Author comment to H. Löwe, Referee #2

We thank H. Löwe for his review of our work. Please find below our responses to the specific points raised.

General comments

The authors investigate the evolution of snow under a temperature gradient by microcomputed-tomography (CT) and analyze a large number of microstructural parameters for that case. In addition, numerical simulations for two effective macroscopic properties (thermal conductivity and permeability) are compared to the predictions of two homogenization schemes which are computed from parameters derived from the 3D microstructure.

In short, I think this is a very interesting paper with new results which eventually warrant publication. In particular considering the self-consistent (SC) approach as an alternative to previous efforts based on two-point bounds is a very interesting route worth investigating. However, subtleties of the SC method should be elaborated in greater depth, and a comparison to previous results should be included. My main comments are:

Assumptions of the SC approach not valid. The SC approach is based on a certain symmetry assumption, namely that both phases have "the same shape" (p.1421, l.5). Likewise, the depolarization tensor A, which characterizes the anisotropy of both phases is assumed to be equal for the ice and air contribution in Eq. (25). However, the analysis of the air and ice tortuosity tensors (Fig. 6), which are geometrical characterizations of the individual phases, clearly reveals that the ice-phase anisotropy A(tau_i) (cyan line) is different from the air-phase anisotropy A(tau_a) (purple line), in contradiction to the underlying assumption. This requires some clarification. In fact, where are the expected limits of applicability of the SC approach?

Missing explanations for the SC method. The SC scheme, if presented as an "analytical model", pretends to be a rigorous approach which can be applied without ambiguity. As far as I know this is not the case (cf. also first comment). In general, there is always some fundamental information missing in effective medium theories which cannot be provided by the method itself. Namely: what should be taken as the structural "ellipsoid-unit" which is represented by the depolarization tensor A? As a consequence, the effective ellipsoid must be guessed in the first place. This is in contrast to the series expansion, which is a rigorous, formally closed-form solution, where Q is unambiguously related to the correlation function. Surprisingly, the guess "use the correlation lengths" actually works quite well, at least for the time series analyzed here. This is however already a finding which does not follow from the SC method. These things should be explained to the reader in sufficient detail. The claim that the SC approximation, equipped with the exponential correlation lengths as definition for the effective ellipsoid, yields an estimate of the effective quantities of snow "as is", without any adjustment of prefactors, is a strong statement. However the generality of the statement (p. 1432, l.10) cannot cope with the amount of data (7 samples, one density). The statement should acknowledge the limitations or a few more samples should be analyzed to demonstrate that the SC can actually replace the bound-based parametrization which was derived, after all, from 176 snow samples.

In order to describe the transverse isotropic behavior of the snow, we chose to use a self-consistent estimate based on ellipsoidal inclusions. In general, several classes of ellipsoidal inclusions can be considered in the self consistent scheme, each class being characterized by a given orientation, an aspect ratio a/b, a volume fraction and its physical properties (Shafiro and Katchanov, 2000, Giraud et al, 2007, Kushch and Sevostianov, 2014). In practice, the determination of the orientation, the aspect ratio, the volume fraction of each class of ellipsoidal inclusions remains an issue of the method in the case of complex microstructure, as a porous media. Ideally, these properties could be obtained by developing specific algorithms in order to compute from the 3D images the geometric pore size/shape distribution not by using a classical isotropic structural element (sphere), but by taking an anisotropic structural element like an ellipsoid. The geometric ice size/shape distribution can be also computed in a similar manner. However, these computations are not straightforward.

In the present work, for the sake of simplicity, we decided to describe the snow microstructure, i.e. the air phase and the ice phase, by two classes of ellipsoidal inclusions with the same aspect ratio a/b, with a major axis collinear with the z axis of the sample, with volume fractions and (1-) and thermal conductivity k_a and $k_{i.}$ In the case of a porous medium where both phases are connected (as the snow), it seems reasonable, in first order of approximation, to assume that the aspect ratio a/b (of the air or the ice) is the order of (lc_x/lc_z), since the correlation lengths (lc_x, lc_y, lc_z) characterize the typical sizes of the heterogeneities (air and ice without distinction). As a consequence, in this particular case the depolarization tensor is equal in both phases and each phase is treated symmetrically. Let us remark that this "symmetry of the self consistent scheme" is not true if different classes of ellipsoidal inclusions with different aspect ratios are considered.

As underlined by the reviewer, both air and ice tortuosity tensor present different anisotropy. These tensors have been obtained by solving the boundary value problem (12), (13), (15), (16) over a REV in the case where (k_a =0, $k_{i.}$ =1) or (k_a =1, $k_{i.}$ =0). The anisotropy of these tensors reflects the coupling between the anisotropy of the microstructure (i.e. an assemblage of heterogeneities) and the physical phenomenon under consideration (diffusion or conduction in the air or ice phase), but this anisotropy c an not be simply linked to the aspect ratio a/b of one heterogeneity as suggested.

An analytical modeling of effective properties of porous media when the pore and solid phase are connected remains a challenge without doing some hypothesis on the microstructure. In many case, analytical models (dilute approximations, self consistent estimates, Mori-Tanaka estimates…) aim at considering the microstructure as an assemblage of "inclusions" within a matrix. The solution of equations for one isolated inclusion embedded in matrix (or in an equivalent medium) is usually sufficient to give an analytical expression of the sought effective properties. In the case of anisotropic materials, an ellipsoidal inclusion is usually chosen (Shafiro and Katchanov, 2000, Giraud et al, 2007, Kushch and V.I., Sevostianov, 2014). As already mentioned, the determination of the properties of each class of ellipsoidal inclusions remains an issue of the method in the case of complex microstructure. In the present paper, we clearly chose to get the simplest representation of the snow microstructure, based on simple measurements on 3D images. Even if this simple modeling seems sufficient at this stage in order to describe the main evolutions of the effective properties computed on the 3D images in this work, it is clear that comparisons with other set of data are needed in the future. Let us remark, that the self consistent estimates can be enriched first (i) by including more classes of ellipsoidal inclusions to describe the ice skeleton and pore network or (ii) by taking more realistic shape of inclusions (extracted from 3 images for example). However, in this latter case, this is possible numerically only. This type of enrichments will be probably necessary if one wants to capture the anisotropy of the snow at low density (see Figure 4).

Finally, as suggested by the reviewer, models based on exact contrast series of expansions appear as another interesting way to describe the evolution of the effective properties without hypothesis on the microstructure a priori. However, these series are complex and involves microstructural parameters (like the 3 or 4 - point correlation functions) which have been computed for a restricted number of microstructures in the literature to our knowledge (Torquato, 1991, 1998, Roberts, 2002).

The revised version of the manuscript has been modified in order to clarify some points discussed above.

Comparison to previous results. It seems appropriate to include a comparison of your results to the parametrization we developed before. This is just plug'n'play since your approach is based on the same parameter Q. Likewise, in the introduction it seems appropriate to state, besides “need to refine the parametrization” (p.1410, l.26) and “more systematic investigations are required” (p.1410, l.4/5), that in fact (Löwe etal 2013) already implemented a “refinement” exactly in the vein of the present paper, i.e. using standard homogenization methods which predict the relevance of the anisotropy parameter Q that is computed from the correlation function. I completely agree though, that the bounds have an aesthetic drawback of requiring an empirical correction, which is motivation enough to aim at improvements.
We added the following description: “Recently, Löwe et al (2013) proposed a refined parameterization of the effective thermal conductivity tensor of snow based on anisotropic, second order bounds. Their results show the importance of taking into account the microstructural anisotropy for the estimation of the effective thermal conductivity during a TG metamorphism.”
Figure 1 shows the comparison of the time evolution of the effective thermal conductivity from computations on 3-D images, the SC estimate and the parameterization of Löwe et al (2013). Figure 2 shows the same comparison for the time evolution of the anisotropy coefficient of the effective thermal conductivity.
In overall, both estimates are in agreement.

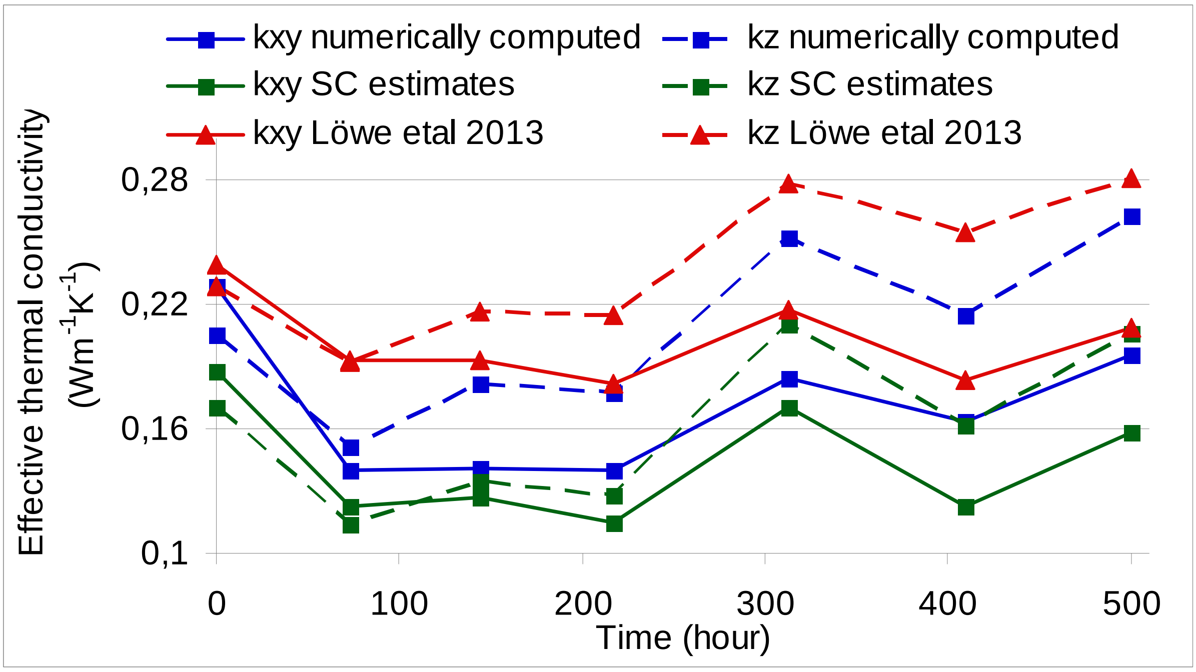

Figure 1 Effective thermal conductivity versus time estimated using three different methods.

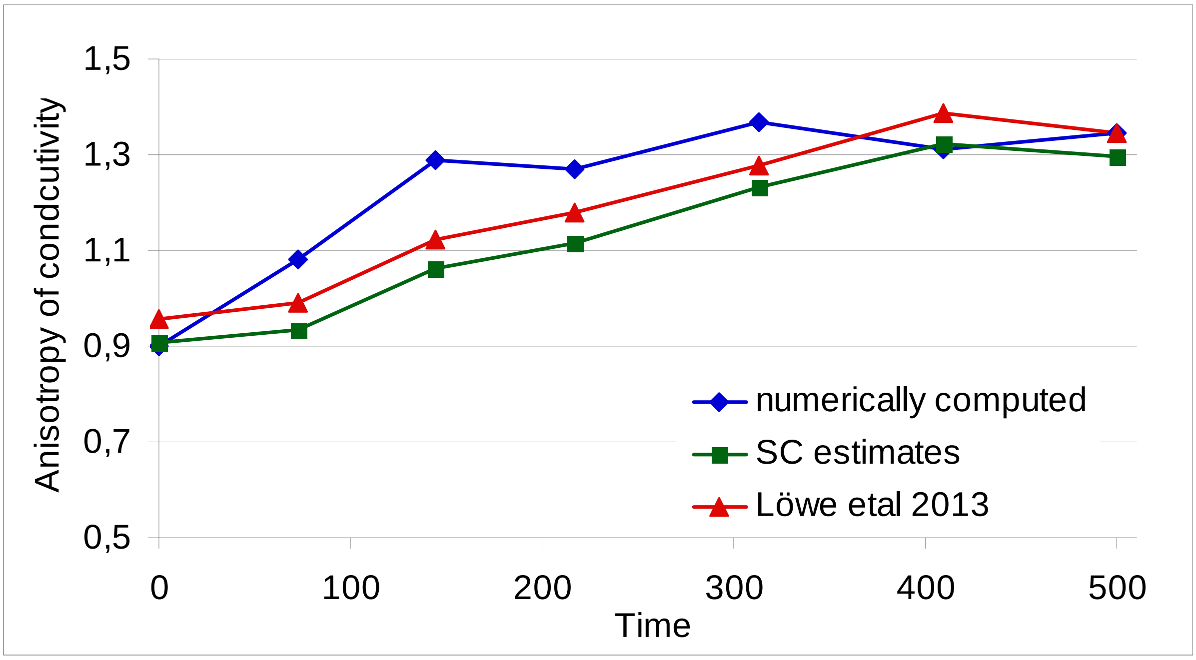

Figure 2 Anisotropy coefficient of effective thermal conductivity estimated using three different methods.

Curvature analysis. The most unique contribution of the present paper is certainly the analysis of curvature distributions for TG metamorphism and the resulting time evolution of the asymmetry between sublimating and growing surfaces, which has never been addressed before. Unfortunately, simply plotting the bare distributions (Fig. 7/8) gives only limited insight. Explicitly providing the mean and the variance of all distributions as a function of time seems to be illustrative for various reasons:

1. First, this allows to make a statement about the rescaling properties of the distribution if plotted over the scaling variable H/H(t) where H(t) is the area-averaged mean curvature at time t. (Note, that rescaling the curvature-axis also requires to rescale the vertical axis if interpreted as a probability). It seems that this rescaling should be possible for the upward faces, but this rescaling will clearly fail for the growing surfaces, at least in the beginning. This is suggested by Fig.7 where the distributions at t = 0 and t = 73 have clearly different mean values but roughly the same peak heights, which is not compatible with a scaling form p(H/H(t)) for the distribution.
We normalized each mean curvature distribution by the area-averaged mean curvature, for the upward and downward surfaces of ice (see figure below where "CM_moy" refers to the area-averaged mean curvature). The rescaled distributions seem roughly self-consistent for the upward surfaces but not for the downward surfaces where the peaks are not centered on a same value. The study of a self-consistent evolution of mean curvature is interesting but seems not straightforward for depth hoar.

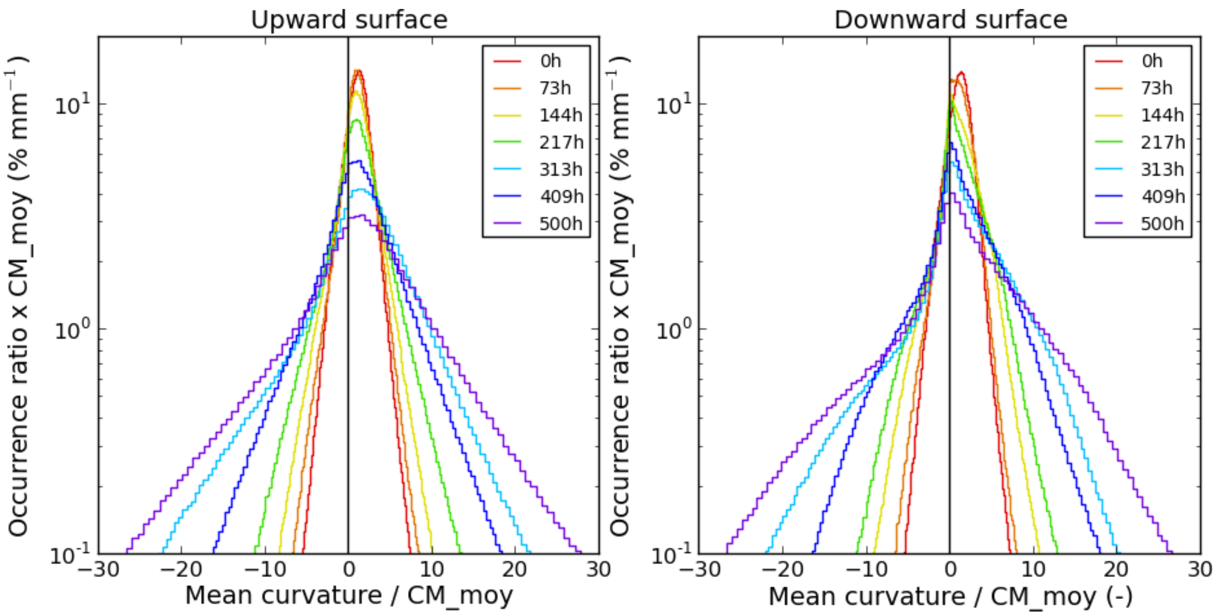

Figure 3 Distributions of the mean curvature rescaled by the area-averaged mean curvature value for the upward and downward surfaces.

2. Second, you could actually quantitatively check your claim from (p.1431, l.7) about different metamorphism stages (destruction of connection between grains in the beginning) from your data: You could evaluate the Euler characteristic X, as a measure for the connectivity, via the Gauss–Bonnet theorem which relates the integral Gaussian curvature to the connectivity/ topology of the structure. According to your claim, you should see a two-stage evolution of X, one related to decreasing connectivity (initial stage) and one related to a rather constant connectivity. Since X can be written as a product of the surface area and the area-averaged Gaussian curvature, this assessment can be done on-the-fly upon evaluation of the time evolution of the area-averaged mean and Gaussian curvature. A two-stage evolution w.r.t. the connectivity would be also interesting in comparison to the isothermal case (http://www.the-cryosphere-discuss.net/8/1795/2014/) where the very initial stage, if at all, is accompanied by an increase of the connectivity, at least for very low density snow.
Yes, we agree that the Euler characteristic X could confirm our observations. X is a good indicator of the topology of the microstructure, but its precise numerical estimation, and its quantitative interpretation, does not seem straightforward to us. In addition, this

parameter, does not take into account the precise shape nor the typical sizes of the microstructure, for which the distribution of Gaussian curvature (former Fig 8) seems more appropriate.

3. Finally, you could also prove your claim from (p. 1427, l.12) that the structure becomes monodisperse by showing that the standard deviation of the curvature distribution (normalized by the mean) decreases with time. The mere fact, that the width of the distribution decreases and the mean moves to zero does not necessarily imply that the structure becomes "monodisperse".
We modified the description. We added the values of area-averaged mean and Gaussian curvatures as well as the standard deviation for each distribution in Table 1 of the revised manuscript.

Overall, the paper might also benefit from language improvements (some suggestions given below, cf. Technical comments) and from figure improvements (cf. last remarks in Specific comments).

Specific comments
Thank you for the interesting comments. They helped to improve the manuscript. Please find below our responses to the specific points raised.

p.1408, l.15: The "bed of ellipsoids" and the "self consistent scheme" are different things, the first is a microstructure model and the second a homogenization scheme. I wouldn't refer to them on the same footing as analytical models.
We modified and referred to "analytical estimates".

p.1412, l.20: Were the cores drilled in horizontal directions?
The cores were drilled in the vertical direction.

p.1412, l.14: What do you mean by "stuck"?
The impregnated snow sample is "glued" on the upper part of the copper sample holder by a droplet of chloronaphthalene which, when refreezing, fixes the sample on the copper.
What are the dimensions of the final cylindrical core?
The snow cores which have been scanned to obtain the images 0A to 4A have a diameter of 0.9 cm and a height of around 1.3 cm. The snow cores which have been scanned to obtain the images 5G and 7G have a diameter of 1.6 cm and a height of around 2 cm.

What are the temperature fluctuations for the sample in the cryo-cell?
We do not know what the temperature fluctuations for the sample are. The temperature at the base of the sample is about -30°C and it is sufficient to avoid melting of our impregnated sample.

p.1412, l.20-25: Where is the vapor condensation avoided?
Inside the sample holder?
A schematic of the cryo-stage might be helpful.
We added an illustration of our cryogenic cell in the revised paper.
The impregnated snow sample is mounted on the sample holder and isolated from the air circulation by a Plexiglas cap. The dry air circulation in the chambers prevents the

outside air vapor to condense on the external sides (colder parts) of the Plexiglas interfaces.

p.1413, l.1: The given references contain a lot of image processing, please indicate briefly what was used from what.
We added the main stages of the applied image processing.

p.1413, l.22: I do understand what is meant here but a "surface area in x direction" might cause confusion for some readers, a surface area doesn't have a direction. One possibility is to define SSA_{β}^{-1} as a length scale λ_{β}, which is also a correlation length, as we have done it in (Löwe etal 2011). The other possibility is to spend more words on that, e.g. refer to specific surface area estimates SSA_{β} from different directions and explain the relation to the SSA in isotropic and anisotropic microstructures.
We now refer to "specific surface area estimates". Equation (1) defines unambiguously our expression of SSA_x, SSA_y, and SSA_z.

p.1414, l.22: Since the correlation function of snow is not an exponential, the notion of the correlation length is ambiguous. I would rather refer to it as one possible definition of a correlation length (see also previous comment).
Modified

p.1415, l.3: It might be helpful here to mention explicitly that the correlation length does not discern between ice and pore space.
Added

p.1415, l.9/11: Use either K or G for the Gaussian curvature. In general H,K (instead of C,G) are more common.
Modified

p.1415, l.25: What is meant by "second order estimates"?
It is a mistake, we replaced by "second order derivatives".

p.1416, l.3: "Variational approach" commonly refers to an underlying minimization principle, is this really meant here? What is minimized then?
The formulas (8) and (9) for Gaussian and mean curvatures are often used in variational approaches (see e.g. boundary displacement under curvature effects -Sethian 1999), but actually do not really proceed themselves from a minimization process. The text has been corrected to prevent any confusion.

p.1416, l.6: This is not a partial differential equation.
Modified

p.1418, l.10/11: The tortuosity is linked to the effective diffusion tensor of a nonreactive tracer, since for ka = 1, ki = 0 the internal boundary conditions leads to a zero flux Neumann condition for the diffusing species, i.e. the ice-air interface as a reflecting or non-reactive boundary. This is certainly not true for water vapor.
At the pore scale, on the ice-air interface, the flux of vapor is balanced by the sublimation-condensation effects (see Kaempfer et al 2005). Even if this flux is non-zero

at the interface, we have recently shown using the homogenization method of double scale expansions (Calonne et al 2014) that, under classical temperature gradients, the vapor transfer through the snow at the macroscopic scale is described by a classical diffusion equation with a source term arising from the ice-air interface. We have also shown that the effective diffusion tensor involved in this equation can be obtained by solving the boundary value problem (12-13), (15-16) when $k_a = 1, k_i = 0$. Let us remark, that these results are consistent with other studies involving phase changes at the interface (Geindreau and Auriault, 2002).

p.1418, l.14: Please give a citation for "it can be shown".
The reference (Bear, 1972) has been added

p.1420, l.1: Regarding the readjustment: The fit function $K^{fit}(\rho_s)$ obtained from previous data implicitly averages over all (unknown) values of Q, which were present in the previous data and which might be different from the present anisotropy values. So dividing by $K^{fit}(295)$ does not strictly remove only the influence of density. Any comment on that?
We agree with the comment. Nevertheless, density is the most influent parameter in the fit's expression; so we believe that the most important effect of our re-adjustment is to remove the density influence.

p.1420, l.18: As mentioned before, I wouldn't treat both models on the same footing of being "analytical models".
Modified

p.1420, l.21: (See general comments) This is not true, as far as I know. Using the correlation lengths for the effective ellipsoid is first and foremost an assumption, only for series expansions the relation to the microstructure is unambiguous and given via correlation functions.
Yes, we chose to use the correlation lengths to define the characteristics of the ellipsoid. We clarified this in the manuscript.

p.1421: The order of the section is odd, there is already quite some discussion of Eq (25) going on, before actually stating it.
We modified the structure of the section.

p.1422, Eq (29): Since the z component is by far more important from a practical point of view, it would make sense to give the z-component explicitly in Eq (29) and rather state how the x, y components can be obtained by the replacement $Q \rightarrow (1 - Q)/2$.
Added

p.1424, l.6: State what kind of a tensor product is meant here.
This point was clarified in the revised version.

Given, that you stress the "analytical" character of the the model you should later note in the discussion of the results that this is an ad-hoc modification.

We added the following sentence in the discussion (section 4.3 in the paper): "We recall that the estimate of permeability tensor has been then multiplied by the estimate of air tortuosity tensor to improve the description of the air phase connectivity".

p.1427: I am not sure if i got it right but are "occurrence ratios" meaningful quantities? Aren't these just bin frequencies which will depend on the chosen bin size of the curvature distributions? If not, you should show that.
The occurrence ratio of a given range of curvature corresponds to the ice surface area showing a curvature in the considered range of curvature divided by the total ice surface area and multiplied by 100.
We improved our definition in the manuscript.

Is it possible to estimate the total fraction of faceted surface area. This would be an important result.
Yes, we agree that this would be an important result, but this seems difficult. Facets are characterized by the local coexistence of flat and highly convex shapes and statistic information on curvature only cannot address this issue. To our opinion, this problem requires a real numerical recognition of the faceted shapes in 3D using specific image analysis methods, which seems out of the scope of this paper.

p.1427, l.16: Why do you discern between up/down only for the mean curvature? Please state (or show in a figure) that the Gaussian case is symmetric in view of up and down.
Yes, the Gaussian curvature is quasi symmetric in view of up and down (cf Figure 4). We added this remark in the manuscript.

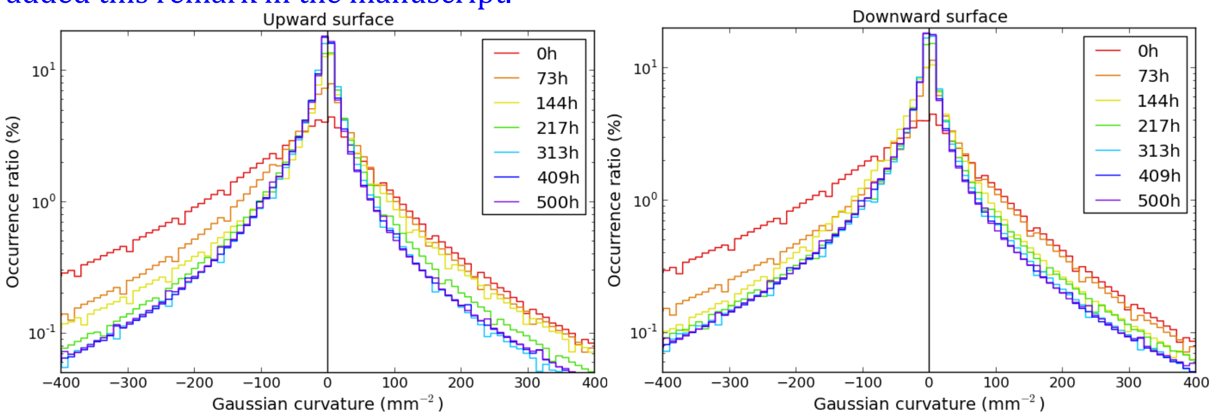

Figure 4 Time evolution of the Gaussian curvature distributions computed for the upward (left) and downward (right) surfaces of ice.

p.1428, l.14: Triple subscripts are cumbersome. Maybe denote the error by E(*), then this "only" leads to a double subscript.
Modified

p.1432, l.4: (and throughout) Referring to the simulations as "computed" is rather confusing since the "model" results are computed from the microstructure as well. I would discern between them by using something like "numerically exact" vs "modeled".
Throughout the paper, we tried to be clearer about the data we are talking about. As the description of the model as been improved, we believe that confusions between "modeled" and "computed" are unlikely. In the revised paper, we use the term "estimate" rather than "model".

p.1442, Figures: In general: The fonts are very small, its hard to read numbers and labels. Some figures (like Fig 6) might also benefit from using different symbols for different quantities.
Modified (see Fig 6)

p.1444, Fig.4: If this will be the size of the figure in the final manuscript, the figure itself is clearly too small, it is impossible to see something. Maybe re-arrange images to fill one column? Or enlarge the present image to have a two-column figure? p.1446, Fig.5: Same thing as before, everything is too small. The height of one subplot should be at least twice the present height.
Concerning the figures, upon acceptance of our article for final publication, we will adapt the font sizes to ensure readability in the final layout of *The Cryosphere* (different from *The Cryosphere Discussions*).

p.1446, Fig.6: Wrong legend, should be A(Ti), A(k), ... In the caption: coefficient → coefficients
Modified

p.1448, Fig.7: I think a semi-log scale might be advantageous in order to have visual access to the tails of the distribution.
We adopted the semi-log scale for the Gaussian curvature distribution (Figure 8). This scale seems less advantageous for the mean curvature distribution.

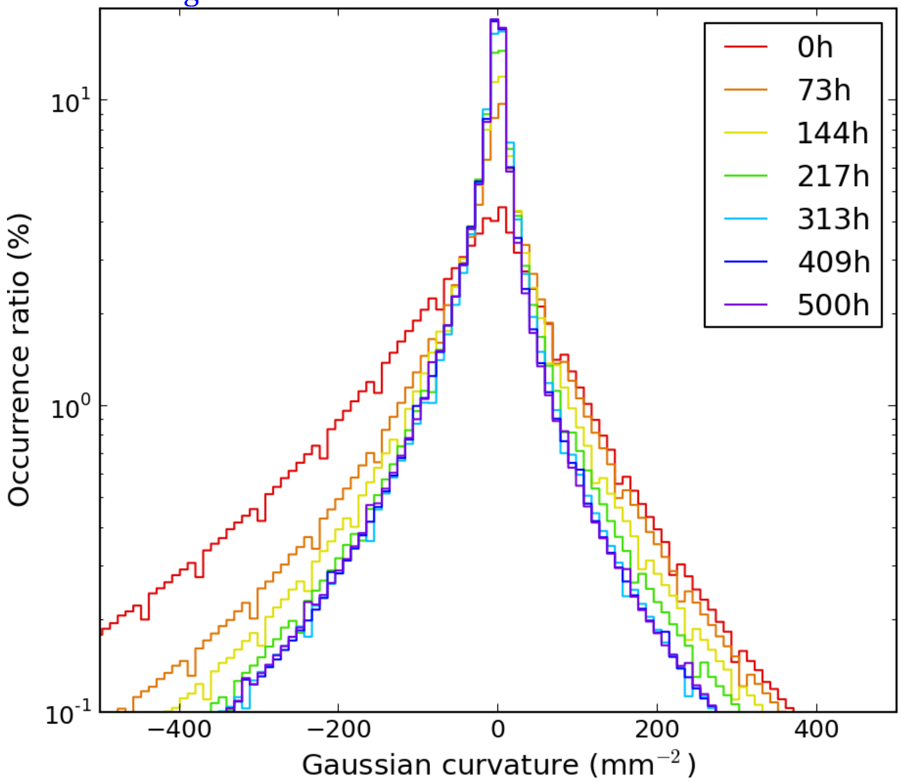

Figure 5 Time evolution of the Gaussian curvature distributions expressed in semi-log scale.

Technical comments

We thank H. Löwe for his technical comments. They have been taken into account in the revised version of the manuscript.
p.1408, l.2: during a temperature → during temperature (cf. also the title)
p.1408, l.3: along the vertical → in vertical direction

p.1408, l.6: panel → set
p.1408, l.9: delete “a”
p.1409, l.2: What does “close environment” mean?
p.1410, l.5: specific anisotropic behaviors → anisotropic behaviour
p.1410, l.14: undergone → encountered
p.1410, l.15: sampled → drilled
p.1410, l.18: delete “a”
p.1411, l.26: punctually → temporarily
p.1412, l.6: porosities → pores
p.1412, l.8: impregnation → casting
p.1412, l.10: delete 2nd “the”
p.1412, l.11: insure → ensure
p.1412, l.11: correct → good
p.1412, l.13: a drill mounted on a lathe operating → a drill which is mounted on a lathe and operated
p.1412, l.19: operates → is operated
p.1413, l.12: noted → denoted by
p.1414, l.2: delete “a”
p.1416, l.20: on → of
p.1418, l.12: is constituted of → constitutes of
p.1418, l.15: strongly → highly
p.1419, l.9: note → denote by
p.1421, l.22: The inversion symbol (•)−1 and sc-superscript read more like sc − 1. I suggest to use brackets: ksc−1 → [ksc]−1
p.1422, l.17: reported → shown
p.1422, l.15: depicts → shows
p.1425, l.13: close from → close to
p.1425, l.26: “all directions combined” unclear, not a full sentence.
p.1426, l.11: see last comment.
p.1426, l.26: pointed out above with the Fig → pointed out above and shown in Fig
p.1427, l.6: proportion → ratio
p.1428, l.11: → In order to compare models and experiments quantitatively, we use the mean relative difference E, defined as
p.1430, l.5: can be considered as constituted by → can be considered as being comprised of
p.1430, l.22: proportion → volume fraction
p.1430, l.22: “way for heat conduction” unclear, not a sentence
p.1441, Fig1: forward → front

References

Bear, J.: Dynamics of Fluids in Porous Media, Dover, 1972.
Calonne, N., Geindreau, C., and Flin, F.: Macroscopic modelling for heat and mass transport in dry snow using a homogenization method, Journal of Physical Chemistry B, in prep, 2014.
Geindreau, C. and Auriault, J.-L.: Transport phenomena in saturated porous media undergoing liquid-solid phase change, Arch. Mech., 53, 385-420, 2001.

Giraud, A., Gruescu, C., Do, D. P., Homand, F., and Kondo, D.: Effective thermal conductivity of transversely isotropic media with arbitrary oriented ellipsoidal inhomogeneities., International Journal of Solids and Structures, 44(9), 2627-2647, 2007.

Kaempfer, T., Schneebeli, M., and Sokratov, S.: A microstructural approach to model heat transfer in snow, Geophys. Res. Lett., 32, L21503, doi:10.1029/2005GL023873, 2005.

Kushch, V. I. and Sevostianov, I.: Dipole moments, property contribution tensors and effective conductivity of anisotropic particulate composites, International Journal of Engineering Science, 74, 15–34, doi:http://dx.doi.org/10.1016/j.ijengsci.2013.08.002, 2014.

Löwe, H., Riche, F., and Schneebeli, M.: A general treatment of snow microstructure exemplified by an improved relation for thermal conductivity, The Cryosphere, 7, 1473-1480, doi:10.5194/tc-7-1473-2013, 2013.

Roberts, A. P., and Garboczi, E. J.: Computation of the linear elastic properties of random porous materials with a wide variety of microstructure, Proceedings of the Royal Society of London, 458, 2021, 1033-1054, 2002.

Shafiro, B., and Kachanov, M.: Anisotropic effective conductivity of materials with nonrandomly oriented inclusions of diverse ellipsoidal shapes, Journal of Applied Physics, 87, 8561-8569, 2000.

Torquato, S.: Random heterogeneous media: microstructure and improved bounds on the effective properties, Appl. Mech. Rev, 44, 37–76, 1991.

Torquato, S.: Effective stiffness tensor of composite media: ii. application to isotropic dispersions, J. Mech. Phys. Solids, 46, 1411-1440, 1998.