er-like crystals (Ebner et al., 2015b). Whisker-like crystals are very small (~10-30 μm) elongated monocrystals. A flow rate dependence of the deposition rate of water vapor deposition at the ice interface was observed, asymptotically approaching an average estimated maximum volumetric deposition rate on the whole sample of $1.05 \cdot 10^{-4}$ kg m^{-3} s^{-1} (Ebner et al., 2015b). Contrarily, if the temperature gradient acts in the same direction of the airflow, the airflow through the snow brings cold and relatively dry air into a warmer area, causing that the pore space air becomes undersaturated, and surrounding ice sublimates. Here, we investigate specifically this last effect.

Sublimation of snow is a fundamental process that affects its crystal structure (Sturm and Benson, 1997), and thus is important for ice core interpretation (Stichler et al., 2001; Ekaykin et al., 2009), as well as calculation of surface energy balance (Box and Steffen, 2001) and mass balance (Déry and Yau, 2002). Kaempfer and Plapp (2009) suggest that condensation of water vapor will have a noticeable effect on the microstructure of snow using a 3D phase-field model, which is also confirmed by a two dimensional finite-element model using airflow velocities, vapor transport and sublimation rates of Albert (2002). Neumann et al. (2009) determined that there is no energy barrier to be overcome during sublimation, and suggest that snow sublimation is limited by vapor diffusion into pore space, rather than by sublimation at the crystal surface.

In the present work, we studied the surface dynamics of snow metamorphism under an induced temperature gradient and saturated airflow in a controlled laboratory experiments. Cold saturated air at around -14 °C was blown into the snow samples and warmed up to around -12.5 °C while flowing across the sample. Sublimation of ice was analyzed by in-situ time-lapse experiment with microcomputer tomography (micro-CT) (Pinzer and Schneebeli, 2009; Chen and Baker, 2010; Pinzer et al., 2012; Wang and Baker, 2014; Ebner et al., 2014) to obtain the discrete-scale geometry of snow. By using discrete-scale geometry, all structures are resolved with a finite resolution corresponding to the voxel size.

2. Time-Lapse tomography experiments

Temperature gradient experiments with fully saturated airflow across snow samples (Ebner et al., 2014) were performed in a cooled micro-CT (Scanco Medical μ-CT80) in a cold laboratory temperature of T_{lab} = -15°C. Cold saturated air was blown into the snow samples and warmed up while flowing across the sample. Aluminum foam including a heating wire was used to warm the side of the snow opposite to the entering air-

② this is a length: 18 μm or 18x18x18 μm³ = 5832 μm³ — this is a volume

flow. We analyzed the following flow rates: a volume flow of 0 (no advection), 0.3, 1.0, and 3.0 liter/min. Higher flow rates were experimentally not possible as shear stresses by airflow destroyed the snow structure (Ebner et al., 2015a). Nature identical snow produced in a cold laboratory (Schleef et al., 2014) was used for the snow sample preparation (water temperature: 30 °C; air temperature: −20 °C). The snow was sieved with a mesh size of 1.4 mm into a box, and was sintered for 27 days at -5°C to increase its strength. The sample holder (diameter: 53 mm; height: 30 mm) was filled by cutting out a cylinder from the sintered snow and pushing into the sample holder without mechanical disturbance of the core. The snow samples were measured with a voxel size of 18 μm³ over 108 h with time-lapse micro-CT measurements taken every 3 h, producing a sequence of 37 images. The size of the cubic voxel size was 18 μm³. The innermost 36.9 mm of the total 53 mm diameter were scanned, and subsamples with a dimension of 7.2 mm × 7.2 mm × 7.2 mm were extracted for further processing. The imaged volume was in the centre of the sample (Fig. 1 c)). A linear encoder with a resolution of less than 1 voxel (< 2 μm) was used to verify that the scans were taken at the same position. The reconstructed micro-CT images were filtered by using a 3 × 3 × 3 median filter followed by a Gaussian filter (σ = 1.4, support = 3). The clustering-based Otsu method (Otsu, 1979) was used to automatically segment the grey-level images into ice and void phase. Morphological properties of the two-phase system were determined based on the geometry obtained by the micro-CT. The segmented data were used to calculate a triangulated ice matrix surface and tetrahedrons inscribed into the ice structure. Morphological parameters such as porosity (ε) and specific surface area (SSA) were then calculated. An opening-based morphological operation was applied to extract the mean pore size of each micro-CT scan (d_{mean}) (Haussener et al., 2012). As additional physical and structural parameter, the effective thermal conductivity k_{cond} was estimated by direct pore-level simulations (DPLS) to determine the influence of changing microstructure. DPLS determined the effective thermal conductivity by solving the governing steady-state heat conduction equations within the solid phase and the stagnant fluid phase (Kaempfer et al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., 2013).

redundant information — 18x18x18 μm³

add information about the position correction procedure mentioned in response #6

→ Specify if 7.2x7.2x7.2 mm³ samples or subsamples (and of which sizes) have been used for the computations described between 109 and 119.

3. Results

Time-lapse tomographic scans were performed with temperature gradients between 43-53 K m^{-1} (Table 1). Small fluctuations of the measured inlet and outlet temperature were due to temperature regulation both inside the cold chamber and inside the micro-CT (Ebner et al., 2014). A shift of $\Delta t < 10$ min between inlet and outlet temperature indicated that a fast equilibrium between the temperature of the snow and the airflow was reached (Albert and Hardy, 1995; Ebner et al., 2015b). The morphological evolution was similar between all four experiments and only a slight rounding and coarsening was visually observed, shown in Fig. 2. The initial ice grains did not change with time and the locations of sublimation and deposition for "ota3" and "ota4" is shown in Fig. 3. Sublimation of 7.7 % and 7.6 % of the ice matrix and deposition of 6.0 % and 9.6 % on the ice matrix were observed. The data were extracted by superposition of vertical cross-sections at 0 and 108 hours with an uncertainty of 6%. The mass sublimated preferentially at locations of the ice matrix with low radii and was relocated leading to a smoothing of the ice surface and to an increase in the size of pores (Fig. 4 a)). The pore size (uncertainty ~6 %) increased by 3.4 %, 3.6 %, 5.4 % and 6.5 % for 'ota1', 'ota2', 'ota3', and 'ota4', respectively.

Loss of ice due to sublimation could not be detected by the micro-CT scans due to limited accuracy and no flow rate dependence was observed during any of the four experiments. The temporal evolution of the porosity, shown in Fig. 4 b), did not change with time and the influence of sublimation of water vapor was not observed. Only 'ota2' showed a slight drop in the temporal evolution of the porosity until 18 h into the experiment but kept constant afterwards. This slight drop (≈ 0.5 %) was probably caused by settling of the snow. Coarsening was observed for each experiment but the influence of changing airflow was not visible, confirmed by the temporal SSA evolution, shown in Fig. 4 c).

The repositioning of water molecules led to a smoothing of the ice grains, but did not affect the thermal conductivity of snow. This quantity (standard deviation ~0.025 W m^{-1}) slightly increased after applying airflow to the temperature gradient, shown in Fig. 4 d), but no flow rate dependence was observed. Every third scan was used to extract the thermal conductivity and a change of -2.6 %, 3.6 %, 2.2 %, and 2.7 % for 'ota1', 'ota2', 'ota3', and 'ota4' was detected.

5. Discussion

The rate of deposition onto the ice surface depends on the flow rate where warm saturated air cooled down while flowing through the sample, as shown in previous experiments (Ebner et al., 2015b). Its deposition rate asymptotically reached a maximum of $1.05 \cdot 10^{-4}$ kg m^{-3} s^{-1}. In this study, changing the temperature gradient leads to a warming up of a cold saturated flow, and resulted in a sublimation rate too small for the analyzed period of the experiment to measure a flow rate dependence by the micro-CT and an influence on the temporal density gradient. A smoothing of ice grains and an increase of the pore space was measured but the airflow velocity did not affect the relocation process of water molecules.

A structural change of the ice grains and repositions of water molecules was observed but the total net flux of the snow was not affected. The superposition of a vertical cross-section in Fig. 3 shows a big effect on reposition of water molecules on the ice structure. However, the temporal porosity (Fig. 4 b) was not affected and the total water vapor net flux was negligible for the analyzed volume. Continued sublimation and deposition of water molecules due the temperature gradient led to a saturation of the pore space. The vapour pressure of the air in the pore was in equilibrium with the water pressure of the ice, given by the local temperature. The entering air warmed up, allowing vapour sublimating from the snow sample to be incorporated into the airflow. As time passed, the snow grains in the sample became more rounded as convexities sublimated. As a result of the reduced curvature, the rate of sublimation decreased and less vapour was deposited in concavities and therefore the surface asperities persisted longer. Finally, the "Kelvin-effect" had a longer impact on the structural change of the ice grains and the reposition of water molecules. In addition, the uptake of water molecules and their transport due to warming during advection was counteracted by diffusion of water molecules due to the temperature gradient. As thermally induced diffusion was opposite to the airflow gradient, a backflow of water vapor occurred and the two opposite fluxes counteracted each other. The Peclet numbers ($Pe = u_D \cdot d_{mean}/D$, where D is the diffusion coefficient of water vapor in air), describing the ratio of mass transfer between diffusion and advection, measured during each experiment, showed that diffusion was still dominant (Table 1). Therefore, water molecules were diffused along the opposite direction to the temperature gradient and advected along the flow direction leading to a back and forth transport of water molecules.

As a Peclet higher than 1 is not possible in snow (Ebner et al., 2015a), advection of cold saturated air into a slightly warmer snowpack has a significant influence not on the total net mass change but on the structural change of the ice grains due to redistribution

Based on the preceding explanations, it seems that these observations can occur either when sublimation is diffusion limited or when it is "reaction"-limited (see comment #2) (5)

personally not convinced by argumentation

of water vapour on the ice matrix. Also the increasing pore size has an influence on the flow field leading to a deceleration of the flow and therefore the interaction of an air-parcel with the ice matrix in the pores increases due to higher residence time. In addition, the diffusive transport rises whereas the advective transport decreases changing the mass transport in the pores. Our results support the hypothesis of Neumann et al. (2009) that sublimation is limited by vapor diffusion into the pore space rather than sublimation at crystal faces. This is supported by the temporal evolution of the porosity (Fig. 4 b)) and the SSA (Fig. 4 c)), as no velocity dependence was observed and the structural changes were too small to be detected by the micro-CT.

The influence of diffusion of water vapor in the direction of the temperature gradient and the influence of the residence time of an air-parcel in the pores were also confirmed by a low mass change at the ice-air interface. Overlapping two consecutive 3D images, the order of magnitude of freshly sublimated ice was detected. The absolute mass change at the ice-air interface (kg m^{-3} s^{-1}) estimated by the experimental results is defined as

$$S_{\mathrm{m,exp}} = \left| \rho_{\mathrm{i}} \frac{\Delta(1-\varepsilon)}{\Delta t} \right| \quad (1)$$

where $\Delta(1\text{-}\varepsilon)$ is the change in the porosity between two images separated by the time step Δt, and ρ_{i} is the density of ice. Albert and McGilvary (1992) and Neumann et al. (2009) presented a model to calculate sublimation rates directly in an aggregate snow sample

$$S_{\mathrm{m}} = \left| h_{\mathrm{m}} \mathrm{SA_V} (\rho_{\mathrm{sat}} - \rho_{\mathrm{v}}) \right| \quad (2)$$

where $\mathrm{SA_V}$ is the specific surface area per volume of snow, and h_{m} is the mass-transfer coefficient (m s^{-1}) given by Neumann et al., (2009)

$$h_{\mathrm{m}} = (0.566 \cdot \mathrm{Re} + 0.075) \cdot 10^{-3} \quad (3)$$

assuming that the sublimation occurs within the first few mm of the sample. Re ($\mathrm{Re} = u_{\mathrm{D}} \cdot d_{\mathrm{mean}} / \nu$ where ν is the kinematic viscosity of the air) is the corresponding Reynolds-number of the flow. The absolute sublimation rate is driven by the difference between the local vapor density (ρ_{v}) and the saturation vapor density (ρ_{sat}) (Neumann et al., 2009; Thorpe and Mason, 1966). Table 2 shows the estimated absolute sublimation rate by the experiment (Eq. (1)) and the model (Eq. (2)). The very small change in porosity due to densification during the first 18 h for 'ota2' was not taken into account. The estimated sublimation rates by the experiment were two orders of magnitude lower than the modeled values and also two orders of magnitude lower than during a negative temperature gradient along an airflow experiment (Ebner et al., 2015b). As the air in the pore space is always saturated (Neumann et al., 2009), the back diffusion of water vapor in the opposite direction of the temperature gradient led to a lower mass transfer rate of sublimation. The flow rate dependence for the model described is shown by the mass-transfer coefficient (Eq. 3), increasing with higher airflow. However, the values calculated from the experiment showed a different trend. Increasing the flow rate led to a lower mass transfer rate due to a lower residence time of the air in the pores. Transfer of heat toward and water vapor away from the sublimating interface may also limit the sublimation rate. In general, the results of the model by Neumann et al. (2009) have to be interpreted with care, as his model was set up to saturate dry air under isothermal conditions. Ice crystals sublimated as dry air enters the snow sample; water vapor was advected throughout the pore space by airflow until saturation vapor pressure was reached, preventing further sublimation. The model by Neumann et al. (2009) does not consider the influence of a temperature gradient and the additional vapor pressure gradient. However, our results concluded that a positive temperature gradient along the airflow has a significant impact on the sublimation rate decreasing the rate by two orders of magnitude.

In the experiments by Neumann et al. (2009), sublimation of snow using dry air under isothermal condition showed a temperature drop for approximately the first 15 min after sublimation started and stayed constant because the latent heat absorption of sublimation for a given flow rate and heat exchange with the sample chamber equalized each other. Such a temperature drop was not observed in our experiments. In the experiments by Neumann et al. (2009) the amount of energy used for sublimation was between -10 and -40 J min^{-1} for saturation of dry air. Using the expected mass change at the ice-air interface $S_{m,exp}$ (Eq. (1)) and the latent heat of sublimation ($L_{sub} \approx 2834.1 \cdot 10^3$ J kg^{-1}) the energy needed for sublimation ranged between -2 and -12 J min^{-1} for our experiments. Our estimated values are a factor up to five lower than the estimated numbers of Neumann et al. (2009), because the entering air was already saturated (with reference to the cold temperature) at the inlet. The needed energy for sublimation could be balanced between the sensible heat carried into and out of the sample, and the exchange of the snow sample with the air stream and the surrounding prevented a temperature drop.

Thermal conductivity changed insignificantly in these experiments, especially for 'ota 1'. This indicates that air warming by a positive temperature gradient along the airflow and an open system reduces or suppresses the increase in thermal conductivity usually observed by temperature gradient metamorphism (Loewe et al., 2013; Calonne et al., 2014). Compared to closed temperature gradient experiment, the applied temperature gradient ~~and the open system induced~~ an air movement and therefore reduced the impact on the thermal conductivity, at least on the short term.

timescales are quite different between experiments, also.

(snow metamorphism and (?))

NB: air movement should be negligible for ota1

→ this is useful to recall that thermal conductivity has been numerically estimated from the geometrical information of the sample only. (no air movement taken into consideration)

6. Summary and conclusion

We performed four experiments of temperature-gradient metamorphism of snow under saturated advective airflow during 108 h. Cold saturated air was blown into the snow samples and warmed up while flowing across the sample. The temperature gradient varied between 43 and 53 K m^{-1} and the snow microstructure was observed by X-ray micro-tomography every 3 h. The micro-CT scans were segmented, and porosity, specific surface area, and the mean pore-size were calculated. Effective thermal conductivity was calculated in direct pore-level simulations (DPLS).

Compared to deposition (shown in Ebner et al., 2015b), sublimation showed a small effect on the structural change of the ice matrix. A change in the pore size was most likely due to sublimation of ice crystals with small radii but a significant loss of water molecules of the snow sample and mass transfer away from the ice interface due to sublimation and advective transport could not be detected by the micro-CT scans and no flow rate dependence was observed. The interaction of mass transport of advection and diffusion of water vapor in the opposite direction of the temperature gradient and the influence of the residence time of an air-parcel in the pores led to a negligible total mass change of the ice. However, a strong reposition of water molecules on the ice grains was observed.

The energy needed for sublimation was too low to see a significant temperature drop because the needed energy was balanced between the sensible heat carried into and out of the sample, and the exchange of the snow sample with the air stream and the surrounding.

This is the third paper of a series analyzing an advective airflow in a snowpack in depth of more than 1 cm. Previous work showed that: (1) under isothermal conditions, the Kelvin-effect leads to a saturation of the pore space in the snow but did 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 and creation of whisker-like structures due to deposition of water molecules on the ice matrix (Ebner et al., 2015b); and (3) a positive temperature gradient along to the flow had a negligible total mass change of the ice but a strong reposition effect of water molecules on the ice grains, shown in this paper. Conditions (1) and (3) showed that they have a negligible effect on the porosity evolution of the ice matrix. Porosity changes can be neglected to improve models for snow compaction and evolution at the surface. In contrast, conditions (2) showed a significant impact on the structural evolution and seems to be essential for such snowpack models and other numerical simulations. Nevertheless, the strong reposition of water molecules on the ice grains observed for all conditions (1) – (3) can have a significant impact on atmospheric chemistry and isotopic changes in snow.

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