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Interactive comment on "Revisiting the vapor diffusion coefficient in dry snow" *by* Andrew Hansen

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1 General comments

This paper addresses a long standing and vividly debated topic in the snow science community and makes a contribution by unifying the definition of effective diffusion. The paper converts the wide variety of predicted and measured effective diffusive coefficient D_s to reasonable range. The paper starts from a mixture theory representation of transport of mass and energy in snow and defines an upper bound for the effective vapor diffusion coefficient as a function the density of snow. By introducing an objective definition of vapor transport one can compare several previous experimental studies on effective vapor transport. The paper shows that all measurements are well below the upper bound. In addition it formulates two models based on diffusion with and without

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tortuosity.

I agree with the author that effective vapor diffusion is a mind boggling topic and can become confusing from time to time. The author attempts to make a first step in clearing the clutter by starting from the first principles of mixture theory. He identifies 4 ways the diffusion path of water can be altered with respect to pure vapor diffusion without an ice matrix interfering. 1) Blockage of diffusion paths by ice, 2) Shortened diffusion paths due to the phase transition at the interface. 3) Enhanced local diffusion by difference in heat conduction in both phases. 4) Tortuosity, lengthening diffusion paths. The author states that any model for the effective diffusion coefficient these 4 principles should be accounted for.

The author introduces an artificial velocity of the vapor molecules that is calculated by the path length that it travels divided by the time spend in the vapor phase only, effectively deleting the time it spends in the ice phase. Hereby the definition of the effective diffusion coefficient is enhanced. Based on this concept, the author defines an upper bound for D_s, which is a density correction based on this ice volume fraction.

The first model, which is a superposition of a layered and a tubular microstructure, leads to a slightly different density correction. The last model that should include tortuosity and is based on the method developed by Calonne et al. (2014). They calculate the effective diffusion by changing transport coefficients from the heat conductivity problem in terms of the transport of the vapor flux coefficients. Here the transport in the ice phase was set to 0. In this work the ice transport coefficient is set to 100 times that of vapor, to implement the idea of shortened pathways or intrinsic/artificial velocity.

My main concern is on the introduction of the intrinsic time or intrinsic/artificial velocity. It is introduced to delete the time the vapor molecules spend in the ice phase, yielding an effective higher velocity to the vapor molecules, since the distance is that it apparently travelled through the ice phase is still counted. If this approach of apparent mass transfer is taken, the apparent movement of the ice matrix in opposite direction of the flux should be accounted for as well. The latter is important, because without it the mass balance of your snow pack wouldn't be complete. The intrinsic time is confusing and as concluded in the end of the paper is not really necessary. However, it is still the basis to compute the effective diffusion coefficient, which raises the question if the meaning of this definition is physical or not.

The main crux is to assess whether the hand-to-hand vapor transport is physically related to mass flux. Since the water molecules diffusivity in ice is orders of magnitudes slower giving it a large of infinite average velocity is not physical. Mechanisms to which actual transport is being enhanced is if the local vapor flux in the pore space is on average more enhanced, than the reductions that are expected. It is known that tortuosity τ is leading to an upper bound for $D_s^* = (1 - \phi_i) D_{v-a}/\tau$ (Pismen, 1974). Note that this is only a reduction of D_{v-a}. A heterogeneous distribution of temperature gradients in porous media will lead to actual locally enhanced vapor flux. Bounds for these fields could be found/developed based on e.g. Torquato (2002), but as far as I know, cannot exceed a density correction factor, as the author suggests for his upper bound.

My interpretation of the given models is that they use the linearization between the vapor concentration gradient and the temperature gradient at the microscale and find density corrections for the latter given 1) a layered, 2) a mix between tubular and layered microstructures. The physical interpretation of the last model is not clear to me, especially how tortuosity is included. If the models are restated in the context of enhanced temperature gradients they might be more physical/useful.

In conclusion, the debate around the effective diffusion coefficient is in need for a uniform and clear definition. The approach which is chosen in this paper, is interesting, but also highly confusing. In my opinion the effective diffusion coefficient is a linear response of water vapor and ice transport to a thermal driving force. Since the phase transitions that take place at the microscale serve as a temporal storage of vapor, i.e. ice, it should in principle reduce the effective transport, and therefore reduce the effective diffusion coefficient. Given that it is the authors choice to define it in a different manner, I will respond with some specific comments below.

2 Specific comments

2.1 Section 1: History

- General: Introduction is quit sparse regarding the explanation of the different mechanisms for enhanced vapor diffusion. For understanding the introduction can be more detailed. example:
- p.2. l.6: More explanation of this experiment of Sommerfeld et al. (1987) would clarify how this was measured, which assumptions are made etc...same for the studies Pinzer et al. (2012) and Calonne et al. (2014)
- p.2 l.32: What is physical difference between blockage of diffusion paths and tortuosity?
- General: From the history or the introduction, it should become clear to the reader which studies have been performed and where the discrepancies lie. To me it is not clear without reading the actual referenced papers what has been measured, modeled or simulated. Suggestion: start with stating that differences in studies are mainly based on the assumptions of contributing mechanisms and the lack of a consistent definition, and depart from there by describing which assumptions they made.

2.2 Section 2: Mass transfer in snow

- p.5. l.8: If isotropy can be relaxed please discuss it somewhere at a later stage.

- p.6. l.11: The introduction of apparent time is confusing to me. If we were to couple measured diffusion to mass transport, we would need to add the residence time spend in the crystal to the time travelled in humid air? If you make the connection to residence time, one could 'track' single molecules.
- p.7. l.15: With eq.11 the definition of D_s is given, which is basically the effective linear response to the physical driving force. Maybe consider this as a natural bridge to describe what it is effected by given the physics at the microscale.
- p.7. l.20: In this paragraph we suppose to be convinced that it is useful to frame D_s in connection to the balance equations. We know that they are dependent on D_s but a quick calculation of the relative importance of the second term is less than 5 ‰.
- p.8. l.9: Despite the title, there is no formal definition of $D_s = \ldots$. One could use eq.(11) but then it means that is connected to the flux of snow, which can only be understood in terms of mass flux.
- p.8. Fig.3: This figure is confusing, suggesting that the bonds are colder than the grains, by the colorbar. The RVE is also on the small side, usually one expects the system size to be roughly 10 times the element (grain) size.
- p.9. l.15: By taking out $d\gamma_v/d\theta$ of the integral, you make quite some assumptions, i.e. the temperature gradient and the vapor gradient are locally always reacting the same, regardless of its surroundings. In principle one should solve the Laplace equation for your vapor density, given the appropriate boundary conditions, which can include the interface dynamics of the phase transition. Stating clearly which assumptions you make is in place here.
- p.9. l.22: Suggesting linking macro to micro scale by explicitly relating j_s to $\overline{j_v}$, which gives you an expression for D_s that shows by what physics it is influenced.

- p.10 l.1: I believe 1) and 4) are in principle the same. From a microstructural perspective I would suggest that there are two mechanisms: 1) Tortuosity that increases the free path length of water vapor molecules. and 2) Phase transitions that, driven by temperature gradients at the ice-air interface, change the boundary conditions for the mass transfer of the vapor concentration and therefore alters the diffusion paths in a structural manner.

- p.10. l.9: My general thought is that it would not be correct to use v^*, since actual molecules aren't travelling through the ice phase with (in)finite speed, and if one looks at it from an apparent perspective, one has to account for the apparent movement of the ice crystals in the opposite direction that is represented by the ice interface velocity in opposite direction.

2.3 Section 3: Models

- p.14 l.4 & Fig 5: The argument that blockage and shortening of diffusion paths are equally frequent is based on the frequency of the ice phase, which is, for isotropic media the same. Given that the chance for shortening or blocking is given by underlying physics one still need to solve the vapor diffusion equation including the correct boundary conditions. Therefore the relative chances can still be very different.

- p.15. l.1: Is the underlying assumption that the microstructure is a superposition of a layered and pore structures?

- p.15. l.4: the naming Pore, is highly confusing. I would suggest something like tubular microstructure.

- p.15. l.7: There is only a slight difference in figure 7, but there is no information on how well this fit is doing to predict real conductivities.

- p.16. Fig. 7: What is the purpose of this figure? In the caption you write finite element predictions, but this should be a fit to finite element simulations.
- p.17. l.16: This is just not physical in terms of the [Calonne.2014] paper.
- p.18. l.6: This equation is not derived but stated? It is hard to follow this line of thought without understanding the appendix of Calonne et al. (2014)
- p.18. l.7: How is tortuosity exactly included? Tortuosity is generally described by the path that molecules travel through a porous media without altered paths due to phase transitions, which is basically governed by laminar flow in an inert media. How is this relevant or included in this model?
- p.19. Fig.8: Redundant figure.
- p.20. l.10: There are 6 values plotted, but I can't retrace them to measurements from the [Pinzer.2012] paper, a little more information could be helpful on how you did this, which figures or data you actually used.

2.4 Section 4: Conclusion

- As the author states eq.(59) should be the vapor mass flux in snow. There is no need for an introduction of shortened diffusion paths, or intrinsic velocity, because it is not physical. In the conclusion it is stated that in principle the definition of D_s is the same as eq.59. This to me is confusing because you use v^* in eq.(11). Maybe I'm missing the general point of the paper, but one way or the other this should be clarified.

Figures: In general the figures are not illustrative to the paper. Fig 3 is very confusing, suggesting bonds are colder than grains. My suggestion is to replace Fig 1,2,3 with a slice of a 3D image of actual snow and indicate the different processes leading to vapor diffusion (tortuosity and phase transitions at the interface)

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3 Technical comments

- Notation: The author decides on using θ as a symbol for temperature and ξ as symbol for coordinates. This might be the convention in mixture theory, but for readability and adaptation to the snow community I would advise on T and x respectively.
- Use of the word ‘may’ in e.g. p.7.13 sounds to me as if the followed expression is lucrative in the sense that we may also use something else. The word ‘can’ in this context would be more appropriate, however, it is a matter of taste.
- p.3 l.19: typo: appropriate.
- p.7 l.9: typo: $4(10)$
- p.16 l.1: sans, without
- p.16 l.10: missing normal vector.
- p.26 l.17: M.sc. thesis is usually not peer reviewed.

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