

**Comments on Manuscript #tc-2019-143:**

***“Revisiting the vapor diffusion coefficient in dry snow”***

***Andrew Hansen***

**General Comments:**

This manuscript proposes to revisit the concept of Apparent Diffusion Coefficient (ADC) in dry snow, which is known to have led to a quite extensive literature, exhibiting a high dispersion in the quantitative results. The author first presents the main studies in a concise, balanced and objective way. He then proposes a definition for the ADC with 4 specific mechanisms that need to be taken into account for its accurate estimation. The author then proposes 3 theoretical models of increasing difficulty to express the ADC in terms of pore and ice volume fractions. He then revisits most of the literature studies, and takes into account their peculiarities to provide ADC estimations that are consistent with those of the proposed definition, showing that ADC values are generally between 1 and 1.3, with a much lower dispersion than that given by the original literature studies.

This manuscript is a sound and interesting contribution to the vapor coefficient literature, and allows better deciphering the high dispersion of the quantitative results observed. In overall, it is clear, didactic and well written, but would benefit from (i) a clearer statement of the main model hypotheses and (ii) additional explanations (e.g. ice blockage exactly compensated by shortened diffusion paths).

Here are some suggestions the author may find useful to improve the manuscript:

**1. Objectives and definitions:**

To my understanding, the diffusion coefficient the author is interested in is not really an effective diffusion coefficient, but an apparent one: it takes into account mechanisms that are not really based on diffusion processes (e.g. phase changes), but consider them as contributing to the overall diffusion.

To my opinion, the estimation of the ADC is close to an ill-defined problem: I think it would be better not to hide specific processes in an apparent diffusion coefficient, but to address each physical process separately. It is important (i) for a better understanding of the different processes and (ii) for a better modeling of the vapor transfer through the snow cover (modular approach, with possible separate improvements). As the author knows, such a modeling has been e.g. proposed in Calonne et al 2014 and Calonne et al 2015, where specific source terms are used to describe phase changes. Microscale studies of TG metamorphism in controlled conditions (e.g. Pinzer et al, 2012, Calonne et al, 2015, etc.) and appropriate simulations (e.g. Kaempfer and Plapp, 2009) are also very promising means to better understand and identify the different processes, leading to a better quantification of macroscopic vapor transfers in snow.

Given the extensive (and extremely confusing) literature on the vapor diffusion coefficient of snow, I think this manuscript is a sound and interesting contribution to snow science, as it provides simple ways to estimate the order of magnitude of the apparent diffusion coefficient, and, last but not least, clarifies and reconciles most of literature results.

However, I would suggest the following improvements:

- stating more clearly from the start of the manuscript that the diffusion coefficient addressed here is an apparent diffusion coefficient (e.g. in the title, abstract, introduction, definition part...).
- recalling the objectives of ADC estimations, and if adequate, explaining the interest of ADC computations over other approaches (e.g. considering the effective diffusion coefficient and phase change effects separately).

2. Exact compensation of ice blockage by shortened diffusion paths (see p. 17 and 22):

I had some difficulties to understand this point. Adding some explanations would help the reader (see detailed comments).

3. Surface Kinetics:

From the manuscript, I understand the models would work especially well when diffusion through pores is very slow as compared to sublimation/deposition processes. Would the model still be valid if surface kinetics phenomena are of the same order or longer than diffusion times? At least a small paragraph would be welcome on this topic.

4. Discussion of the model parameters and domain of validity of the proposed results.

From the 3 equations (55 to 57) used by the author to model the ADC and from the related figures (Fig. 8-9), it clearly appears that the diffusion enhancement only depends on the volume fraction of ice (1 parameter only). This description has the advantage to be very simple, but raise several questions, e.g.:

-What about the impact of the ice microstructure (grain and pore size, connectivity, ice shapes, anisotropy...) on the real ADC? E.g., are the results the same for an horizontally (cf Fig. 6) or a vertically oriented layered microstructure of same volume fractions? Have large open pore structures the same ADC than small closed pore structures of the same volume fractions? Formulas have been obtained under strong hypotheses (isotropy, e.g.), but problems related to ADC are inherently linked to snow microstructures obtained under strong TG, i.e., which typically involve the formation of vertically elongated anisotropic structures.

-What about temperature effects? When temperature decreases (e.g. from -0.5 to -5°C), metamorphism is known to be strongly inhibited while diffusion coefficient and conductivities stay nearly the same (cf e.g. Massmann, 1998 and Calonne et al, 2011). Is the real ADC impacted by temperature and how is it reflected by the proposed formulas?

All these questions are probably difficult to precisely answer in this manuscript but some hints could be given in the discussion.

At least, the main hypotheses of the model should be recalled in the conclusion part.

#### Detailed and minor comments:

p. 2, lines 3-4: *"The net effect is that ice grains act as an instantaneous source and sink of water vapor, thereby shortening the diffusion path of a water molecule"*.

I would replace "instantaneous" by "quasi-instantaneous" (here, and everywhere in the paper) : depending on the conditions (temperature, facet orientation, etc.), the kinetics at the interface may strongly impede the phase change and impact the resulting geometry. See e.g. Yokoyama, 1990; Libbrecht et al, 2005; Flin and Brzoska, 2008; Libbrecht and Rickerby, 2013, etc.

p. 3, lines 1-2: *"For instance, the numerical studies of Criston (1990), Pinzer et al. (2012), and Calonne et al. (2014) all use different methods to evaluate the mass flux and/or the diffusion coefficient."*

Actually, as truly explained by the author just a line before, the main problem is probably not the method, but the definition. E.g., in Calonne et al, 2014, we did not consider the sublimation condensation effects (mechanism #2 of the author's definition), as it is not a real diffusion process, but a phase change that can be viewed as contributing to the overall apparent diffusion process. Replacing "methods" by "definitions" might probably be more accurate.

p. 4, lines 15-19: *"Convection is neglected. Convection only occurs in extreme weather conditions such as near the top of a snowpack in the presence of a **strong wind** or extremely large temperature gradients. Foslien (1994) provides support for this assumption through the calculation of a Rayleigh*

*number for porous media. His results show the Rayleigh for snow is an order of magnitude below the number required for the onset of convection...*

What kind of convection the author actually wants to address here?

-1) Forced convection as in Calonne et al, 2015 (i.e., wind pumping -with e.g. Reynolds number, etc.)

or

-2) Natural convection as in Kaempfer et al (2005) (i.e., natural convection, due to the fact that cold air is heavier than warm one -with Rayleigh number)?

As the author know, the physical mechanisms (and associated characteristic numbers) are different for these two distinct processes. It would be better to address these two process in distinct paragraphs.

p. 8, line 15-16: *"Vapor diffusion in snow is driven by temperature gradients and, therefore, it is useful to examine relations between macroscale and microscale temperature gradients."*

This is a small detail, but strictly speaking, the first part of the sentence is not perfectly true, as vapor diffusion in snow can be also caused by curvature effects (see e.g. Brzoska et al, 2008), at least at microscale. Replacing "Vapor diffusion in snow" by "Macroscale vapor diffusion in snow" or "Large scale vapor diffusion in snow" would be more accurate.

p. 6, line 10: *"To account for this altered time scale for water vapor diffusion, the notion of intrinsic time,..."*

For me, the terminology "intrinsic" is unclear in this context. I would suggest to replace "intrinsic" by "apparent", here and everywhere in the text.

p. 16, Fig. 7: *"x-axis: kg m<sup>-3</sup>"*

=> blank spaces to be suppressed.

p. 16, Fig. 7: *"Figure 7. Thermal conductivity analytical prediction of Foslien (1994) versus finite element predictions of Riche and Schneebeli (2013)."*

To my understanding, the comparison made here is a comparison with a very specific fit of the dataset of Riche and Schneebeli (2013), namely the vertical component of the thermal effective conductivity, and for FC and DH only (see Fig 7, p 224: "kz FC and DH").

In addition, I am not sure the red curve of Riche and Schneebeli (2013) can be really considered as "a finite element prediction": for me, it is rather a fit computed from a subset of kz values (FC and DH only), obtained from their whole dataset.

p. 16, line 10: missing brackets or parentheses.

p. 17, lines 14-15: *"Preserving the ratio of (ki/kha) is a necessity to preserve the local temperature gradient field obtained from the heat transfer analysis."*

At first glance, I found this idea quite difficult to understand. The author should add some explanations (or an example on a specific configuration, with a figure?).

p. 17, lines 26-27: *"Note that the averaging process is done over the humid air volume as opposed to the total volume as the effects of ice blockage and the shortened diffusion paths from hand-to-hand vapor transport cancel each other out."*

Same as just before: some additional explanations would be welcome.

p. 21, lines 20-25: *"Perhaps the most extensive numerical study of mass transport is found in Calonne et al. (2014). They performed finite element analyses for 35 RVE's of snow spanning a density range from 100–500 kgm<sup>-3</sup>. Unfortunately, it is not possible to reconcile their results with the analytical models presented, as they solved a fundamentally different boundary value problem for mass transfer driven by the relation substitutions shown in (51) where the thermal conductivity of ice was set to zero. In contrast, in the analytical models, the influence of ki is retained to obtain the local temperature gradient field. The influence of ki is then removed in the calculation of the diffusion coefficient."*

I agree with the author. However, it should be mentioned that, in the model of Calonne et al, phase change can be described using specific sources terms (see Calonne et al, 2014 and Calonne et al, 2015).

p. 22, lines 15-16: *“Arguments from stereology, combined with experimental observation, suggest that the influence of ice blocking diffusion paths is canceled out by the shortened diffusion paths from hand to hand vapor transport.”*

Same as p. 17, lines 26-27: some additional explanations or justifications would be welcome.

p. 22, lines 26-27: *“Importantly, Eq. (59) represents the mass flux per unit area of snow and not just the flux across the humid air phase as the ice blockage impeding diffusion is exactly countered by the shortened diffusion paths of water molecules”*

Same as just before: some additional explanations or justifications would be welcome.

Minor typo: a “t” is missing in “importantly”.

## References

- Brzoska, J.-B., F. Flin and J. Barckicke, 2008. Explicit iterative computation of diffusive vapour field in the 3-D snow matrix : preliminary results for low flux metamorphism, *Ann. Glaciol.*, 48, 13-18, doi: 10.3189/172756408784700798.
- Calonne, N., C. Geindreau, F. Flin, 2014. Macroscopic modeling for heat and water vapor transfer in dry snow by homogenization, *J. Phys. Chem. B*, 118 (47), 13393–13403, doi : 10.1021/jp5052535.
- Calonne, N., C. Geindreau, F. Flin, 2015. Macroscopic modeling of heat and water vapor transfer with phase change in dry snow based on an upscaling method : Influence of air convection, *J. Geophys. Res. : Earth Surf.*, 120, 2476-2497, doi : 10.1002/2015JF003605.
- Calonne, N., et al., 2015. CellDyM: A room temperature operating cryogenic cell for the dynamic monitoring of snow metamorphism by time-lapse X-ray microtomography, *Geophysical Research Letters*, 42 (10), 3911–3918.
- Flin, F. and J.-B. Brzoska, 2008. The temperature gradient metamorphism of snow : vapour diffusion model and application to tomographic images, *Ann. Glaciol.*, 49, 17-21, doi: 10.3189/172756408787814834.
- Foslien, W., 1994. A modern mixture theory applied to heat and mass transfer in snow, M.S. thesis, University of Wyoming, Laramie, WY, USA.
- Kaempfer, T. U., and M. Plapp, 2009. Phase-field modeling of dry snow metamorphism, *Physical Review E*, 79 (3), 031,502.
- Kaempfer, T. U., M. Schneebeli, M., and S. A. Sokratov, 2005. A microstructural approach to model heat transfer in snow, *Geo. Res. Lett.*, 32, <https://doi.org/10.1029/2005GL023873>.
- Libbrecht, K. G., 2005. The physics of snow crystals, *Reports on Progress in Physics*, 68 (4), 855–895, doi: 10.1088/0034-4885/68/4/R03.
- Libbrecht, K. G., and M. E. Rickerby, 2013. Measurements of surface attachment kinetics for faceted ice crystal growth, *Journal of Crystal Growth*, 377, 1–8.
- Massman, W. A review of the molecular diffusivities of H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>, CO, O<sub>3</sub>, SO<sub>2</sub>, NH<sub>3</sub>, N<sub>2</sub>O, NO, and NO<sub>2</sub> in air, O<sub>2</sub> and N<sub>2</sub> near STP. *Atmos. Environ.* 1998, 32, 1111–1127.
- Pinzer, B. R., M. Schneebeli, and T. U. Kaempfer, 2012. Vapor flux and recrystallization during dry snow metamorphism under a steady temperature gradient as observed by time-lapse micro-tomography, *The Cryosphere*, 6, 1141–1155, <https://doi.org/10.5194/tc-6-1141-2012>.
- Riche, F. and M. Schneebeli, 2013. Thermal conductivity of snow measured by three independent methods and anisotropy considerations, *The Cryosphere*, 7, 217–227, <https://doi.org/10.5194/tc-7-217-2013>, 2013.
- Yokoyama, E., and T. Kuroda, 1990. Pattern formation in growth of snow crystals occurring in the surface kinetic process and the diffusion process, *Physical Review A*, 41 (4), 2038.