_{air}$), average inlet temperature of the humidifier and at the inlet ($T_{in,mean}$), average outlet temperature at the outlet ($T_{out,mean}$), and average temperature gradient ($\nabla T_{ave}$). Experiment (1) corresponds to the isothermal conditions; Experiment (2) to air warming; and Experiment (3) to air cooling in the snow sample.

| | $\rho$ | $\varepsilon$ | SSA | $d_{mean}$ | $u_D$ | Re | $T_{in,mean}$ | $T_{out,mean}$ | $\nabla T_{ave}$ |
|---|---|---|---|---|---|---|---|---|---|
| | kg m$^{-3}$ | – | m$^2$ kg$^{-1}$ | mm | m s$^{-1}$ | – | °C | °C | K m$^{-1}$ |
| Experiment (1) | 201.74 | 0.78 | 28.0 | 0.39 | 0.03 | 0.76 | -15.5 | -15.5 | – |
| Experiment (2) | 201.74 | 0.78 | 29.7 | 0.36 | 0.03 | 0.70 | -15.4 | -14.0 | +47 |
| Experiment (3) | 220.08 | 0.76 | 27.2 | 0.37 | 0.031 | 0.74 | -12.3 | -14.1 | -60 |

**Table 2:** $\delta^{18}O$ in the vapor in the humidifier ($\delta^{18}O_{hum}$) and of the snow in the sample holder ($\delta^{18}O_s$) at the beginning ($t$ = 0) and end ($t$ = end) of each experiment and the final $\delta^{18}O$ content of the snow in the sample holder at the inlet ($z$ = 0 mm) and outlet ($z$ = 30 mm). Experiment (1) corresponds to the isothermal conditions; Experiment (2) to air warming; and Experiment (3) to air cooling in the snow sample.

| | $\delta^{18}O_{hum}$ ‰ | | $\delta^{18}O_{s,\ t=0}$ ‰ | $\delta^{18}O_{s,\ t=\text{end}}$ ‰ | |
|---|---|---|---|---|---|
| | $t$ = 0 | $t$ = end | | $z$ = 0 mm | $z$ = 30 mm |
| Experiment (1) | -68.2 | -67.5 | -10.97 | -17.75 | -15.72 |
| Experiment (2) | -66.3 | -66.1 | -11.94 | -19.60 | -16.60 |
| Experiment (3) | -62.8 | -62.2 | -10.44 | -25.53 | -15.00 |

## Figure captions

**Fig. 1.** Schematic of the experimental setup. A thermocouple (TC) and a humidity sensor (HS) inside the humidifier measured the the mean temperature and humidity of the airflow. Two thermistors (NTC) close to the snow surface measured the inlet and outlet temperature of the airflow (Ebner et al., 2014). The Picarro Analyzer measured the isotopic composition $\delta^{18}O$ of the outlet flow. Inset: 3D structure of 110 × 42 × 110 voxels (2 × 0.75 × 2 $mm^3$) obtained by the μCT.

**Fig. 2.** Temporal isotopic composition of $\delta^{18}O$ of the outflow for each of the experimental runs. The spikes in the $\delta^{18}O$ were due to small temperature changes in the cold laboratory (Ebner et al., 2014). Exp. (1) corresponds to the isothermal conditions; Exp. (2) to air warming; and Exp. (3) to air cooling in the snow sample. The higher the recrystallization rate of the snow the slower the adaption of $\delta^{18}O$ of the outlet air to the inlet air. The illustration in the lower right corner shows the relation between $\delta^{18}O$ of the initial snow, inlet, and outlet of the air.

**Fig. 3.** Spatial isotopic composition of $\delta^{18}O$ of the snow sample at the beginning ($t$ = 0) and at the end ($t$ = end) for each experiment. The air entered at $z$ = 0 mm and exited at $z$ = 30 mm. Exp. (1) corresponds to the isothermal conditions; Exp. (2) to air warming; and Exp. (3) to air cooling in the snow sample.

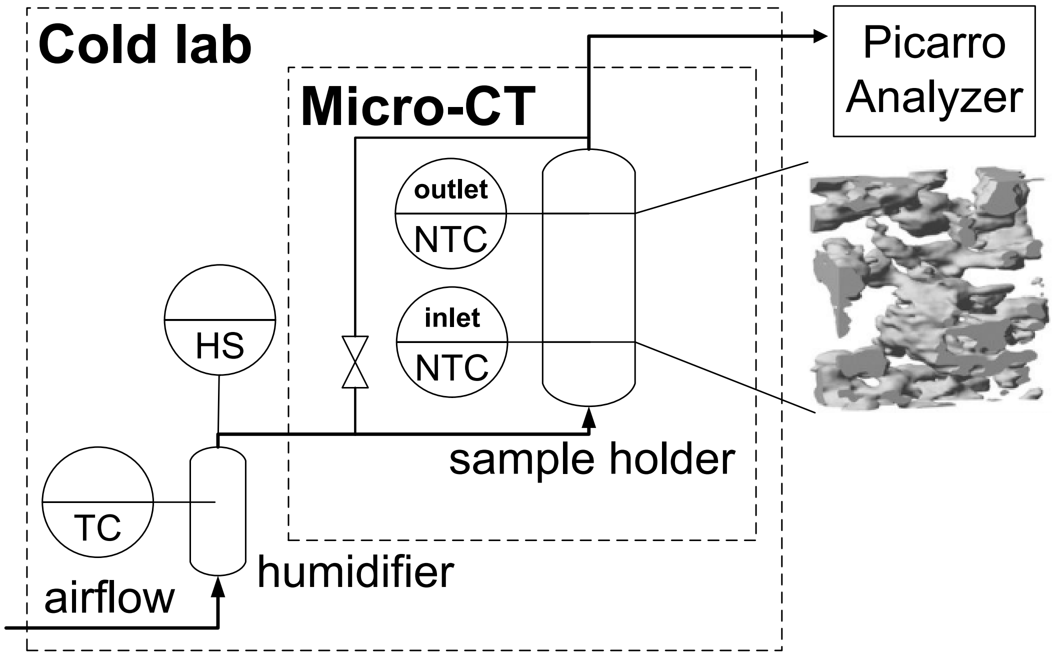
Cold lab
Micro-CT
Picarro Analyzer
outlet
NTC
inlet
NTC
HS
TC
sample holder
humidifier
airflow

Fig. 1

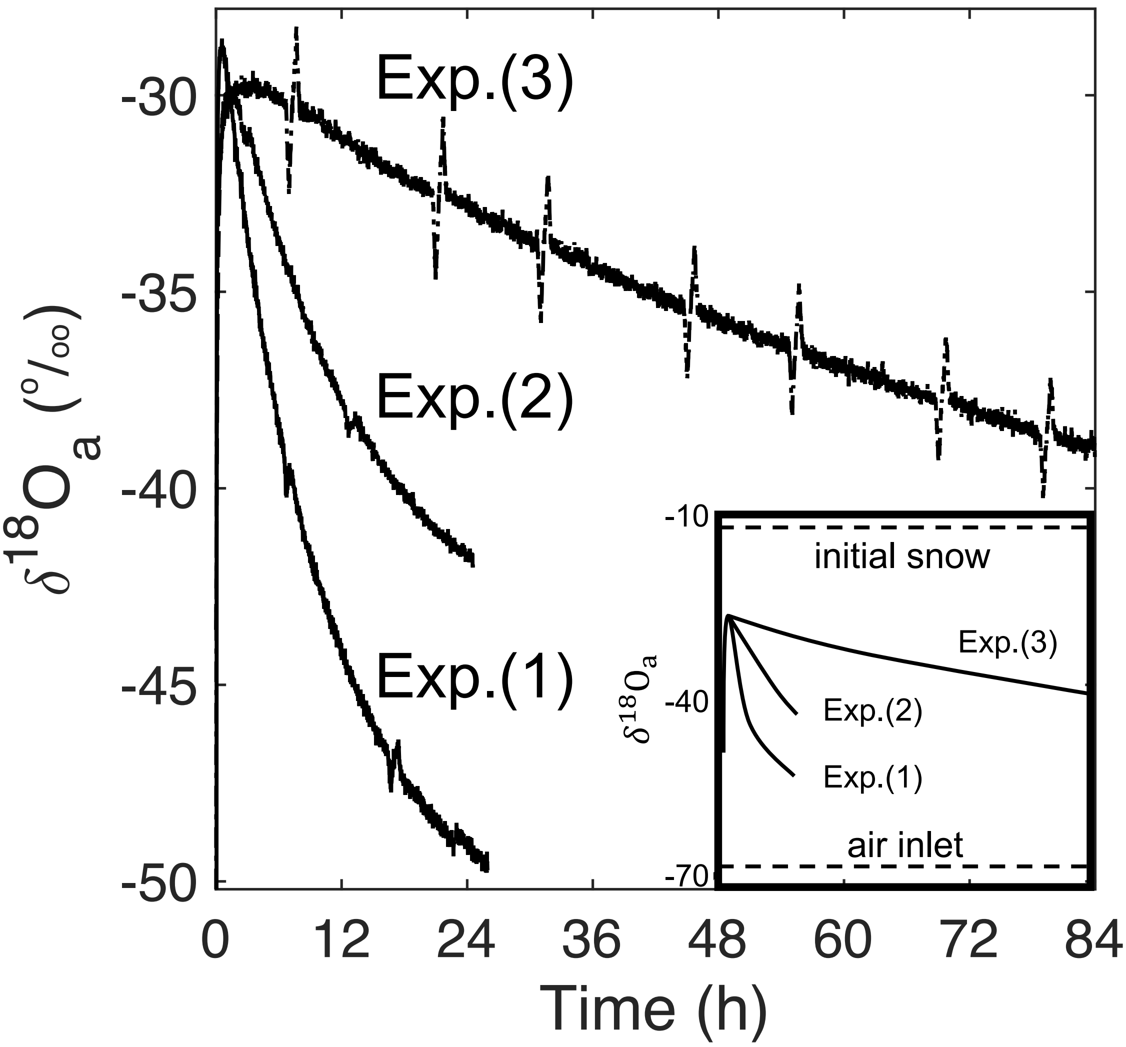
Exp.(3)
Exp.(2)
Exp.(1)
-30
-35
-40
-45
-50
$\delta^{18}O_a$ (‰)
0
12
24
36
48
60
72
84
Time (h)
-10
initial snow
-40
-70
air inlet
$\delta^{18}O_a$

Fig. 2

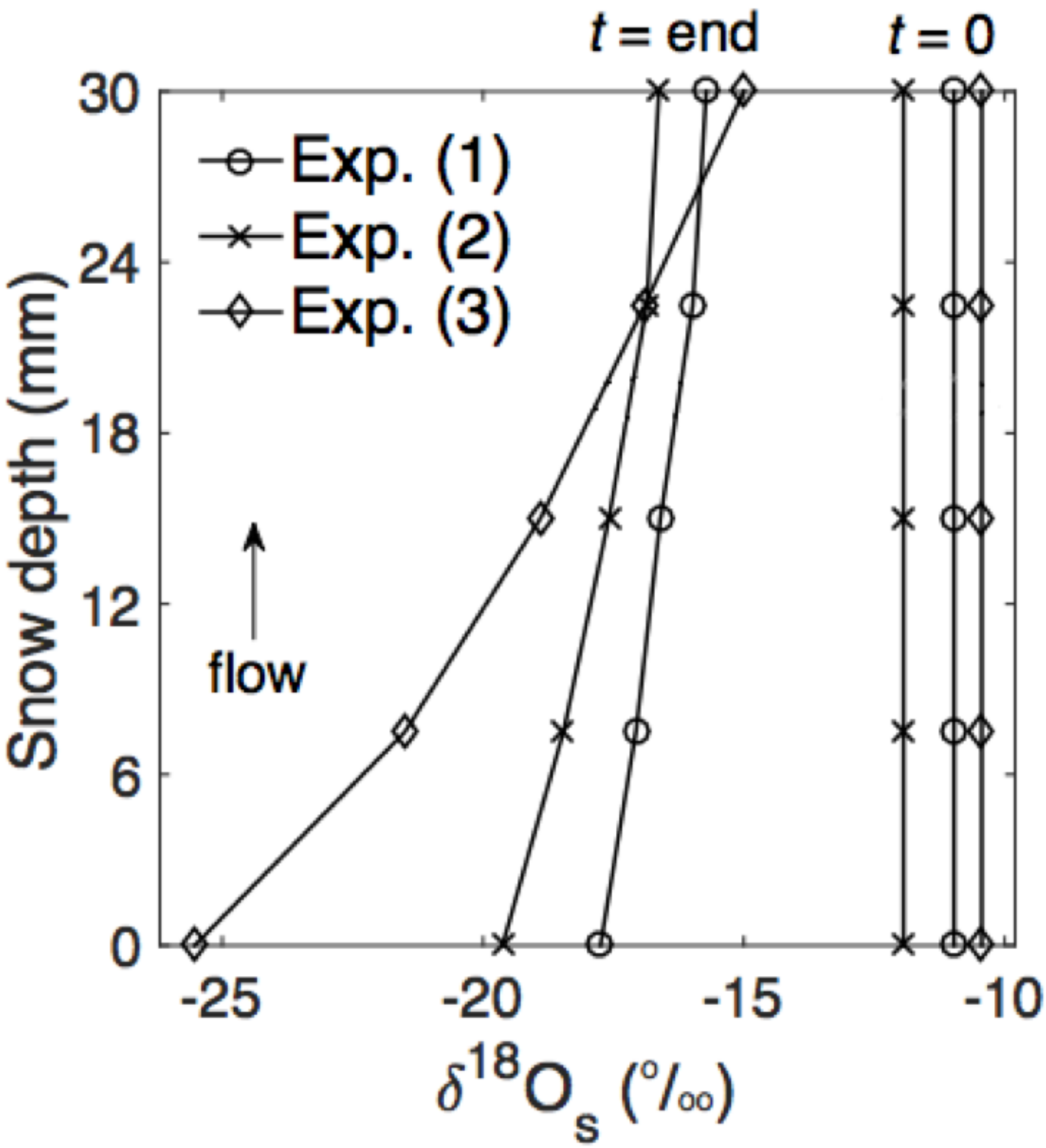
t = end
t = 0
Exp. (1)
Exp. (2)
Exp. (3)
flow
Snow depth (mm)
30
24
18
12
6
0
-25
-20
-15
-10
δ18Os (°/oo)

Fig. 3