

**RESPONSE TO REFEREES' F. FLIN COMMENTS
TO MANUSCRIPT TC-2015-158**

Title: Metamorphism during temperature gradient with undersaturated advective airflow in a snow sample

Authors: Pirmin Philipp Ebner, Martin Schneebeil, and Aldo Steinfeld

We thank the referee F. Flin for the constructive comments and corrections. We agree with his comments and changed the corresponding typos and unclear statements (see below).

The authors

er-like crystals (Ebner et al., 2015b). Whisker-like crystals are very small ($\sim 10\text{-}30\text{ }\mu\text{m}$) 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 $1.05 \cdot 10^{-4}\text{ kg m}^{-3}\text{ s}^{-1}$ (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 $-14\text{ }^{\circ}\text{C}$ was blown into the snow samples and warmed up to around $-12.5\text{ }^{\circ}\text{C}$ 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 $\mu\text{-CT80}$) in a cold laboratory temperature of $T_{\text{lab}} = -15^{\circ}\text{C}$. 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-

90 flow. We analyzed the following flow rates: a volume flow of 0 (no advection), 0.3, 1.0,
 91 and 3.0 liter/min. Higher flow rates were experimentally not possible as shear stresses
 92 by airflow destroyed the snow structure (Ebner et al., 2015a). Nature identical snow
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 94 ration (water temperature: 30 °C; air temperature: -20 °C). The snow was sieved with a
 95 mesh size of 1.4 mm into a box, and was sintered for 27 days at -5 °C to increase its
 96 strength. The sample holder (diameter: 53 mm; height: 30 mm) was filled by cutting out
 97 a cylinder from the sintered snow and pushing into the sample holder without mechani-
 98 cal disturbance of the core. The snow samples were measured with a voxel size of 18
 99 μm^3 over 108 h with time-lapse micro-CT measurements taken every 3 h, producing a
 100 sequence of 37 images. The size of the cubic voxel size was 18 μm . The innermost 36.9
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 103 in the centre of the sample (Fig. 1 c)). A linear encoder with a resolution of less than 1
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 105 reconstructed micro-CT images were filtered by using a $3 \times 3 \times 3$ median filter followed
 106 by a Gaussian filter ($\sigma = 1.4$, support = 3). The clustering-based Otsu method (Otsu,
 107 1979) was used to automatically segment the grey-level images into ice and void phase.
 108 Morphological properties of the two-phase system were determined based on the geom-

109 etry obtained by the micro-CT. The segmented data were used to calculate a triangulat-
 110 ed ice matrix surface and tetrahedrons inscribed into the ice structure. Morphological
 111 parameters such as porosity (ϵ) and specific surface area (SSA) were then calculated. An
 112 opening-based morphological operation was applied to extract the mean pore size of
 113 each micro-CT scan (d_{mean}) (Haussener et al., 2012). As additional physical and struc-
 114 tural parameter, the effective thermal conductivity k_{cond} was estimated by direct pore-
 115 level simulations (DPLS) to determine the influence of changing microstructure. DPLS
 116 determined the effective thermal conductivity by solving the governing steady-state heat
 117 conduction equations within the solid phase and the stagnant fluid phase (Kaempfer et
 118 al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., 2013).

119 *> Specify if $7.2 \times 7.2 \times 7.2 \text{ mm}^3$ samples or subsamples have been used for the computations described between 109 and 119.*

120 **3. Results**

121 Time-lapse tomographic scans were performed with temperature gradients between
 122 43-53 K m^{-1} (Table 1). Small fluctuations of the measured inlet and outlet temperature

②
 this is a length
 18 μm
 or
 $18 \times 18 \times 18 \mu\text{m}^3$
 $= 5832 \mu\text{m}^3$
 this is a volume



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

*redundant information
 $18 \times 18 \times 18 \mu\text{m}^3$*

(and of which size)

during the 108h experiment

123 were due to temperature regulation both inside the cold chamber and inside the micro-
 124 CT (Ebner et al., 2014). A shift of $\Delta t < 10$ min between inlet and outlet temperature in-
 125 dicated that a fast equilibrium between the temperature of the snow and the airflow was
 126 reached (Albert and Hardy, 1995; Ebner et al., 2015b). The morphological evolution
 127 was similar between all four experiments and only a slight rounding and coarsening was
 128 visually observed, shown in Fig. 2). The initial ice grains did not change with time and
 129 the locations of sublimation and deposition for “ota3” and “ota4” is shown in Fig. 3.
 130 Sublimation of 7.7 % and 7.6 % of the ice matrix and deposition of 6.0 % and 9.6 % on
 131 the ice matrix were observed. The data were extracted by superposition of vertical
 132 cross-sections at 0 and 108 hours with an uncertainty of 6%. The mass sublimated pref-
 133 erentially at locations of the ice matrix with low radii and was relocated leading to a
 134 smoothing of the ice surface and to an increase in the size of pores (Fig. 4 a)). The pore
 135 size (uncertainty ~ 6 %) increased by 3.4 %, 3.6 %, 5.4 % and 6.5 % for ‘ota1’, ‘ota2’,
 136 ‘ota3’, and ‘ota4’, respectively.

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

146 The repositioning of water molecules led to a smoothing of the ice grains, but did
 147 not affect the thermal conductivity of snow. This quantity (standard deviation ~ 0.025 W
 148 m^{-1}) slightly increased after applying airflow to the temperature gradient, shown in Fig.
 149 4 d), but no flow rate dependence was observed. Every third scan was used to extract
 150 the thermal conductivity and a change of -2.6 %, 3.6 %, 2.2 %, and 2.7 % for ‘ota1’,
 151 ‘ota2’, ‘ota3’, and ‘ota4’ was detected.

152

153 5. Discussion

154 The rate of deposition onto the ice surface depends on the flow rate where warm
 155 saturated air cooled down while flowing through the sample, as shown in previous ex-

156 periments (Ebner et al., 2015b). Its deposition rate asymptotically reached a maximum
 157 of $1.05 \cdot 10^{-4} \text{ kg m}^{-3} \text{ s}^{-1}$. In this study, changing the temperature gradient leads to a
 158 warming up of a cold saturated flow, and resulted in a sublimation rate too small for the
 159 analyzed period of the experiment to measure a flow rate dependence by the micro-CT
 160 and an influence on the temporal density gradient. A smoothing of ice grains and an in-
 161 crease of the pore space was measured but the airflow velocity did not affect the reloca-
 162 tion process of water molecules. *pore size* (porosity is constant.)

163 A structural change of the ice grains and repositions of water molecules was ob-
 164 served but the total net flux of the snow was not affected. The superposition of a vertical
 165 cross-section in Fig. 3 shows a big effect on reposition of water molecules on the ice
 166 structure. However, the temporal porosity (Fig. 4 b) was not affected and the total water
 167 vapor net flux was negligible for the analyzed volume. Continued sublimation and dep-
 168 osition of water molecules due ^(to) the temperature gradient led to a saturation of the pore
 169 space. The vapour pressure of the air in the pore was in equilibrium with the water pres-
 170 sure of the ice, given by the local temperature. The entering air warmed up, allowing
 171 vapour sublimating from the snow sample to be incorporated into the airflow. As time
 172 passed, the snow grains in the sample became more rounded as convexities sublimated.
 173 As a result of the reduced curvature, the rate of sublimation decreased and less vapour
 174 was deposited in concavities and therefore the surface asperities persisted longer. Final-
 175 ly, the “Kelvin-effect” had a longer impact on the structural change of the ice grains and
 176 the reposition of water molecules. In addition, the uptake of water molecules and their
 177 transport due to warming during advection was counteracted by diffusion of water mol-
 178 ecules due to the temperature gradient. As thermally induced diffusion was opposite to
 179 the airflow gradient, a backflow of water vapor occurred and the two opposite fluxes
 180 counteracted each other. The Peclet numbers ($Pe = u_D \cdot d_{\text{mean}} / D$, where D is the diffusion
 181 coefficient of water vapor in air), describing the ratio of mass transfer between diffusion
 182 and advection, measured during each experiment, showed that diffusion was still domi-
 183 nant (Table 1). Therefore, water molecules were diffused along the opposite direction to
 184 the temperature gradient and advected along the flow direction leading to a back and
 185 forth transport of water molecules.

186 As a Peclet higher than 1 is not possible in ^{material} snow (Ebner et al., 2015a), advection of
 187 cold saturated air into a slightly warmer snowpack has a significant influence not on the
 188 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

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

198 The influence of diffusion of water vapor in the direction of the temperature gradi-
199 ent and the influence of the residence time of an air-parcel in the pores were also con-
200 firmed by a low mass change at the ice-air interface. Overlapping two consecutive 3D
201 images, the order of magnitude of freshly sublimated ice was detected. The absolute
202 mass change at the ice-air interface ($\text{kg m}^{-3} \text{s}^{-1}$) estimated by the experimental results is
203 defined as

$$204 \quad S_{\text{m,exp}} = \left| \rho_i \frac{\Delta(1-\varepsilon)}{\Delta t} \right| \quad (1)$$

205 where $\Delta(1-\varepsilon)$ is the change in the porosity between two images separated by the time
206 step Δt , and ρ_i is the density of ice. Albert and McGilvary (1992) and Neumann et al.
207 (2009) presented a model to calculate sublimation rates directly in an aggregate snow
208 sample

$$209 \quad S_{\text{m}} = |h_{\text{m}} \text{SA}_{\text{v}} (\rho_{\text{sat}} - \rho_{\text{v}})| \quad (2)$$

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

$$212 \quad h_{\text{m}} = (0.566 \cdot \text{Re} + 0.075) \cdot 10^{-3} \quad (3)$$

213 assuming that the sublimation occurs within the first few mm of the sample. Re ($\text{Re} =$
214 $u_{\text{D}} \cdot d_{\text{mean}} / \nu$ where ν is the kinematic viscosity of the air) is the corresponding Reynolds-
215 number of the flow. The absolute sublimation rate is driven by the difference between
216 the local vapor density (ρ_{v}) and the saturation vapor density (ρ_{sat}) (Neumann et al., 2009;
217 Thorpe and Mason, 1966). Table 2 shows the estimated absolute sublimation rate by the
218 experiment (Eq. (1)) and the model (Eq. (2)). The very small change in porosity due to
219 densification during the first 18 h for 'ota2' was not taken into account. The estimated
220 sublimation rates by the experiment were two orders of magnitude lower than the mod-

221 eled values and also two orders of magnitude lower than during a negative temperature
 222 gradient along an airflow experiment (Ebner et al., 2015b). As the air in the pore space
 223 is always saturated (Neumann et al., 2009), the back diffusion of water vapor in the op-
 224 posite direction of the temperature gradient led to a lower mass transfer rate of sublima-
 225 tion. The flow rate dependence for the model described is shown by the mass-transfer
 226 coefficient (Eq. 3), increasing with higher airflow. However, the values calculated from
 227 the experiment showed a different trend. Increasing the flow rate led to a lower mass
 228 transfer rate due to a lower residence time of the air in the pores. Transfer of heat to-
 229 ward and water vapor away from the sublimating interface may also limit the sublima-
 230 tion rate. In general, the results of the model by Neumann et al. (2009) have to be inter-
 231 preted with care, as his model was set up to saturate dry air under isothermal conditions.
 232 Ice crystals sublimated as dry air enters the snow sample; water vapor was advected
 233 throughout the pore space by airflow until saturation vapor pressure was reached, pre-
 234 venting further sublimation. The model by Neumann et al. (2009) does not consider the
 235 influence of a temperature gradient and the additional vapor pressure gradient. Howev-
 236 er, our results concluded that a positive temperature gradient along the airflow has a
 237 significant impact on the sublimation rate, decreasing the rate by two orders of magni-
 238 tude.

239 In the experiments by Neumann et al. (2009), sublimation of snow using dry air un-
 240 der isothermal condition showed a temperature drop for approximately the first 15 min
 241 after sublimation started and stayed constant because the latent heat absorption of sub-
 242 limation for a given flow rate and heat exchange with the sample chamber equalized
 243 each other. Such a temperature drop was not observed in our experiments. In the exper-
 244 iments by Neumann et al. (2009) the amount of energy used for sublimation was be-
 245 tween -10 and -40 J min^{-1} for saturation of dry air. Using the expected mass change at
 246 the ice-air interface $S_{m,exp}$ (Eq. (1)) and the latent heat of sublimation ($L_{sub} \approx 2834.1 \cdot$
 247 10^3 J kg^{-1}) the energy needed for sublimation ranged between -2 and -12 J min^{-1} for our
 248 experiments. Our estimated values are a factor up to five lower than the estimated num-
 249 bers of Neumann et al. (2009), because the entering air was already saturated (with ref-
 250 erence to the cold temperature) at the inlet. The needed energy for sublimation could be
 251 balanced between the sensible heat carried into and out of the sample, and the exchange
 252 of the snow sample with the air stream and the surrounding prevented a temperature
 253 drop.

254 Thermal conductivity changed insignificantly in these experiments, especially for
 255 'ota 1'. This indicates that air warming by a positive temperature gradient along the air-
 256 flow and an open system reduces or suppresses the increase in thermal conductivity
 257 usually observed by temperature gradient metamorphism (Loewe et al., 2013; Calonne
 258 et al., 2014). *temperatures are quite different between experiments, also.*

259 ture gradient and the open system induced an air movement and therefore reduced the
 260 impact on the thermal conductivity, at least on the short term. *air movement should be negligible for ota?*

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

263 We performed four experiments of temperature-gradient metamorphism of snow
 264 under saturated advective airflow during 108 h. Cold saturated air was blown into the
 265 snow samples and warmed up while flowing across the sample. The temperature gradi-
 266 ent varied between 43 and 53 K m⁻¹ and the snow microstructure was observed by X-ray
 267 micro-tomography every 3 h. The micro-CT scans were segmented, and porosity, spe-
 268 cific surface area, and the mean pore-size were calculated. Effective thermal conductivi-
 269 ty was calculated in direct pore-level simulations (DPLS).

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

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

284 This is the third paper of a series analyzing an advective airflow in a snowpack in
 285 depth of more than 1 cm. Previous work showed that: (1) under isothermal conditions,

286 the Kelvin-effect leads to a saturation of the pore space in the snow but did not affect
 287 the structural change (Ebner et al., 2015a); (2) applying a negative temperature gradient
 288 along the flow direction leads to a change in the microstructure and creation of whisker-
 289 like structures due to deposition of water molecules on the ice matrix (Ebner et al.,
 290 2015b); and (3) a positive temperature gradient along the flow had a negligible total
 291 mass change of the ice but a strong reposition effect of water molecules on the ice
 292 grains, shown in this paper. Conditions (1) and (3) showed that they have a negligible
 293 effect on the porosity evolution of the ice matrix. *except when settling is concerned*
 294 Porosity changes can be neglected to
 295 improve models for snow compaction and evolution at the surface. In contrast, condi-
 296 tions (2) showed a significant impact on the structural evolution and seems to be essen-
 297 tial for such snowpack models and other numerical simulations. Nevertheless, the strong
 298 reposition of water molecules on the ice grains observed for all conditions (1) – (3) can
 299 have a significant impact on atmospheric chemistry and isotopic changes in snow.

300 Acknowledgements

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 303 constructive reviews and M. Jaggi, S. Grimm, H. Löwe for technical and modelling
 304 support.

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Metamorphism during temperature gradient with undersaturated advective airflow in a snow sample

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Abstract

Snow at or close to the surface commonly undergoes temperature gradient metamorphism under advective flow, which alters its microstructure and physical properties. Time-lapse X-ray micro-tomography is applied to investigate the structural dynamics of temperature gradient snow metamorphism exposed to an advective airflow in controlled laboratory conditions. Cold saturated air at the inlet was blown into the snow samples and warmed up while flowing across the sample with a temperature gradient of around 50 K m⁻¹. Changes of the porous ice structure were observed at mid-height of the snow sample. Sublimation occurred due to the slight undersaturation of the incoming air into the warmer ice matrix. Diffusion of water vapor opposite to the direction of the temperature gradient counteracted the mass transport of advection. Therefore, the total net ice change was negligible leading to a constant porosity profile. However, the strong recrystallization of water molecules in snow may impact its isotopic or chemical content.

Keywords: snow, temperature gradient, metamorphism, advection, sublimation, tomography

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24 **1. Introduction**

25 Snow has a complex porous microstructure and consists of a continuous ice struc-
26 ture made of grains connected by bonds and inter-connecting pores (Löwe et al., 2011).
27 It has a high permeability (Calonne et al, 2012, Zermatten et al., 2014) and under ap-
28 propriate conditions airflow through the snow structure can occur (Sturm and Johnson,
29 1991) due to variation of surface pressure (Colbeck, 1989; Albert and Hardy, 1995),
30 simultaneous warming and cooling, and induced temperature gradients (Sturm and
31 Johnson, 1991). Both diffusive and advective airflows affect heat and mass transport in
32 the snowpack and influence chemical concentrations (Gjessing, 1977; Waddington et
33 al., 1996). Various airflow conditions in a snow sample occur, namely: isothermal air-
34 flow, air cooling by a negative temperature gradient along the airflow leading to a local
35 supersaturation of the air, and air warming by a positive temperature gradient along the
36 airflow leading to a local undersaturation of the air (Fig. 1). In general, in a natural
37 snowpack close to the surface ($< 1\text{ cm}$) two additional conditions can occur: (1) warm air
38 enters a snowpack having a positive temperature gradient leading to a supersaturation of
39 the air at the entrance, and (2) cold air enters a snowpack having a negative temperature
40 gradient leading to an undersaturation of the air at the entrance. However, because snow
41 has a high heat capacity compared to the air, a high specific surface area, and therefore a
42 high convective heat transfer to the air, a “quasi” thermal equilibrium (the term “quasi”
43 is used because normally the snow structure continuously changes and therefore the
44 equilibrium conditions as well) is usually assumed inside the snowpack ($> 1\text{ cm}$). In this
45 paper, only conditions deeper than $> 1\text{ cm}$ inside a snowpack are considered. Under iso-
46 thermal condition, the continuous sublimation and deposition of ice is due to higher va-
47 por pressure over convex surfaces and lower vapor pressure over concave surfaces, re-
48 spectively (Kelvin-effect) (Neumann et al., 2008; Ebner et al., 2014). However, apply-
49 ing a fully isothermal saturated airflow across a snow sample has been shown to have
50 no influence on the coarsening rate that is typical for isothermal snow metamorphism
51 independently of the transport regime in the pores at a physically possible Peclet-
52 number (Ebner et al., 2015a). When applying a temperature gradient, the effect of sub-
53 limation and deposition in the snow results from interaction between snow temperature
54 and the local relative humidity in the pores. If vapor is advected from a warmer zone
55 into a colder zone, the air becomes supersaturated, and some water vapor deposits onto
56 the surrounding ice grains. This leads to a change in the microstructure creating whisk-

er-like crystals (Ebner et al., 2015b). Whisker-like crystals are very small ($\sim 10\text{-}30\text{ }\mu\text{m}$) 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 $1.05 \cdot 10^{-4}\text{ kg m}^{-3}\text{ s}^{-1}$ (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 $-14\text{ }^{\circ}\text{C}$ was blown into the snow samples and warmed up to around $-12.5\text{ }^{\circ}\text{C}$ 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 of $18\text{ }\mu\text{m}$.

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 $\mu\text{-CT}80$) in a cold laboratory temperature of $T_{\text{lab}} = -14^{\circ}\text{C}$. 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-

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 99 μm^3 over 108 h with time-lapse micro-CT measurements taken every 3 h, producing a
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 101 mm of the total 53 mm diameter were scanned, and subsamples with a dimension of 7.2
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 104 voxel ($< 2 \mu\text{m}$) was used to verify that the scans were taken at the same position. ~~Addi-~~
 105 ~~tionally, a cross-correlation function was applied to suppress all erroneous data from the~~
 106 ~~dataset.~~ The reconstructed micro-CT images were filtered by using a $3 \times 3 \times 3$ median
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 110 based on the geometry obtained by the micro-CT. The segmented data were used to cal-
 111 culate a triangulated ice matrix surface and tetrahedrons inscribed into the ice structure.
 112 Morphological parameters such as porosity (ϵ) and specific surface area (SSA) were
 113 then calculated ~~of subsamples of the size of 6.3 mm \times 6.3 mm \times 6.3 mm samples.~~ An
 114 opening-based morphological operation was applied to extract the mean pore size of
 115 each micro-CT scan (d_{mean}) (Haussener et al., 2012). As additional physical and struc-
 116 tural parameter, the effective thermal conductivity k_{cond} was estimated by direct pore-
 117 level simulations (DPLS) to determine the influence of changing microstructure. DPLS
 118 determined the effective thermal conductivity by solving the governing steady-state heat
 119 conduction equations within the solid phase and the stagnant fluid phase (Kaempfer et
 120 al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., 2013).

121

122 3. Results

123 Time-lapse tomographic scans were performed with temperature gradients between
124 43-53 K m⁻¹ (Table 1). Small fluctuations of the measured inlet and outlet temperature
125 were due to temperature regulation both inside the cold chamber and inside the micro-
126 CT (Ebner et al., 2014). A shift of $\Delta t < 10$ min between inlet and outlet temperature in-
127 dicated that a fast equilibrium between the temperature of the snow and the airflow was
128 reached (Albert and Hardy, 1995; Ebner et al., 2015b). The morphological evolution
129 was similar between all four experiments and only a slight rounding and coarsening was
130 visually observed during the 108 h experiment shown in (Fig. 2). The initial ice grains
131 did not change with time and the locations of sublimation and deposition for “ota3” and
132 “ota4” is shown in Fig. 3. Sublimation of 7.7 % and 7.6 % of the ice matrix and deposi-
133 tion of 6.0 % and 9.6 % on the ice matrix were observed. The data were extracted by
134 superposition of vertical cross-sections at 0 and 108 hours with an uncertainty of 6%.
135 The mass sublimated preferentially at locations of the ice matrix with low radii and was
136 relocated leading to a smoothing of the ice surface and to an increase in the size of pores
137 (Fig. 4 a)). The pore size (uncertainty ~6 %) increased by 3.4 %, 3.6 %, 5.4 % and 6.5
138 % for ‘ota1’, ‘ota2’, ‘ota3’, and ‘ota4’, respectively.

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

148 The repositioning of water molecules led to a smoothing of the ice grains, but did
149 not affect the thermal conductivity of snow. This quantity (standard deviation ~ 0.025 W
150 m⁻¹) slightly increased after applying airflow to the temperature gradient, shown in Fig.
151 4 d), but no flow rate dependence was observed. Every third scan was used to extract
152 the thermal conductivity and a change of -2.6 %, 3.6 %, 2.2 %, and 2.7 % for ‘ota1’,
153 ‘ota2’, ‘ota3’, and ‘ota4’ was detected.

154

155 5. Discussion

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

165 A structural change of the ice grains and repositions of water molecules was ob-
 166 served but the total net flux of the snow was not affected. The superposition of a vertical
 167 cross-section in Fig. 3 shows a big effect on reposition of water molecules on the ice
 168 structure. However, the temporal porosity (Fig. 4 b) was not affected and the total water
 169 vapor net flux was negligible for the analyzed volume. Continued sublimation and dep-
 170 osition of water molecules due to the temperature gradient led to a saturation of the pore
 171 space. The vapour pressure of the air in the pore was in equilibrium with the water pres-
 172 sure of the ice, given by the local temperature. The entering air warmed up, allowing
 173 vapour sublimating from the snow sample to be incorporated into the airflow. As time
 174 passed, the snow grains in the sample became more rounded as convexities sublimated.
 175 As a result of the reduced curvature, the rate of sublimation decreased and less vapour
 176 was deposited in concavities and therefore the surface asperities persisted longer. Final-
 177 ly, the “Kelvin-effect” had a longer impact on the structural change of the ice grains and
 178 the reposition of water molecules. In addition, the uptake of water molecules and their
 179 transport due to warming during advection was counteracted by diffusion of water mol-
 180 ecules due to the temperature gradient. As thermally induced diffusion was opposite to
 181 the airflow gradient, a backflow of water vapor occurred and the two opposite fluxes
 182 counteracted each other. The Peclet numbers ($Pe = u_D \cdot d_{\text{mean}} / D$, where D is the diffusion
 183 coefficient of water vapor in air), describing the ratio of mass transfer between diffusion
 184 and advection, measured during each experiment, showed that diffusion was still domi-
 185 nant (Table 1). Therefore, water molecules were diffused along the opposite direction to
 186 the temperature gradient and advected along the flow direction leading to a back and
 187 forth transport of water molecules.

188 As a Peclet higher than 1 is not possible in **natural** snow (Ebner et al., 2015a), ad-
 189 vection of cold saturated air into a slightly warmer snowpack has a significant influence
 190 not on the total net mass change but on the structural change of the ice grains due to re-
 191 distribution of water vapour on the ice matrix. Also the increasing pore size has an in-
 192 fluence on the flow field leading to a deceleration of the flow and therefore the interac-
 193 tion of an air-parcel with the ice matrix in the pores increases due to higher residence
 194 time. In addition, the diffusive transport rises whereas the advective transport decreases,
 195 changing the mass transport in the pores. Our results support the hypothesis of Neu-
 196 mann et al. (2009) that sublimation is limited by vapor diffusion into the pore space ~~fa-~~
 197 ~~ther than sublimation~~ or kinetics effects (reaction effect) at crystal faces. This is sup-
 198 ported by the temporal evolution of the porosity (Fig. 4 b)) and the SSA (Fig. 4 c)), as
 199 no velocity dependence was observed and the structural changes were too small to be
 200 detected by the micro-CT.

201 The influence of diffusion of water vapor in the direction of the temperature gradi-
 202 ent and the influence of the residence time of an air-parcel in the pores were also con-
 203 firmed by a low mass change at the ice-air interface. Overlapping two consecutive 3D
 204 images, the order of magnitude of freshly sublimated ice was detected. The absolute
 205 mass change at the ice-air interface ($\text{kg m}^{-3} \text{ s}^{-1}$) estimated by the experimental results is
 206 defined as

$$207 \quad S_{\text{m,exp}} = \left| \rho_i \frac{\Delta(1-\varepsilon)}{\Delta t} \right| \quad (1)$$

208 where $\Delta(1-\varepsilon)$ is the change in the porosity between two images separated by the time
 209 step Δt , and ρ_i is the density of ice. Albert and McGilvary (1992) and Neumann et al.
 210 (2009) presented a model to calculate sublimation rates directly in an aggregate snow
 211 sample

$$212 \quad S_{\text{m}} = |h_{\text{m}} \text{SA}_v (\rho_{\text{sat}} - \rho_v)| \quad (2)$$

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

$$215 \quad h_{\text{m}} = (0.566 \cdot \text{Re} + 0.075) \cdot 10^{-3} \quad (3)$$

216 assuming that the sublimation occurs within the first few mm of the sample. Re ($\text{Re} =$
 217 $u_{\text{D}} \cdot d_{\text{mean}} / \nu$ where ν is the kinematic viscosity of the air) is the corresponding Reynolds-
 218 number of the flow. The absolute sublimation rate is driven by the difference between
 219 the local vapor density (ρ_v) and the saturation vapor density (ρ_{sat}) (Neumann et al., 2009;

220 Thorpe and Mason, 1966). Table 2 shows the estimated absolute sublimation rate by the
 221 experiment (Eq. (1)) and the model (Eq. (2)). The very small change in porosity due to
 222 densification during the first 18 h for ‘ota2’ was not taken into account. The estimated
 223 sublimation rates by the experiment were two orders of magnitude lower than the mod-
 224 eled values and also two orders of magnitude lower than during a negative temperature
 225 gradient along an airflow experiment (Ebner et al., 2015b). As the air in the pore space
 226 is always saturated (Neumann et al., 2009), the back diffusion of water vapor in the op-
 227 posite direction of the temperature gradient led to a lower mass transfer rate of sublima-
 228 tion. The flow rate dependence for the model described is shown by the mass-transfer
 229 coefficient (Eq. 3), increasing with higher airflow. However, the values calculated from
 230 the experiment showed a different trend. Increasing the flow rate led to a lower mass
 231 transfer rate due to a lower residence time of the air in the pores. Transfer of heat to-
 232 ward and water vapor away from the sublimating interface may also limit the sublima-
 233 tion rate. In general, the results of the model by Neumann et al. (2009) have to be inter-
 234 preted with care, as his model was set up to saturate dry air under isothermal conditions.
 235 Ice crystals sublimated as dry air enters the snow sample; water vapor was advected
 236 throughout the pore space by airflow until saturation vapor pressure was reached, pre-
 237 venting further sublimation. The model by Neumann et al. (2009) does not consider the
 238 influence of a temperature gradient and the additional vapor pressure gradient. Howev-
 239 er, our results concluded that a positive temperature gradient along the airflow has a
 240 significant impact on the sublimation rate, decreasing the rate by two orders of magni-
 241 tude.

242 In the experiments by Neumann et al. (2009), sublimation of snow using dry air un-
 243 der isothermal condition showed a temperature drop for approximately the first 15 min
 244 after sublimation started and stayed constant because the latent heat absorption of sub-
 245 limation for a given flow rate and heat exchange with the sample chamber equalized
 246 each other. Such a temperature drop was not observed in our experiments. In the exper-
 247 iments by Neumann et al. (2009) the amount of energy used for sublimation was be-
 248 tween -10 and -40 J min^{-1} for saturation of dry air. Using the expected mass change at
 249 the ice-air interface $S_{m,exp}$ (Eq. (1)) and the latent heat of sublimation ($L_{sub} \approx 2834.1 \cdot$
 250 10^3 J kg^{-1}) the energy needed for sublimation ranged between -2 and -12 J min^{-1} for our
 251 experiments. Our estimated values are a factor up to five lower than the estimated num-
 252 bers of Neumann et al. (2009), because the entering air was already saturated (with ref-
 253 erence to the cold temperature) at the inlet. The needed energy for sublimation could be

254 balanced between the sensible heat carried into and out of the sample, and the exchange
255 of the snow sample with the air stream and the surrounding prevented a temperature
256 drop.

257 Thermal conductivity changed insignificantly in these experiments, especially for
258 ‘ota 1’. This indicates that air warming by a positive temperature gradient along the air-
259 flow and an open system reduces or suppresses the increase in thermal conductivity
260 usually observed by temperature gradient metamorphism (Loewe et al., 2013; Calonne
261 et al., 2014), also the timescales are quite different between experiments. Compared to
262 closed temperature gradient experiment, the applied temperature gradient ~~and the open~~
263 ~~system~~ induced an air movement and therefore reduced the impact on the ~~snow meta-~~
264 ~~morphism and its~~ thermal conductivity, at least on the short term. As a recall, the ther-
265 mal conductivity has been numerically estimated from the geometrical information of
266 the sample only and no air movement was taken into consideration.

267

268 **6. Summary and conclusion**

269 We performed four experiments of temperature-gradient metamorphism of snow
270 under saturated advective airflow during 108 h. Cold saturated air was blown into the
271 snow samples and warmed up while flowing across the sample. The temperature gradi-
272 ent varied between 43 and 53 K m⁻¹ and the snow microstructure was observed by X-ray
273 micro-tomography every 3 h. The micro-CT scans were segmented, and porosity, spe-
274 cific surface area, and the mean pore-size were calculated. Effective thermal conductivi-
275 ty was calculated in direct pore-level simulations (DPLS).

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

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

290 This is the third paper of a series analyzing an advective airflow in a snowpack in
291 depth of more than 1 cm. Previous work showed that: (1) under isothermal conditions,
292 the Kelvin-effect leads to a saturation of the pore space in the snow but did not affect
293 the structural change (Ebner et al., 2015a); (2) applying a negative temperature gradient
294 along the flow direction leads to a change in the microstructure and creation of whisker-
295 like structures due to deposition of water molecules on the ice matrix (Ebner et al.,
296 2015b); and (3) a positive temperature gradient along ~~to~~ the flow had a negligible total
297 mass change of the ice but a strong reposition effect of water molecules on the ice
298 grains, shown in this paper. Conditions (1) and (3) showed that they have a negligible
299 effect on the porosity evolution of the ice matrix **except when sitting is concerned**. Po-
300 rosity changes can be neglected to improve models for snow compaction and evolution
301 at the surface, **however, mechanical processes like compaction strongly impact porosity**.
302 In contrast, conditions (2) showed a significant impact on the structural evolution and
303 seems to be essential for such snowpack models and other numerical simulations. Nev-
304 ertheless, the strong reposition of water molecules on the ice grains observed for all
305 conditions (1) – (3) can have a significant impact on atmospheric chemistry and isotopic
306 changes in snow.

307

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313

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Table 1: Morphological and flow characteristics of the experiments: Volume flow (V), initial superficial velocity in snow ($u_{D,0}$), initial snow density (ρ_0), initial porosity (ε_0), specific surface area (SSA_0), initial mean pore size (d_{mean}), average inlet ($T_{\text{in,ave}}$) and outlet temperature ($T_{\text{out,ave}}$), and the average temperature gradient (∇T_{ave}), corresponding Reynolds number (Re) and Peclet number (Pe).

422

Name	V liter min ⁻¹	$u_{D,0}$ m s ⁻¹	ρ_0 kg m ⁻³	ε_0 —	SSA_0 m ² kg ⁻¹	d_{mean} mm	$T_{\text{in,ave}}$ °C	$T_{\text{out,ave}}$ °C	∇T_{ave} K m ⁻¹	Re —	Pe —
ota1	—	—	284.3	0.69	25.0	0.30	-13.8	-12.5	43.3	—	—
ota2	0.3	0.004	256.8	0.72	26.3	0.33	-14.0	-12.5	50.0	0.07	0.05
ota3	1.0	0.012	256.8	0.72	24.3	0.34	-13.8	-12.3	43.3	0.25	0.19
ota4	3.0	0.036	265.9	0.71	21.7	0.36	-14.6	-13.0	53.3	0.78	0.61

423

424

425

Table 2: Estimated sublimation rate S_m using the mass transfer coefficient h_m determined by Neumann et al. (2009) and the corresponding average surface area per volume $SA_{V,\text{ave}}$. S_m can be compared with the measured sublimation rate of the experiment $S_{m,\text{exp}}$ (Eq. (1)).

430

Name	$SA_{V,\text{ave}}$ mm ⁻¹	h_m m s ⁻¹	S_m kg m ⁻³ s ⁻¹	$S_{m,\text{exp}}$ kg m ⁻³ s ⁻¹
ota1	22.44	$0.75 \cdot 10^{-4}$	$4.83 \cdot 10^{-4}$	$0.68 \cdot 10^{-6}$
ota2	23.98	$1.15 \cdot 10^{-4}$	$2.99 \cdot 10^{-4}$	$4.48 \cdot 10^{-6}$
ota3	21.88	$2.17 \cdot 10^{-4}$	$5.15 \cdot 10^{-4}$	$0.76 \cdot 10^{-6}$
ota4	19.61	$5.16 \cdot 10^{-4}$	$10.9 \cdot 10^{-4}$	$0.08 \cdot 10^{-6}$

431

432 **Figure captions**

433 **Fig. 1.** Schematic of the ice-air interface transport processes: a) Under isothermal
434 conditions Kelvin-effect leads to a saturation of the pore space in the snow
435 but did not affect the structural change (Ebner et al., 2015a); b) Air cooling
436 by a negative temperature gradient along the flow direction leads to a
437 change in the microstructure due to deposition (Ebner et al., 2015b); c) Air
438 warming by a positive temperature gradient along the flow has a negligible
439 total mass change of the ice but a strong reposition effect of water molecules
440 on the ice grains, shown in this paper.

441 **Fig. 2.** Evolution of the 3-D structure of the ice matrix with applied temperature
442 gradient and advective conditions. Experimental conditions (from left to right
443 at different measurement times from beginning to the end (top to bot-
444 tom) of the experiment. The shown cubes are $110 \times 40 \times 110$ voxels ($2 \times$
445 0.7×2 mm³) large with 18 μ m voxel size (a high resolution figure can be
446 found in supplementary material).

447 **Fig. 3.** Superposition of vertical cross-section parallel to the flow direction at time
448 0 and 108 hours for ‘ota3’ (left panel) and ‘ota4’ (right panel). Sublimation
449 and deposition of water vapor on the ice grains were visible with an uncer-
450 tainty of 6 % (a high resolution figure can be found in supplementary mate-
451 rial).

452 **Fig. 4.** Temporal evolution of a) the mean pore size, d_{mean} , of the snow samples ob-
453 tained by an opening-size distribution, b) the porosity, ϵ , obtained by trian-
454 gulated structure surface method, c) the specific surface area, SSA, of the
455 ice matrix obtained by triangulated structure surface method, and d) the ef-
456 fective thermal conductivity of the snow sample, k_{cond} , estimated by DPLS
457 simulations. The sizes of the volumes used for the computation of each
458 property are $350 \times 350 \times 350$ voxels ($6.3 \times 6.3 \times 6.3$ mm³).
459

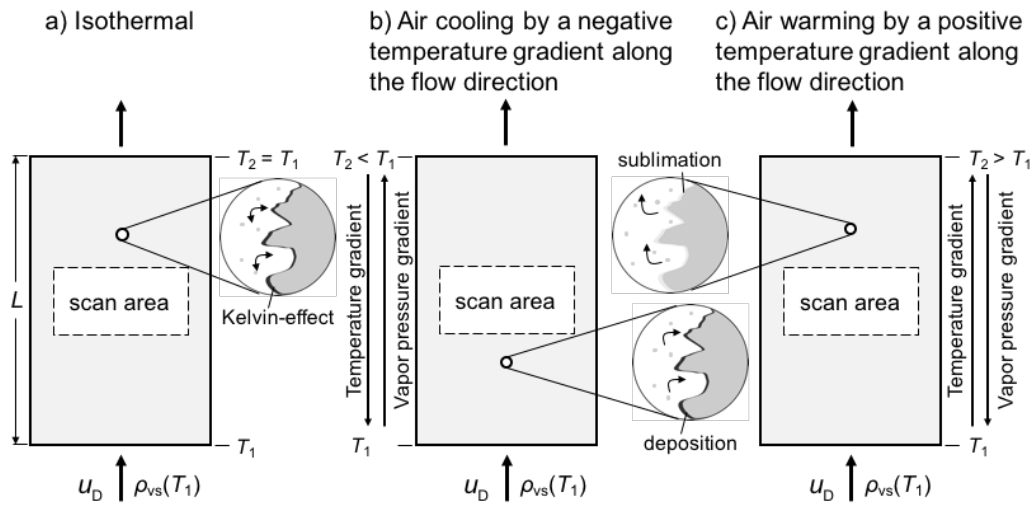


Fig. 1

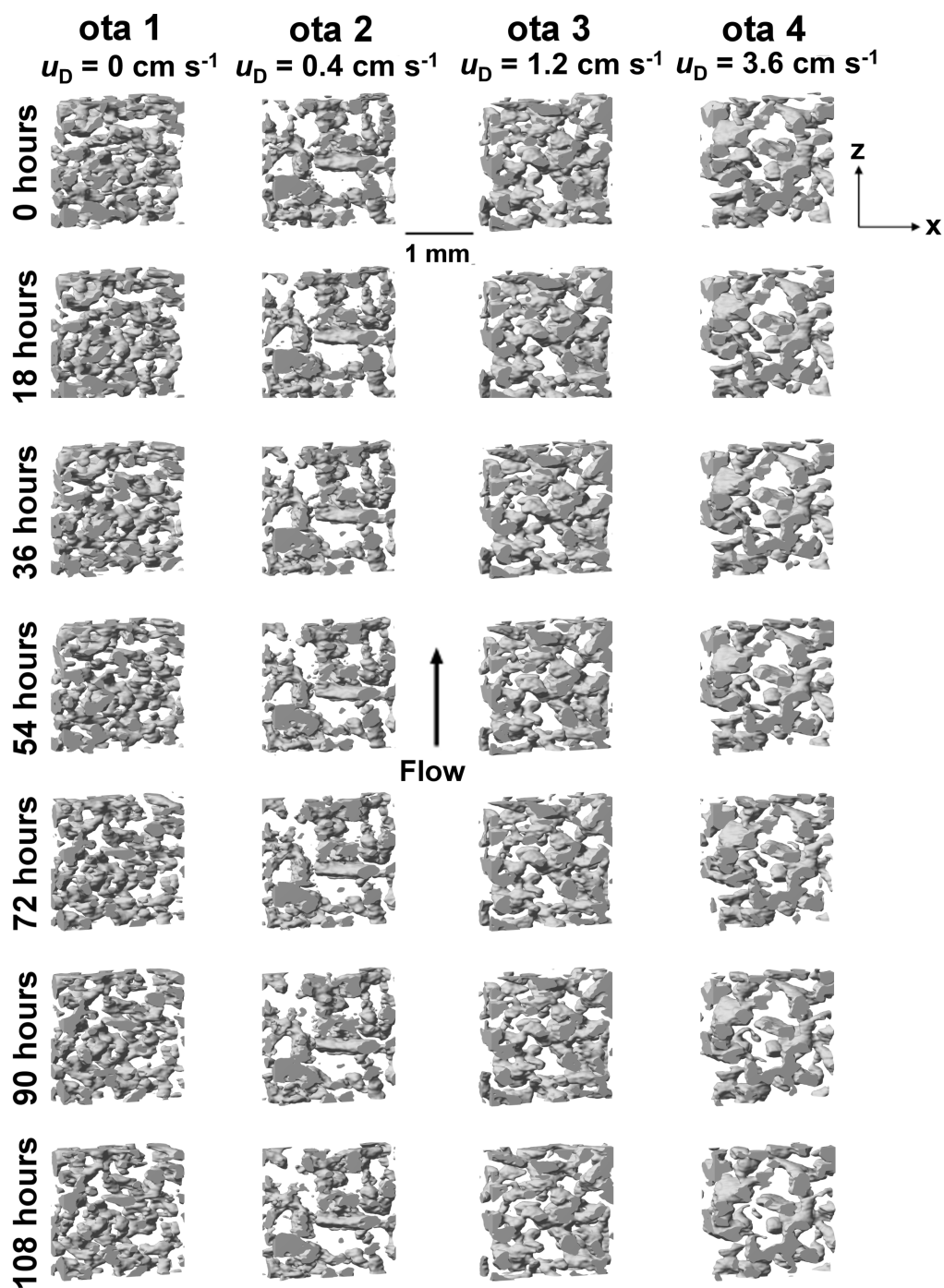
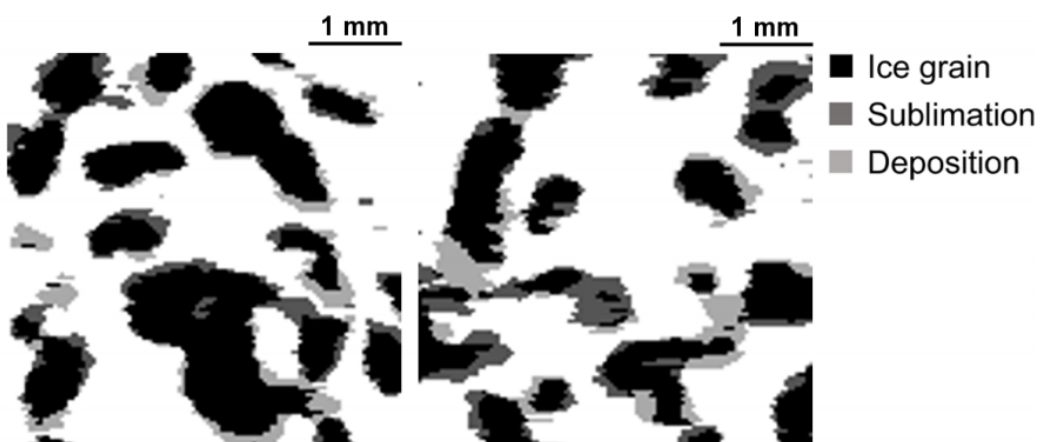


Fig. 2

467



468

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Fig. 3

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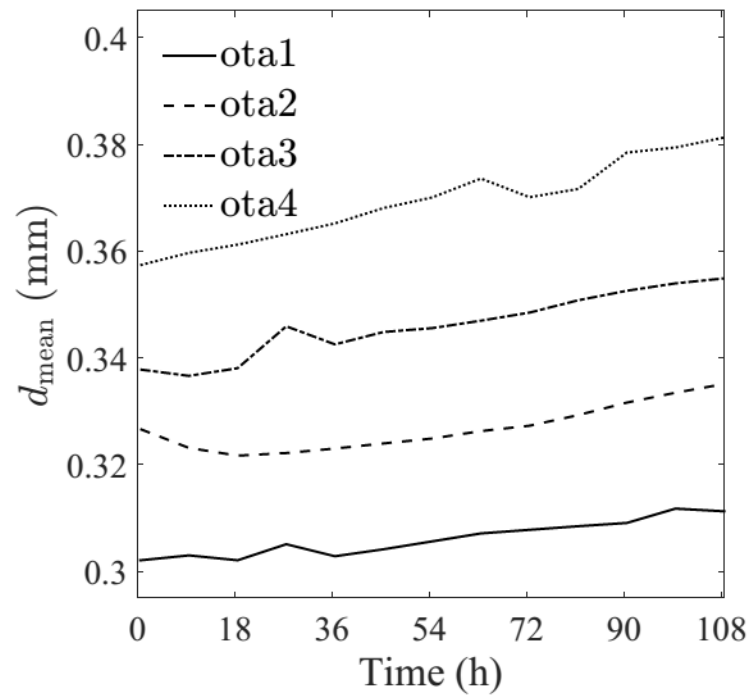


Fig. 4 a)

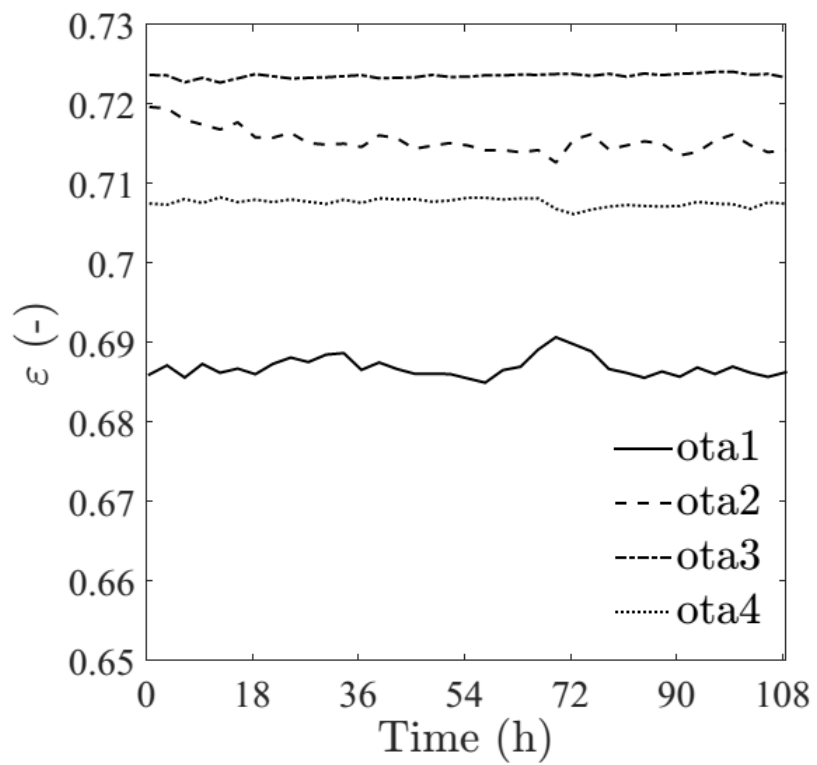


Fig. 4 b)

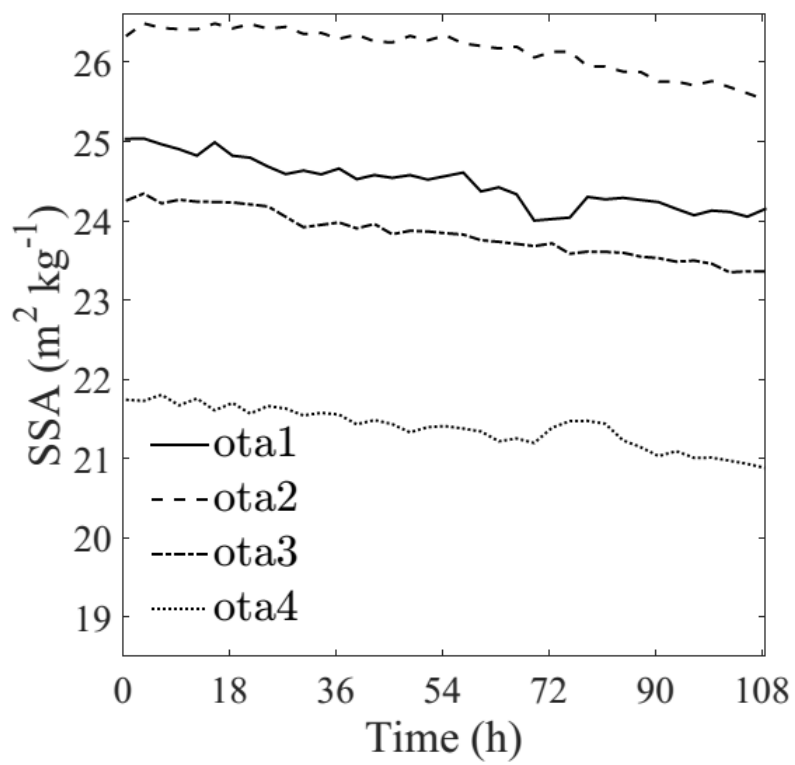


Fig. 4 c)

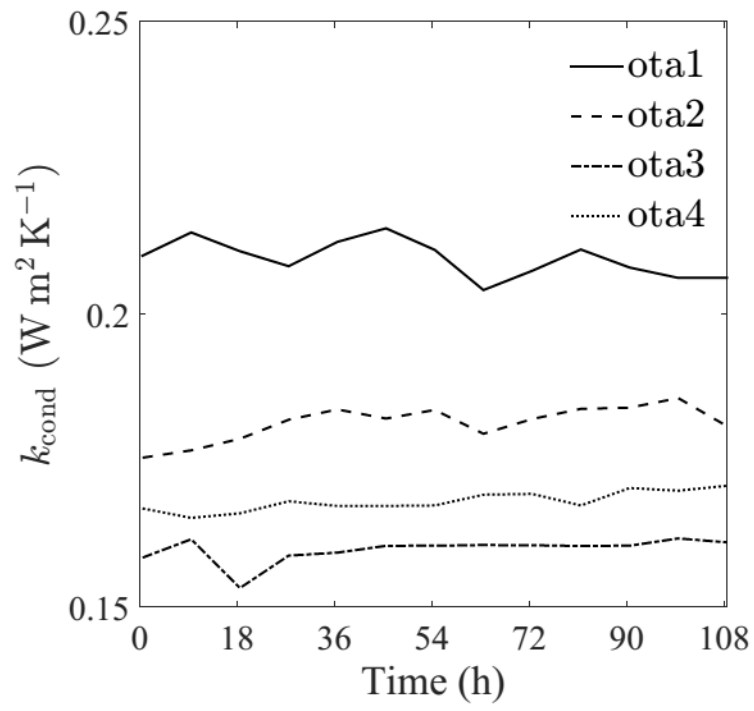


Fig. 4 d)