RESPONSE TO REFEREES' E. A. PODOLSKIY COMMENTS

TO MANUSCRIPT TC-2015-158

Title: Tomography-based observation of sublimation and snow metamorphism under temperature gradient and advective flow

Authors: P. P. Ebner, M. Schneebeli, and A. Steinfeld

We thank the referee E. A. Podolskiy for the constructive comments. All page and line numbers correspond to those of the Discussion Paper.

REVIEWER: E. A. Podolskiy

By employing time-lapse micro-CT approach, the work by Ebner et al. (2015) provides a fresh look onto old and important questions of sublimation and ice reposition within snow, which are indeed crucial for our understanding of snow physics and snow chemistry. Overall, I find the manuscript to be in a good shape, but nevertheless would like to suggest multiple points to consider for improving the readability and clarity of the paper.

Comment #1: p. 4846/14: *Consider giving a broader perspective: "... relevant for atmospheric chemistry." -> "... relevant for atmospheric chemistry {and isotope contents in snow}."*

Revision: Text changed in the revised paper:

> On page 4846, line 13: "However, the strong recrystallization of water molecules in snow may impact its isotopic or chemical content."

Comment #2: p. 4847/1: *In this introduction, I suggest to remind this well familiar effect before giving its name, say: "due to the Kelvin-effect" -> "due to higher vapor pressure over curved surfaces (Kelvin-effect)"*

Revision: Text changed in the revised paper:

> On page 4847, line 1: "… due to higher vapor pressure over convex surfaces and lower vapor pressure over concave surfaces, respectively (Kelvin-effect)."

Comment #3: p. 4847/10: *As non-native English speaker, I cannot correct language. Nevertheless, I note that I never saw "whistler-like crystals" in conventional snow classifications and could not get what was meant by it.*

Response: It is not mention in the conventional snow classifications but was observed in the Paper by Ebner et al. (2015b). However, there is spelling mistake, it is called "whisker-like crystals.

Revision: Text changed in the revised paper:

> On page 4847, line 10: "… whisker-like crystals (Ebner et al., 2015b). Whisker-like crystals are very small (~10-30 μm) elongated monocrystals."

Comment #4: p. 4847/10-11: *Please, reread this sentence, seems to be the opposite? You say the opposite (in p.4850, Lines 18-21).*

"The flow rate dependence on the deposition rate of water vapor on the ice matrix was observed, reaching asymptotically a maximum rate of … (Ebner et al., 2015b)." ->

"The {deposition rate of water vapor on the ice matrix} dependence on the {flow} rate was observed, reaching asymptotically a maximum rate of … (Ebner et al., 2015b)."

Revision: Text changed in the revised paper:

> On page 4847, line 10-11: "A deposition rate of water vapor on the ice matrix dependence on the flow rate was observed, reaching asymptotically a maximum rate of … (Ebner et al., 2015b)."

Comment #5: p. 4847/19: *In regard to ice-core interpretation I also suggest to add a reference to experimental study on isotopic content of snow driven by sublimation.*

Ekaykin, A. A., Hondoh, T., Lipenkov, V. Y. and A. Miyamoto (2009) Post-depositional changes in snow isotope content: preliminary results of laboratory experiments, Clim. Past Discuss., 5, 2239–2267.

Revision: Text changed in the revised paper:

> On page 4847, line 19: "… and thus is important for ice core interpretation (Stichler et al., 2001; Ekaykin et al., 2009)"

Reference added:

> Ekaykin, A. A., Hondoh, T., Lipenkov, V. Y. and A. Miyamoto (2009) Post-depositional changes in snow isotope content: preliminary results of laboratory experiments, Clim. Past Discuss., 5, 2239–2267.

Comment #6: p. 4847/22-23: *About Albert (2002) and his 2D finite-element model ->*

Please note that much more recently Slaughter and Zabaras (2012) took into account such effects on microstructure through a 3D FEM micro-structural model, which considers vapor *deposition and sublimation within the snow. The latter study is indeed very relevant reference here.*

Slaughter AE and Zabaras N (2012), A phase-tracking snow microstructure model. In Proceedings of the International Snow Science Workshop, 16–21 September 2012, Anchorage, Alaska, 395–397 http://arc.lib.montana.edu/snow-science/item/1739

Response: It's correct that this study is a very relevant reference but, based on the Proceeding, this model is not yet finished and no results could be found in the literature. Therefore, it will not make sense to cite this. But we will cite Kaempfer and Plapp (2009)

Revision: Reference added in the revised manuscript

> Kaempfer, T. U. and Plapp, M. (2009) Phase-field modeling of dry snow metamorphism, Physical Review E, 79, http://dx.doi.org/10.1103/PhysRevE.79.031502.

Comment #7: p. 4848/8: *the warm site of the snow -> the warm si{d}e of the snow?*

Revision: Text changed in the revised paper:

> On page 4848, line 8: "… the warm side of the snow …"

Comment #8: p. 4848/10-11: *If available, a range of estimated naturally occurring flow rates would be useful to mention here for indicating how significant the used values were.*

Response: Unfortunately, no directly measured flow rates in a snowpack could be found in the literature, but based on numerical simulation in the PhD thesis of T. Neumann (2003) a maximum flow rate of 0.01 m s^{-1} inside the snow layer (close to the surface) was estimated for a high wind speed (≈10 m s^{-1}).

Comment #9: p. 4848/15-16: *"... and to evaluate the structural change in the earlier stage of metamorphism of new snow." -> I could not follow where in this paper this earlier stage had been discussed? Since it does not seem to be mentioned ever again in this particular manuscript, this phrase could be removed as irrelevant here.*

Revision: Text changed in the revised paper:

> On page 4848, line 15-16: "The snow was sieved with a mesh size of 1.4 mm into a box, and was sintered for 27 days at -5 ºC to increase the strength."

Comment #10: p.4848/19-20: *Please, state the resolution of taken projection X-ray images and reconstructed 3-D scenes.*

Revision: Text changed in the revised paper:

> On page 4848, line 19-20: “The snow samples were measured with a voxel size of 18 μm over 108 h with time-lapse micro-CT measurements taken every 3 h, producing a sequence of 37 images.”

Comment #11: p. 4849/3: *“is analyzed” -> “was analyzed” (for consistency with overall past tense) however, see also below where I suggest to remove this sentence.*

Revision: See Comment #15.

Comment #12: p. 4849/6: *pictures* -> *images*

Revision: Word changed in the revised paper:

Comment #13: p. 4849/7-8: *“was determined by direct pore-level simulations (DPLS) to determine” -> to avoid repetition of the same word, in one place it could be “was estimated by …”*

Revision: Text changed in the revised paper:

> On page 4849, line 7-8: “… was estimated by direct pore-level simulations (DPLS) to determine …”

Comment #14: p. 4849/9: *a reference is made to Löwe et al. (2012), which does not appear in the references.*

Revision: It should mean: Löwe et al. (2013).

Comment #15: p. 4849/2-3 & 6-9: *These two sentences could be easily merged to avoid redundant text and repeating references:*

“As additional physical and structural parameter, the {effective} thermal conductivity{,k_e,} was estimated by direct pore-level simulations (DPLS) to determine the influence of changing microstructure (Kaempfer et al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., 2012).”

Also, for someone who is not familiar with DPSL, an extra sentence introducing the main principle of this computational approach would be informative.

Revision: Text changed in the revised paper:

> On page 4849, line 6-9: “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 corresponding mass and momentum conservation equations (Kaempfer et al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., 2013)."

Comment #16: p. 4849/19: *"were observed for example, Fig …" "were observed{,} for example, Fig …"*

Revision: Text changed in the revised paper:

> On page 4849, line 19: "…were observed, for example, Fig …"

Comment #17: p. 4849/20: *since a reference to test names is made for the first time, it could be smoothed by adding: "for {tests} "ota3" and "ota4".*

Here I also note that Tables 1 and 2 use test names which are different from those mentioned in the main text, Figs. 2 & 4 or a caption of Fig. 3 (e.g., "ta1" vs. "ota1").

Response: The tests names in Tables 1 and 2 are incorrect, it should mean "ota". It is changed in the revised paper.

Revision: Text changed in the revised paper:

> On page 4849, line 20: "Fig. 3 shows the locations of sublimation and deposition for tests "ota3" and "ota4"".

Comment #18: p. 4849/26: *What was an uncertainty for estimated pore size? Similar to the one stated for evaluating reposition?*

Response: It is not relevant to mention the uncertainty for the estimated pore size as the trend is relevant and not the absolute value. But the uncertainty for the estimated pore size is similar to the one stated evaluating reposition.

Revision: Text added in the revised manuscrip

> On page 4849, line 25: "The pore size (uncertainty 6 %) increased …"

Comment #19: p. 4850/1-2: *Should be also mentioned that possible ice loss could not be detected due to limited accuracy (which is almost the same as provided values; p. 4849, Line 21)?*

Revision: Text changed in the revised paper:

On page 4850, line 1-2: “Loss of ice of the snow due to sublimation could not be detected by the micro-CT scans due to limited accuracy and no flow rate …”.

Comment #20: p. 4850/13: *What was the accuracy of estimated thermal conductivity (could be mentioned in DPSL part)?*

Response: It is not relevant to mention the accuracy of the estimated thermal conductivity as the trend is relevant and not the absolute value. However, the standard deviation is only 0.025 W m^{-1}. (Calonne et al., 2011)

Revision: Reference added in the revised manuscript.

Calonne, N., Flin, F., Morin, S., Lesaffre, B., Rolland du Roscoat, S., and Geindreau, C. (2011) Numerical and experimental investigations of the effective thermal conductivity of snow, Geophysical Research Letters, 38, doi:10.1029/2011GL049234.

Comment #21: p. 4850/21: *This phrase is slightly confusing: "In this study, changing flow direction lead …", given that in methods Fig. 1 showed this direction as constant. Was it flipped instead of flipping your heating system? If so, please, check if it is clear in methods. Or if it refers to previously published paper, re-written -> "In the {latter} study, …"*

Revision: Text changed in the revised paper:

On page 4850, line 21: “In this study, changing the temperature gradient leads to a warming up of a cold saturated flow …”

Comment #22: p. 4851/5: *the analyze volume -> the analyzed volume?*

Revision: Word changed in the revised paper:

Comment #23: p. 4851/7: *due to the undersaturated airflow -> In methods, you indicated that the incoming advective flow was initially saturated. Here it is undersaturated. Due to warming during propagation? Perhaps, it could be mentioned here for a sake of clarity.*

Revision: Text changed in the revised paper:

On page 4851, line 7-9: “However, the uptake of water molecules and their transport due to warming during propagation was counteracted by diffusion of water molecules due to the temperature gradient.”

Comment #24: p. 4851/11-12: *Please, explain how mass transfer between diffusion and advection was measured? It remains not very clear from the context.*

Revision: Text changed in the revised paper:

> On page 4851, line 10-14: "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 advection and diffusion, measured during each experiment, showed that diffusion was still dominant (Table 1). Therefore, water molecules were diffused along the temperature gradient and advected along the flow direction leading to a back and forth transport of water molecules."

Comment #25: p. 4851/19-20: *Please, clarify here what causes the increased interaction between air and ice. Residence time? Or air pressure increase due to Bernoulli's principle?*

Revision: Text changed in the revised paper:

> On page 4851, line 19-20: "… and therefore the interaction of an air-parcel with the ice matrix in the pores increases due to higher residence time."

Comment #26: p. 4852/14: *I could not follow how Reynolds number was evaluated in the study, and in Table 1 in particular.*

Revision: Text changed in the revised paper:

> On page 4852, line 14: "Re ($Re = u_D \cdot d_{mean}/\nu$ where ν is the kinematic viscosity of the air) is the corresponding Reynolds-number of the flow."

Comment #27: p. 4852/22: *As the air in the pore spaces are always -> As the air in the pore spaces {is} always*

Revision: Text changed in the revised paper:

> On page 4852, line 22: "As the air in the pore space is always …"

Comment #28: p. 4855/14: *Similarly to Abstract: "... impact on atmospheric chemistry. -> "... impact on atmospheric chemistry {and isotope contents in snow}.*

Revision: Text changed in the revised paper:

> On page 4855, line 14: "… impact on atmospheric chemistry and isotope contents in snow."

Comment #29: Figure 3, caption: *108 h for (left panel) "ota3" and (right panel) "ota4". -> 108 h for "ota3" (left panel) and "ota4" (right panel).*

Revision: Caption changed in the revised paper:

> On page 4863, caption: "Superposition of vertical cross-section parallel to the flow direction at time 0 and 108 h for "ota3" (left panel) and "ota4" (right panel)."

Comment #30: Figure 4, caption: *In the caption a reference to (b) is missing. Also the label of y-axis for (d) uses different symbol from the one used in the main text. Should be consistent (k_e v.s. k_cond).*

Revision: Caption changed in the revised paper:

> On page 4864, caption: "Temporal evolution of (a) the mean pore size, d_{mean}, of the snow samples obtained by opening-size distribution, (b) the porosity, ε, obtained by triangulated structure surface method, (c) the specific surface area, SSA, of the ice matrix obtained by triangulated structure surface method, and (d) the effective thermal conductivity of the snow sample, k_{cond}, estimated by DPLS simulations."

Text changed in the revised manuscript:

> On page 4849, line 3: "… is analyzed by the effective thermal conductivity k_{cond} …"

Minor revisions were made throughout the revised manuscript.

We thank E. A. Podolskiy for his scrutiny and recommendations.

The authors

RESPONSE TO REFEREES' F. FLIN COMMENTS

TO MANUSCRIPT TC-2015-158

Title: Tomography-based observation of sublimation and snow metamorphism under temperature gradient and advective flow

Authors: P. P. Ebner, M. Schneebeli, and A. Steinfeld

We thank the referee F. Flin for the constructive comments. All page and line numbers correspond to those of the Discussion Paper.

We reply to the general questions of the reviewer specifically in the section "Specific comments", as all general questions of the review are given with more details in this section.

<u>REVIEWER: F. Flin</u>

This work deals with the effect of saturated air ($T_{air} = T_d$ = -14°C?) warming up progressively (typically from -14°C to -12.5°C) into snow samples submitted to an imposed temperature gradient (TG) of about 50 K/m. 4 samples were submitted to different flow velocities (from 0 to 3 L min^{-1}) and the evolution of their inner parts (probably 7x7x7 mm3 volumes – this is to be confirmed by the authors) was monitored by X-ray tomography with a pixel size of probably 20 micrometer over 108 h. The evolutions of density, specific surface area (SSA), mean pore diameter and vertical component of thermal effective conductivity were computed from the 3D images obtained. Based on these observations, the authors conclude that the circulation of air, as defined in their settings, impacts the microstructure but not the overall ice mass at the scale of the observed volume. They compare this result to those of a previous paper (Ebner et al, 2015b), where the saturated air is progressively cooled down and propose physical explanations for the differences observed.

This is potentially a good paper that proposes, as the previous papers of Ebner et al, (2015a) and Ebner et al, (2015b) an interesting way to study the impact of air circulation in snow. This time, the case of a saturated air flow progressively warming up in a snow sample is addressed, which consists in one of the possible cases of air advection in snow.

Among several topics, this paper has possible implications for the study of snow subjected to wind (e.g. Seligman 1936; Champollion et al 2013), permeability measurements (e.g. Jordan et al, 1999; Arakawa et al, 2009; Domine et al, 2013) and a better understanding of the matter redistribution mechanisms occurring in TG snow metamorphism (e.g. Flin and Brzoska, 2008; Kaempfer and Plapp, 2009; Pinzer et al, 2012; Calonne et al, 2014; Wang et al, 2014; Krol and Löwe, in press).

This paper needs, however, improvements and clarifications before publication. Here are my main concerns, with some suggestions (see "specific comments" for more details):

1. Missing information and unclear definition of the physical problem:

1.1 The presentation of the physical problem needs to be improved. In particular, the concept of "saturated air", which is extensively used by the authors, is very ambiguous as soon as the snow sample is submitted to a TG. The authors should state more clearly the experimental conditions, and define precisely to which temperature the air is saturated (using the Td notation for the dew point might help). From a strict presentation point of view, such crucial information should appear as soon as possible in the paper.

1.2 Other important information such as voxel size and the size of the region of interest (ROI) used for the computations should be mentioned in the paper.

1.3 Also, no information about the vertical position of the ROI is available. This is, however, an important parameter that should be taken into account in the physical interpretation of the experiment. See e.g. comment 4848/section 2.

2. Problems with the presentation of the physical analysis: Logical links are sometimes difficult to follow and some explanations about the involved physical mechanisms should be improved. See e.g. comments 4845/l6-7 and 4851/21-23.

3. Data post-processing and reliability: The experiment is really interesting, but some "unusual" results obtained need to be checked, clarified or discussed. It is in particular the case of:

- Some erratic translations or changes that can be observed in the image series (see the enlarged version of Fig. 2 in supplementary materials). Has each image that constitutes Fig. 2 been spatially repositioned thanks to adequate references? The beginning of the series ota3 is especially problematic. For most series, some slight but persistent downward translations are observed and should also be commented. See e.g. comment 4862/Fig 2.

- Fig. 3, which is a bit difficult to "read" (no classical rounding or TG effect) and exhibits some post-processing artefacts. See comments 4863/Fig. 3 for suggestions.

- The ota1 series, which does not show any increase of the vertical component of its conductivity when submitted to a TG only. At least a comment should be written on this topic. See comment 4852/27-4853/2.

- Ideally, a characterization of the structural anisotropy of the snow samples would be appropriate. See comment e.g. 4850/7-8.

4. Abstract would benefit from a reformulation: it lacks basic, but important experimental information and do not give a sufficiently clear summary of the physical process occurring in the samples.

5. The title does not describe precisely the experiment and could be improved. Here is a suggestion: "Tomography-based observation of snow metamorphism under a saturated air flow progressively warming up inside the snow sample".

Specific comments:

Comment #1: 4845/Title: *Tomography-based observation of sublimation and snow metamorphism under temperature gradient and advective flow*

1) "temperature gradient and advective conditions": this formulation does not really allow the reader to distinguish the present title from the title of Ebner et al, 2015b.

From the experimental conditions, it appears that snow is always placed in slight but undersaturated air flow, as the incoming air is saturated with respect to the colder temperature of the sample. Mentioning this important fact in the title would be pertinent. At least, from a purely didactic point of view, this would help the reader to understand quickly why sublimation occurs (abstract l. 6).

2) This is a minor point but the wording "sublimation and snow metamorphism" seems unusual as local sublimation is generally assumed (with condensation) as being a part of the metamorphism process itself. Maybe replace the title with: "sublimation during snow metamorphism"?

See also main comment #5.

Response: We reformulated the title

> **“**Metamorphism during temperature gradient with undersaturated advective airflow in a snow sample.**“**

<u>Comment #2:</u> 4846/Abstract: Clear explanations about the direction and intensity of the TG and of the air flow are missing here. Are the air flow velocity and TG collinear? Are they pointing toward the same direction? Which TG and air velocity ranges are concerned? Which part of the sample was observed (top, middle-height, etc.)? These pieces of information are very basic, but mandatory to understand the exact topic of the paper.

Revision: Text added in the revised paper:

> On page 4846, line 6: “Cold saturated air at the inlet was blown into the snow samples and warmed up while flowing across the sample. The temperature gradient in the sample was around 50 K m^{-1} at maximum airflow velocity.”

Text changed in the revised paper:

> On page 4846, line 6: “The sublimation of ice for saturated air flowing across the snow sample was experimentally determined via changes of the porous ice structure in the middle-height of the snow sample.”

<u>Comment #3:</u> 4846/l6-7: *The sublimation of water vapor for saturated air flowing across the snow sample was…*

1) As we are in TG conditions, please define to which temperature the air is saturated (top, middle or base of the snow sample?), i.e., give the dew point of the incoming air.

2) "sublimation of ice" would be preferable to "sublimation of water vapour"

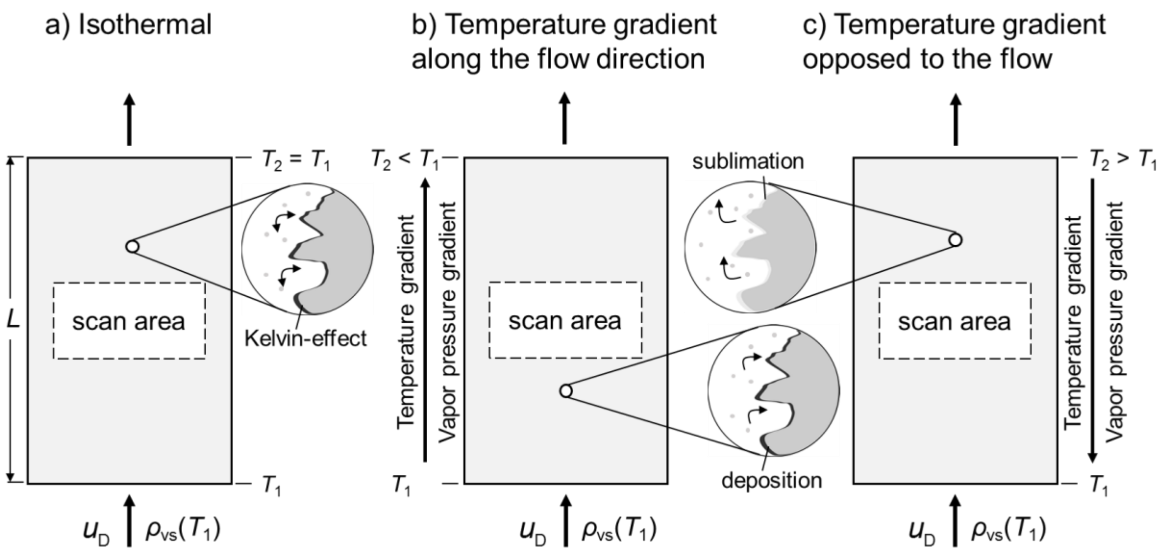
Revision: Text added in the revised paper:

> On page 4845, line 6: “Cold saturated air at the inlet was blown into the snow samples and warmed up while flowing across the sample. The temperature gradient in the sample was around 50 K m^{-1} at maximum airflow velocity.”

Text changed in the revised paper:

> On page 4845, line 6: “The sublimation of ice for saturated air flowing across the snow sample was experimentally determined via changes of the porous ice structure in the middle-height of the snow sample.”

Figure 1 changed in the revised paper:



<u>Comment #4:</u> 4845/8-10: *The results showed that the exothermic gas-to-solid phase change is favorable vis-a-vis the endothermic solid-to-gas phase change, thus leading to more ice deposition than ice sublimation.*

I can hardly understand the meaning of this sentence in the context of the current problem, where all the processes seem to be rather well explained with combined TG and undersaturation effects. Do the authors suspect latent heat effects to take a significant role in the process described? If so, additional explanations should be added to the text. Otherwise, removing this sentence would clarify the conclusion of the paper.

Response: Text removed in the revised paper.

<u>Comment #5:</u> 4846/13-14: *However, the strong reposition process of water molecules on the ice grains is relevant for atmospheric chemistry.*

The sentence seems a bit strange. Why is this process relevant to atmospheric chemistry? No additional element is available in the paper. A possible alternative would be "However, the strong reposition process of water molecules in snow may impact its isotopic or chemical content."

Revision: Text changed in the revised paper:

> On page 4846, line 13: “However, the strong recrystallization of water molecules in snow may impact its isotopic or chemical content.”

<u>Comment #6:</u> 4847/26-28: *In the present work, we studied the surface dynamic of snow metamorphism under an induced temperature gradient and saturated airflow in a controlled laboratory experiments.*

Again, please define more explicitly if the airflow is saturated with respect to the top, the middle or the base of the sample. Giving the typical temperatures of the sample (T_top = -12.5°C and T_base = -14°C) and the temperature and dew point of the incoming air (T_air = - 14°C and Td_air = -14°C?) as soon as possible in the paper would really help the reader.

Revision: Text added in the revised paper:

> On page 4847, line 28: "Cold saturated air at around -14 ºC was blown into the snow samples and warmed up to around -12.5 ºC while flowing across the sample."

<u>Comment #7:</u> 4848/2: What is the meaning of "discrete-scale geometry"? Do you just mean "discrete geometry" (or "digitized geometry") or something more specific?

Response: We use the term "discrete-scale geometry" in contrast to "continuum-scale geometry".

Revision: Text added in the revised paper:

> On page 4848, line 2: "By using discrete-scale geometry, all structures are resolved with a finite resolution corresponding to the voxel size."

<u>Comment #8:</u> 4848/section 2: no information about the voxel size of the images is clearly available. It is however a very crucial point when considering the changes occurring in a metamorphosing snow microstructure (especially for a rather short time period of 108 h at about -14°C).

Important information such as the selected ROI in the sample (size and position inside the sample) needs also to be added. As the authors know, the vertical position of the ROI is especially important as soon as TG metamorphism is concerned. For example, if a snow sample is submitted to vertical gradient (Tbase=-12.5°C, Ttop=-14°C) with no air flow, the base is known to undergo strong sublimation while the top undergoes condensation: people observing only the upper part of the sample would conclude into an ice mass increase, while people looking at the base part would reach exactly opposite conclusions. The central part is however known to be constant in density (Schneebeli and Sokratov, 2004; Srivastava et al, 2009; Calonne et al 2014), which is consistent with what is observed in this experiment. It should however be noticed that additional air flow makes the problem much more complex, exhibiting supplementary reasons for vertical variations depending on air flow velocity (see e.g. Fig. 5 of Calonne et al, 2015).

Revision: Text changed in the revised paper:

> On page 4848, line 19-20: "The snow samples were analyzed 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 μm^3".

Text added in the revised paper:

> On page 4848, line 20: "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 center of the sample (Fig. 1)"

Comment #9: 4848/11-13: *Natural identical snow produced in a cold laboratory [Schleef et al., 2014] was used for the snow sample preparation (water temperature: 30 ºC; air temperature: −20 ºC).*

Such information on water and air temperatures seems particularly system-dependent. Would it be possible for the authors to indicate temperature and humidity (Td?) in the nucleation chamber? Are there specific reasons for the choice of snow type C1f/G6 ("hollow columns with germs" as described by Schleef et al, 2014)?

Response: We agree that this information is system dependent. Schleef et al. (2014) estimated the corresponding temperature and humidity in the chamber. As the snow was isothermally stored during more than 27 days before use (-5 °C), the original snow type was not any more recognizable in the used samples

Comment #10: 4848/25-27: *The segmented data were used to calculate a triangulated ice matrix surface and tetrahedrons inscribed into the ice structure. Morphological parameters such as porosity (epsilon) and specific surface area (SSA) were then calculated.*

As stressed in preceding reviews, several mesh methods are known to provide biased estimations of SSA (see e.g. Flin et al, 2011).

Response: We agree that the calculation of the SSA is not trivial. In our case, we used the algorithms of Haussener et al (2012), which are validated (Zermatten et al., 2014). In addition, we use here mainly the ratio between SSA/SSA_0 to investigate the trend and the evolution of this parameter to see the influence of advective airflow and metamorphism.

Comment #11: 4849/1: *Opening size distribution was applied…* -> "An opening-based morphological operation was applied…"

Revision: Text changed in the revised paper:

<u>Comment #12:</u> 4849/18: *The morphological evolution was similar between all four experiments and only a slight rounding and coarsening was visually observed, shown in Fig. 2, indicating that the initial ice grain did not change with time. Only coarsening processes of the ice grain were observed for example, Fig. 3 shows the locations of sublimation and deposition for "ota3" and "ota4".*

These sentences are not very clear and some assertions seem to contradict each other.

Revision: Text added in the revised paper:

> On page 4849, line 18: "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 grain did not change with time and the locations of sublimation and deposition for "ota3" and "ota4" is shown in Fig. 3."

<u>Comment #13:</u> 4850/3: *the temporal porosity distribution* -> "the temporal evolution of the porosity"

Revision: Text changed in the revised paper:

<u>Comment #14:</u> 4850/7-8: *A coarsening was observed for each experiment but the influence of changing airflow was not visible, confirmed by the temporal SSA evolution, shown in Fig. 4c.*

Have the authors tried to quantify snow structural anisotropy? From detailed observation of the final images of Fig. 2 (see supplementary material), it seems the higher the velocity, the less horizontally-layered the structure.

Response: Although a change in the anisotropy was observed in previous TG experiments we didn't quantify the snow structural anisotropy in our observations.

<u>Comment #15:</u> 4850/9-11: *Although the repositioning of water molecules led to a smoothing of the ice grains, it did not affect the heat transfer in the snow. The thermal conductivity slightly increased after applying airflow to the temperature gradient…*

Heat transfer is actually made of different contributions (conduction, convection, radiation, latent heat…) but the authors limited their computations to the determination of the vertical component of the effective conductivity from the obtained tomographic images. This formulation might be more adequate: "The repositioning of water molecules led to a smoothing of the ice grains, but did not affect the conductivity of snow. This quantity slightly increased after applying airflow to the temperature gradient."

Revision: Text changed in the revised paper:

> On page 4850, line 9-11: “The repositioning of water molecules led to a smoothing of the ice grains, but did not affect the thermal conductivity of snow. This quantity slightly increased after applying airflow to the temperature gradient ...”

Comment #16: 4850/16-18: *The kinetic phase-change from gas to solid is preferable over solid to gas as energy is released rather than consumed leading to more ice deposition rather than ice sublimation.*

Like in the abstract, this sentence sounds very strange to me, and is neither supported by what follows in the discussion nor by the reported experiment. See also lines 4854/21/22 in the text.

Response: Text removed in the revised paper.

Comment #17: 4851/3-4: *The superposition of vertical cross-section in Fig. 3 shows a big effect on reposition of water molecules on the ice structure.*

Due to the acquisition process (slight variability of the X-ray source leading to small differences in the reconstruction parameters, e.g.), the 3D images can generally undergo tiny translations and rotations with time. Has each image that constitutes Fig. 3 been spatially repositioned thanks to adequate references? How was it done? See also comment 4862/Fig. 2.

Response: Yes, we repositioned the images to adequately reference. We used a linear encoder with a resolution of less than 1 voxel to verify that the scans were taken at the same position. We added a sentence to the description of the methods.

Revision: Text added in the revised paper:

> On page 4848, line 20: “A linear encoder with a resolution of less than 1 voxel was used to verify that the scans were taken at the same position.”

Comment #18: 4851/5-6: *Continued sublimation and deposition of water molecules due the Kelvin-effect led to a saturation of the pore space.*

Does this sentence concern the snow samples before starting the experiment? In the described experiment, we have:

1) a significant temperature gradient;

2) an air advection effect, which brings cold and dry air onto slightly warmer ice surfaces; Is really Kelvin effect (also known as curvature effect) occurring in this case? What about Clausius-Clapeyron equation? Some physical explanations of the whole paragraph are rather difficult to follow and should be clarified.

Response: Yes, this sentence concern the snow samples before starting the experiments.

Revision: Text added in the revised paper:

> On page 4851, line 5-6: “The vapor pressure of the air in the pore was in equilibrium with the water pressure of the ice, given by the local temperature.”

Comment #19: 4851/15-18: *...sublimation inside a snowpack has a significant influence not on the total net mass change but on the structural orientation of the ice grains due to redistribution of water vapor on the ice matrix*.

1) What does "sublimation inside a snowpack" actually mean? It seems it would be better to replace this wording by "advection of cold saturated air into a slightly warmer snowpack".

2) It is also important to notice that the experiment presented in the paper does not allow drawing any conclusion on potential "skin effects", i.e. on what happens near the interfaces (snow-air interface, interface between 2 distinct layers in the snowpack…). However, "skin effects" are particularly important as far as snowpack, TG and undersaturated flow are considered.

3) Also, what is actually meant by "structural orientation"? I there a way to quantify this impact? See also comment 4850/7-8. 4851/21-23: *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 crystals faces.*

I could not understand the logical process by which the author reached this conclusion. Would it be possible to improve the related explanations?

Response: Optically we can see a change of the structural orientation. However, as we didn’t calculate the anisotropy we will reformulate the sentence.

Revision: Text changed in the revised paper:

> On page 4851, line 15-18: “…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 of water vapor on the ice matrix."

Comment #20: 4853/27-4853/2: *Thermal conductivity changed insignificantly in these experiments of short duration. This indicates that advective cold airflow opposite to a temperature gradient reduces or suppresses the increase in thermal conductivity usually observed by temperature gradient metamorphism (Riche and Schneebeli, 2013).*

It should be noticed that this stable conductivity is also true for experiment ota1, which is a pure TG experiment as it occurs without any air advection: why an increase in the vertical component of the thermal conductivity is not observed in this particular case? Are the results sufficiently reproducible (difficulty of precise temperature control on a small snow sample, problem of representative elementary volume for the computation of thermal conductivity, dependence of the morphology with the vertical position of the investigated sample, etc.) to draw conclusions on this topic? Are there other reasons to explain the conductivity evolution of ota1?

Response: An increase of the vertical component of the thermal conductivity is not observed for 'ot1' because one reason could be that we had an open TG experiment system. Compared to closed TG experiment, the temperature gradient induce an air movement and therefore has an additional influence on the thermal conductivity.

Revision: Text changed in the revised paper:

> On page 4853, line 27 – page 4853, line 2: "Thermal conductivity changed insignificantly in these experiments. This indicates that advective cold airflow opposite to a temperature gradient and an open system reduces or suppresses the increase in thermal conductivity usually observed by temperature gradient metamorphism (Riche and Schneebeli, 2013)."

Comment #21: 4855/8-10: *Conditions (1) and (3) showed that they have a negligible effect on the structural changes of the ice matrix and can be neglected to improve models for firn compaction and evolution.*

For conditions (3), this sentence seems in contradiction with some parts of the paper, where the morphological changes are considered as significant in 108 h of experiment (see e.g Fig. 3 and 4851/3-4).

Response: Correct, but looking at the temporal porosity evolution there is no change observable and therefore it can be neglected to improve models for snow compaction and evolution at the surface.

Revision: Text changed in the revised paper:

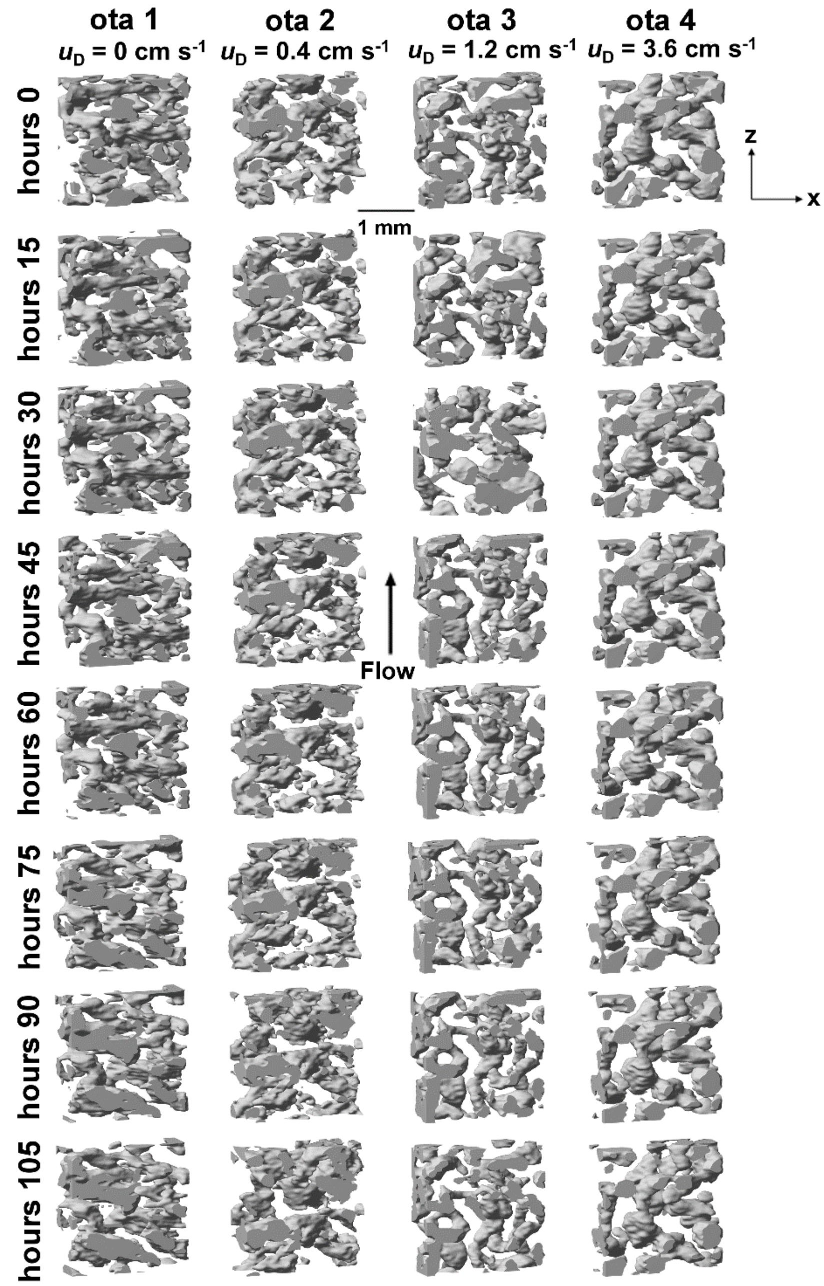
> On page 4855, line 8-10: "Conditions (1) and (3) showed that they have a negligible effect on the porosity evolution of the ice matrix and can be neglected to improve models for snow compaction and evolution at the surface."

Comment #22: 4862/Fig. 2: Please add flow velocities in the figure and voxel size in the caption. From the enlarged view (supplement file) it can be noticed that microstructures show erratic translations with time: it is the case for ota1 (between 0 and 15 h) and ota2 (between 75 and 90 h) where fast downward translations occur. Is it an artefact of the image acquisition process or is it linked to a physical process (snow settling under its own weight (Schleef et al, 2014a), which is generally considered as unlikely under significant TG, sublimation of underlying snow, etc.)? Artefacts should be corrected and physical processes explicitly mentioned in the article's text.

For ota3, the series show clearly erratic vertical translations between 0 and 30 h, with a completely different structure at 30 h, before returning to something more like the original image after 45 h. Could these positioning problems be related to the strong variations observed in conductivity (Fig. 4 – ota3) between 0 and 30 h? Please check.

See also comment of 4850/7-8.

Revision: Figure 2 and caption changed:



Caption Figure 2:

> "Evolution of the 3-D structure of the ice matrix with applied temperature gradient and advective conditions. Experimental conditions (from left to right) at different measurements times from beginning to the end (top to bottom) of the experiment. The shown cubes are 110 × 40 × 110 voxels (2 mm × 0.7 mm × 2 mm) large with 18 µm voxel size (a high resolution figure can be found in Supplement)."

Text added in the revised paper:

> On page 4849, line 19: "The change of structural change "ota 3" at 30 h is due to an error in the scan."

Comment #23: 4863/Fig. 3: is a bit deceiving: in particular, it exhibits a voxel-size horizontal layering, which is typically obtained when image processing algorithms (median and Gaussian filters, threshold…) are only applied in 2D on horizontal cross-sections without considering the vertical direction. I suggest really using a 3D threshold method to improve the quality and reliability of the images. If necessary, some indications and references can be found in e.g., Hagenmuller et al, 2013 (p. 862-863).

It seems also difficult to deduce a general trend for sublimation and deposition sites. Maybe a 3D view with a color code proportional to the measured interface speed or an adequate graph (see e.g. Krol and Löwe, in press) would help.

Response: For the µ-CT80 (Scanco Medical) it would not make sense to use a 3D threshold method. The device scans the sample stack by stack having a slightly difference in the intensity for each stack. Therefore, we decided to segment each 2D section separately using the Otsu method.

A measured interface speed graph is out of the scope for this paper as we are not interested in the interface speed and if the editor agrees, we suggest not to include such graphs.

Comment #24: 4864/Fig. 4: please specify (caption and/or text) the sizes of the samples on which these properties have been obtained. Are volumes representative for the considered properties?

Revision: Text added in the revised paper:

> On page 4848, line 20: "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."

Technical comments:

Comment #1: 4847/28: *experiments* -> "experiment"

Revision: Text changed in the revised paper:

Comment #2: 4847/10 *whistler-like crystals* -> "whisker-like crystals"

Revision: Text changed in the revised paper:

<u>Comment #3:</u> 4848/6: *in a cold laboratory temperature* -> “at a cold laboratory temperature”

Revision: Text changed in the revised paper:

<u>Comment #4:</u> 4848/11: *Natural identical snow* -> “Nature identical snow”

Revision: Text changed in the revised paper:

<u>Comment #5:</u> 4849/18: *initial ice grain* -> initial ice grains

Revision: Text changed in the revised paper:

<u>Comment #6:</u> 4849/19: *the ice grain* -> the ice grains

Revision: Text changed in the revised paper:

<u>Comment #7:</u> 4851/5: *analyze volume* -> analyzed volume

Revision: Text changed in the revised paper:

<u>Comment #8:</u> 4855/11: *seem* -> “seems”

Revision: Text changed in the revised paper:

<u>Comment #9:</u> 4864/Fig. 4: A "(b)" is missing in the caption.

Revision: Text insert in the revised paper:

Minor revisions were made throughout the revised manuscript.

We thank F. Flin for his scrutiny and recommendations.

The authors

~~Tomography-based observation of sublimation and snow metamorphism under temperature gradient and advective flow~~

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. The temperature gradient in the sample was around 50 K m^{-1} at maximum airflow velocity. The sublimation of ~~water vapor~~ ice for saturated air flowing across the snow sample was experimentally determined via ~~variations of the porous ice structure~~ changes of the porous ice structure in the middle-height of the snow sample. ~~The results showed that the exothermic gas-to-solid phase change is favorable vis-a-vis the endothermic solid-to-gas phase change, thus leading to more ice deposition than ice sublimation.~~ Sublimation has a marked effect on the structural change of the ice matrix but diffusion of water vapor in 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. ~~reposition process of water molecules on the ice grains is relevant for atmospheric chemistry.~~

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

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1. Introduction

Snow has a complex porous microstructure and consists of a continuous ice structure made of grains connected by bonds and inter-connecting pores (Löwe et al., 2011). It has a high permeability (Calonne et al, 2012) and under appropriate conditions airflow through the snow structure can occur (Sturm and Johnson, 1991) due to variation of surface pressure (Colbeck, 1989; Albert and Hardy, 1995), simultaneous warming and cooling, and induced temperature gradients (Sturm and Johnson, 1991). Both diffusive and advective airflows affect heat and mass transport in the snowpack and influence chemical concentrations (Gjessing, 1977; Waddington et al., 1996). Various airflow conditions in a snow sample occur, namely: isothermal airflow, temperature gradient along the flow direction, and temperature gradient opposite to the airflow (Fig. 1). Under isothermal condition, the continuous sublimation and deposition of ice due to higher vapor pressure over convex surfaces and lower vapor pressure over concave surfaces, respectively (Kelvin-effect).~~to the Kelvin-effect leads to a saturation of the pore space in the snow~~ (Neumann et al., 2008; Ebner et al., 2014). However, applying a fully isothermal saturated airflow across a snow sample has been shown to have no influence on the coarsening rate that is typical for isothermal snow metamorphism independently of the transport regime in the pores (Ebner et al., 2015a). When applying a temperature gradient, the effect of sublimation and deposition in the snow results from interaction between snow temperature and the local relative humidity in the pores. If vapor is advected from a warmer zone into a colder zone, the air becomes supersaturated, and some water vapor deposits onto the surrounding ice grains. This leads to a change in the microstructure creating whisker-like crystals (Ebner et al., 2015b). Whisker-like crystals are very small (~10-30 µm) elongated monocrystals. ~~whistler-like crystals (Ebner et al., 2015b) The flow rate dependence on the deposition rate of water vapor on the ice matrix was observed,~~ A deposition rate of water vapor on the ice matrix dependence on the flow rate was observed, reaching asymptotically a maximum rate of $1.05 \cdot 10^{-4}$ kg m^{-3} s^{-1} (Ebner et al., 2015b). Contrarily, if the temperature gradient acts in the opposite 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). Albert (2002) suggest that condensation of water vapor will have a noticeable effect on the microstructure of snow using airflow velocities, vapor transport and sublimation rates calculated using a two-dimensional finite-element model, which is also confirmed by a 3D phase-field model of Kaempfer and Plapp (2009).. 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 crystal surface.

In the present work, we studied the surface dynamic of snow metamorphism under an induced temperature gradient and saturated airflow in a controlled laboratory experiments. Cold saturated air at around -14 ºC was blown into the snow samples and warmed up to around -12.5 ºC while flowing across the sample. Sublimation of ice was analyzed by in-situ time-lapse ~~experiments~~ experiment with microcomputer tomography (micro-CT) (Fig. 1 c)) (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~~ at 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 generate the warm side of the snow, opposite to the entering airflow. 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). ~~Natural~~ Nature identical snow produced in a cold laboratory (Schleef et al., 2014) was used for the snow sample preparation (water temperature: 30 ºC; air temperature: −20 ºC). 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 the strength. ~~and to evaluate the structural change in the earlier stage of metamorphism of new snow.~~ 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 μm ~~analyzed~~ 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 μm^3. 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 center of the sample (Fig. 1 c)). A linear encoder with a resolution of less than 1 voxel 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 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 of the two-phase system were determined based on the exact 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. ~~Opening size distribution was applied~~ An opening-based morphological operation was applied to extract the mean pore size of each micro-CT scan (d_{mean}) (Haussener et al., 2012). ~~The influence of structural changes on the heat transfer in the snow is analyzed by the effective conductivity k_e (Kaempfer et al., 2005; Petrasch et al., 2008; Calonne et al., 2011).~~ Morphological parameters such as porosity, specific surface area and the initial mean pore size were extracted from the micro-CT ~~pictures~~ images to study the ice-air interface dynamic. As additional physical and structural parameter, the effective thermal conductivity k_{cond} was ~~determined~~ estimated by direct pore-level simulations (DPLS) to determine the influence of changing microstructure. DPLS determined the effective thermal conductivity by solving the corresponding mass and momentum conservation equations (Kaempfer et al., 2005; Petrasch et al., 2008; Calonne et al., 2011; Löwe et al., ~~2012~~ 2013).

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 Δt < 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 change of structural change "ota 3" at 30 h is due to an error in the scan.~~ indicating that the initial ice grain did not change with time. Only coarsening processes of the ice grain were observed, for example, Figure 3 shows the locations of sublimation and deposition for tests 'ota3' and 'ota4'.~~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 grain with low radii and was relocated leading to a smoothing of the ice grain 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 of the snow 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 porosity distribution~~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. A 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).

~~Although the repositioning of water molecules led to a smoothing of the ice grains, it did not affect the heat transfer in the snow. The thermal conductivity~~ 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} (Calonne et al., 2011)) slightly increased after applying airflow to the temperature gradient ~~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 kinetic phase-change from gas to solid is preferable over solid to gas as energy is released rather than consumed leading to more ice deposition rather than ice sublimation. 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 flow direction 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 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 Kelvin-effect led to a saturation of the pore space. The vapor pressure of the air in the pore was in equilibrium with the water pressure of the ice, given by the local temperature. However, the uptake of water molecules and their transport due to warming during propagation was counteracted by diffusion of water molecules due to the temperature gradient. to the undersaturated airflow 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 cancelled each other out. 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 temperature gradient and advected along the flow direction leading to a back and forth transport of water molecules., thus indicating that water molecules were transported back and forth due to diffusive and advective transport.

As a Peclet higher than 1 is not possible in nature (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 of water vapor on the ice matrix. ~~sublimation inside a snowpack has a significant influence not on the total net mass change but on the structural orientation of the ice grains due to redistribution of water vapor 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 crystals faces. This is also 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

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

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

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

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)

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

assuming that the sublimation occurs within the first few mm of the sample. Re (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 modelled values and also two orders of magnitude lower than for the temperature gradient along an airflow experiment (Ebner et al., 2015b). As the air in the pore spaces is always saturated (Neumann et al., 2009), the back diffusion of water vapor in the 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 the 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 was not analyzed.

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 began 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. This indicates that advective cold airflow opposite to a temperature gradient reduces or suppresses the increase in thermal conductivity usually observed by temperature gradient metamorphism~~

~~(Riche and Schneebeli, 2013).~~ Thermal conductivity changed insignificantly in these experiments. This indicates that advective cold airflow opposite to a temperature gradient and an open system reduces or suppresses the increase in thermal conductivity usually observed by temperature gradient metamorphism (Riche and Schneebeli, 2013).

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 microtomography 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 crystal with low 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 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 kinetic phase-change from gas to solid is preferable as energy is released compared to solid to gas where energy is required, thus leading to more water molecule deposition than water molecule sublimation. 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. Previous work showed that: (1) under isothermal conditions 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 temperature gradient along the flow direction leads to a change in the microstructure and creation of whistler-like structures due to deposition of water molecules on the ice matrix (Ebner et al., 2015b); and (3) a temperature gradient opposed 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 and can be neglected to improve models for snow compaction and evolution at the surface. ~~on the structural changes of the ice matrix and can be neglected to improve models for firn compaction and evolution~~. 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 isotope contents in snow.

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Table 1: Morphological and flow characteristics of the experiments: Volume flow ($\dot{V}$), 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 ($T_{in,ave}$) and outlet temperature ($T_{out,ave}$), and the average temperature gradient (∇T_{ave}), corresponding Reynolds number (Re) and Peclet number (Pe).

Name	$\dot{V}$ 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	$T_{in,ave}$ °C	$T_{out,ave}$ °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

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 $SA_{V,ave}$. S_m can be compared with the measured sublimation rate of the experiment $S_{m,exp}$ (Eq. (1)).

Name	$SA_{V,ave}$ mm^{-1}	h_m m s^{-1}	S_m kg m^{-3} s^{-1}	$S_{m,exp}$ 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	$5.16 \cdot 10^{-4}$	$10.9 \cdot 10^{-4}$	$0.08 \cdot 10^{-6}$

Figure captions

Fig. 1. Schematic of the ice-air interface transport processes: a) Under isothermal conditions Kelvin-effect leads to a saturation of the pore space in the snow but did not affect the structural change (Ebner et al., 2015a); b) Temperature gradient along the flow direction leads to a change in the microstructure due to deposition (Ebner et al., 2015b); c) Temperature gradient opposite to the flow has a negligible total mass change of the ice but a strong reposition effect of water molecules on the ice grains, shown in this paper.

Fig. 2. Evolution of the 3-D structure of the ice matrix with applied temperature gradient and advective conditions. Experimental conditions (from left to right) at different measurement times from beginning to the end (top to bottom) of the experiment. The shown cubes are 110 × 40 × 110 voxels (2 × 0.7 × 2 mm^3) large with 18 µm voxel size (a high resolution figure can be found in supplementary material).

Fig. 3. Superposition of vertical cross-section parallel to the flow direction at time 0 and 108 hours for 'ota3' (left panel) and 'ota4' (right panel). Sublimation and deposition of water vapor on the ice grain were visible with an uncertainty of 6 % (a high resolution figure can be found in supplementary material).

Fig. 4. Temporal evolution of a) the mean pore size, d_{mean}, of the snow samples obtained by opening-size distribution, b) the porosity, ε, obtained by triangulated structure surface method, c) the specific surface area, SSA, of the ice matrix obtained by triangulated structure surface method, and d) the effective thermal conductivity of the snow sample, k_{cond}, estimated by DPLS simulations.

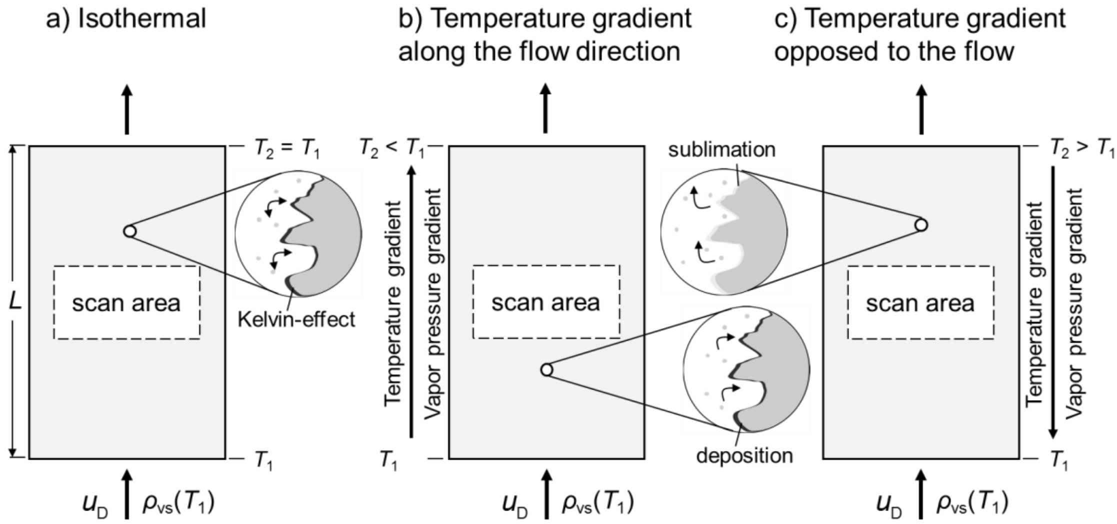

a) Isothermal
b) Temperature gradient along the flow direction
c) Temperature gradient opposed to the flow
$T_2 = T_1$
$T_2 < T_1$
$T_2 > T_1$
T_1
L
scan area
Kelvin-effect
sublimation
deposition
Temperature gradient
Vapor pressure gradient
u_D
$\rho_{vs}(T_1)$

Fig. 1

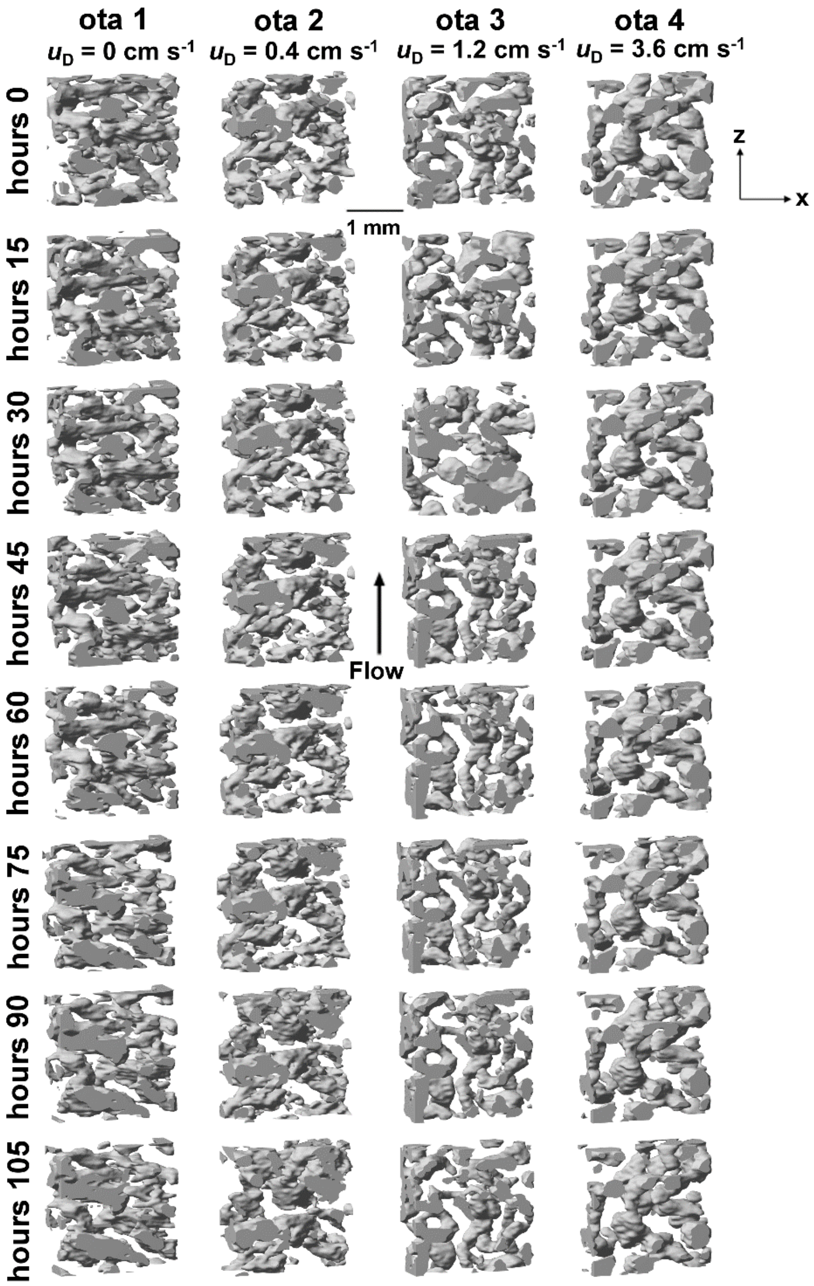

ota 1
uD = 0 cm s-1
ota 2
uD = 0.4 cm s-1
ota 3
uD = 1.2 cm s-1
ota 4
uD = 3.6 cm s-1
hours 0
hours 15
hours 30
hours 45
hours 60
hours 75
hours 90
hours 105
1 mm
Flow
z
x

Fig. 2

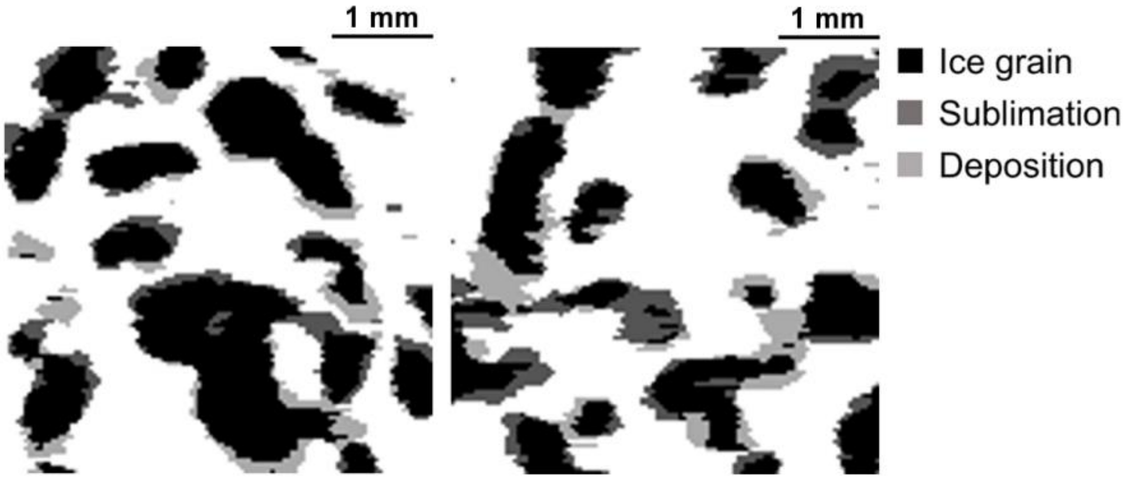


Fig. 3

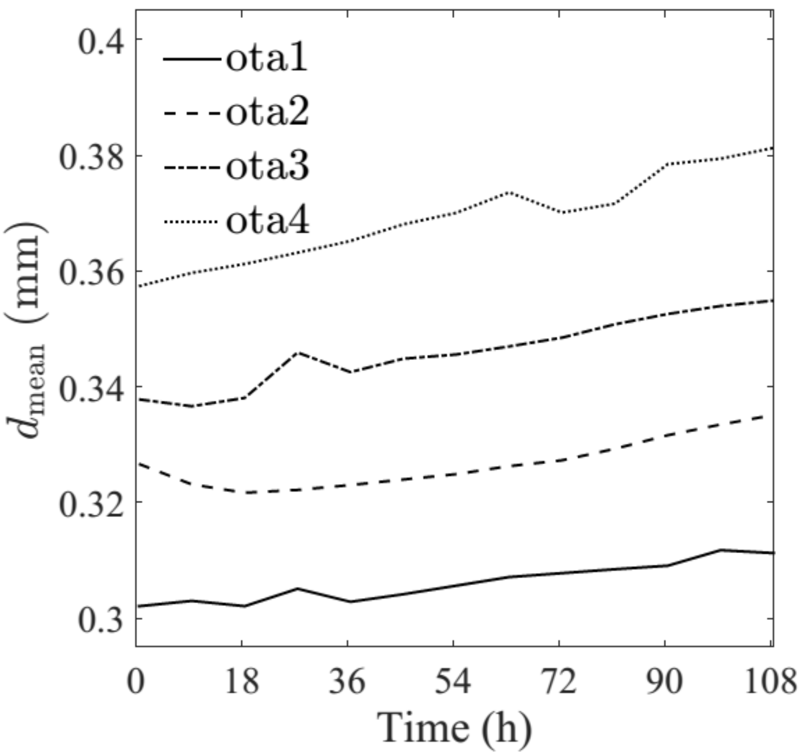

ota1
ota2
ota3
ota4
d_{mean} (mm)
0.4
0.38
0.36
0.34
0.32
0.3
0
18
36
54
72
90
108
Time (h)

Fig. 4 a)

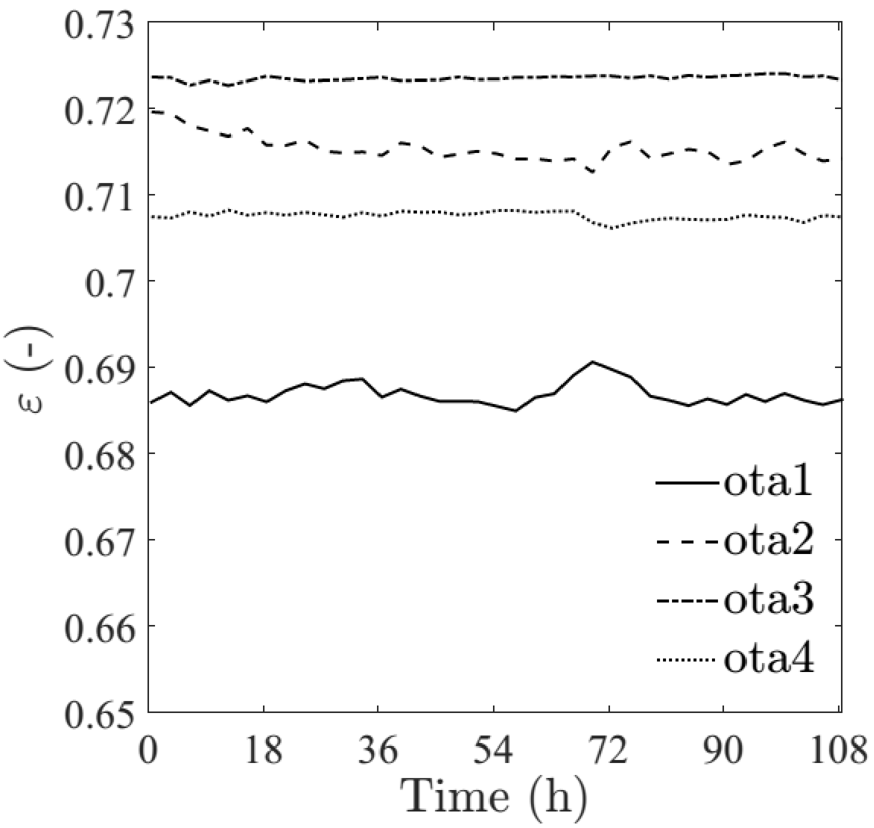
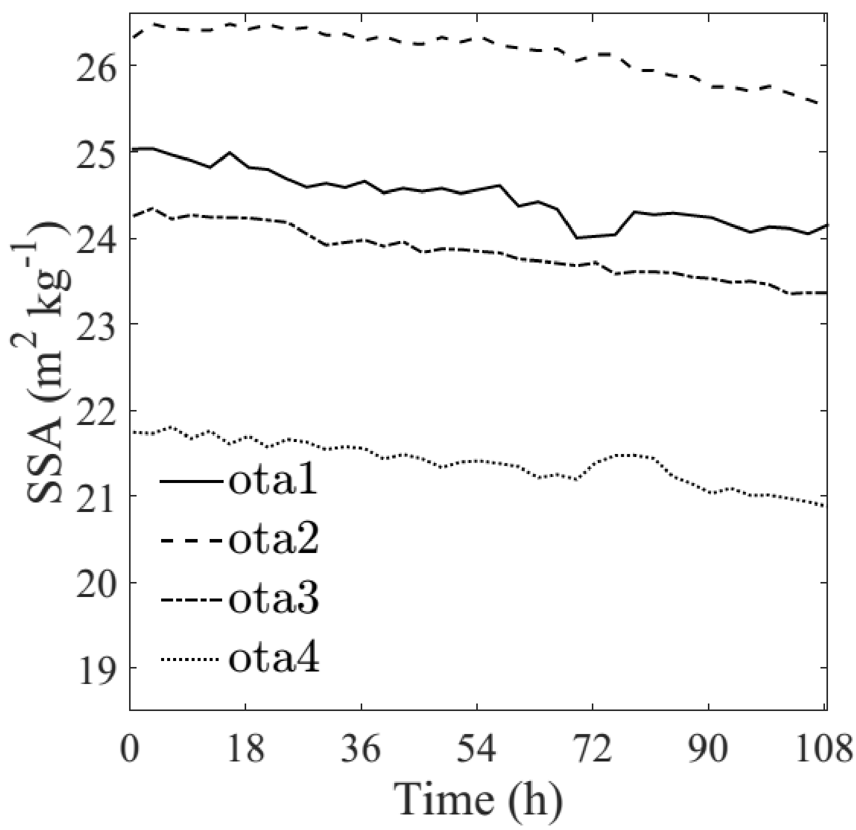

0.73
0.72
0.71
0.7
0.69
0.68
0.67
0.66
0.65
ε (-)
0
18
36
54
72
90
108
Time (h)
ota1
ota2
ota3
ota4

Fig. 4 b)



SSA (m^2 kg^-1)
26
25
24
23
22
21
20
19
ota1
ota2
ota3
ota4
0
18
36
54
72
90
108
Time (h)

Fig. 4 c)

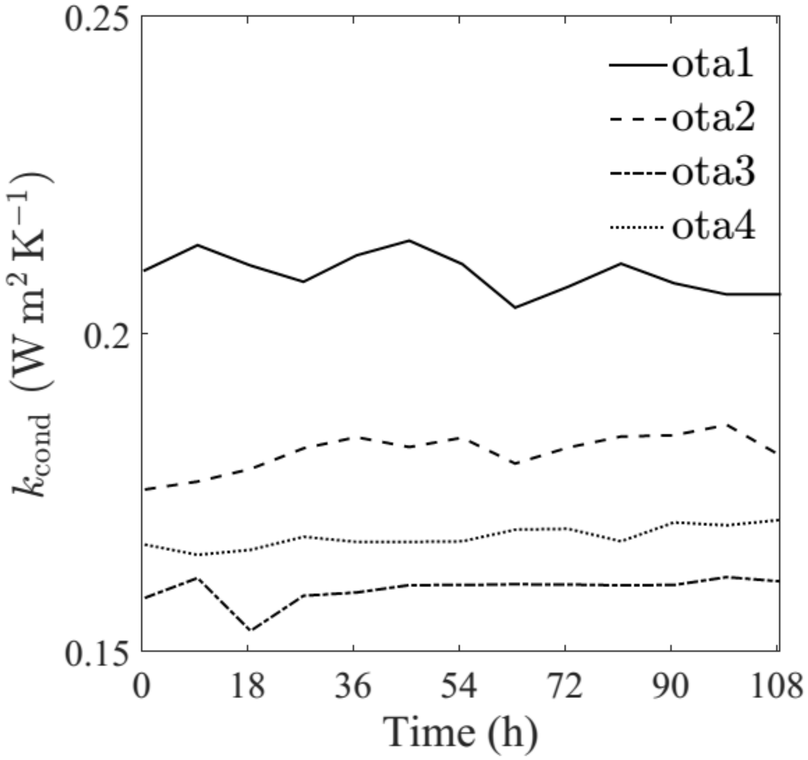

0.25
0.2
0.15
0
18
36
54
72
90
108
Time (h)
k_{cond} $(\mathrm{W\,m^2\,K^{-1}})$
ota1
ota2
ota3
ota4

Fig. 4 d)