Response to Andrew Hansen’s comment on tc-2020-317

Andrew Hansen supports the idea that water vapor diffusive fluxes in snow (expressed as mass fluxes per surface of snow) can be enhanced compared to those occurring in free air under similar vapor concentration gradients. For that two arguments are advanced:

- The “hand-to-hand” mechanism, in which the deposition of one water molecule on one side of an ice grain and the concurrent sublimation of another molecule on the other side of the grain is viewed as a real mechanism of mass transfer (as if the depositing molecule instantly traveled through the ice phase).
- An analytical expression of the effective thermal conductivity of a layered snow structure, including latent heat effects, from which the macroscopic vapor flux is derived. This derivation appears in the technical document of the comment, in Hansen and Folsien (2015), and in Hansen (2019).

Our brief answers to these two points are:

- The proposed mechanism of water molecules instantly traveling through the ice phase is nonphysical. During the concurrent deposition and sublimation of water molecules on two opposite sides of an ice grain, no mass is physically moved in the process (the condensing molecule stay below the ice grain and the sublimating molecule already was above). This is completely different from the situation in which this molecule would actually be transported on the other side of the ice grain, as proposed by the hand-to-hand mechanism. This has been addressed in detail in Fourteau et al. (2021) and their Figure 1 and relevant text in their paper explain this in detail.
- The derivation of Eq 16 in the technical document proposed by Andrew Hansen, and which is also present in Hansen and Folsien (2015) and Hansen (2019), is invalid.

As explained in Appendix C of Fourteau et al. (2021), there are multiple ways to rewrite the heat flux q of a layered microstructure under the form (A + B L dcsat/dT) grad(T). One cannot simply directly identify the term B with the diffusion coefficient as done by Andrew Hansen. This analytical solution therefore cannot be received as a proof of the reality of the hand-to-hand mechanism and as an argument against our results.

For clarity, we also want to reiterate our point of view on the macroscopic diffusive vapor flux, as exposed in Fourteau et al. (2021):

- At the microscopic scale only the diffusion of water vapor in the pore space contributes to the mass flux. Contrary to the mechanism advanced in Hansen (2019), the deposition of some molecules and the sublimation of others do not count as a mass transport mechanism, as no mass is transported during such processes (molecules physically stay where they are during phase change). The indistinguishably of water molecules does not help either, as permuting two water molecules does not result in a net mass transport. This is illustrated in Figure 1 of Fourteau et al. (2021).
- For a random heterogeneous structure, the macroscopic vapor flux can be computed as the microscopic vapor flux crossing any given boundary in a REV. Again, only diffusion in the pores count for the vapor flux, but the total mass transport should be normalized by the total surface (including surface in the ice phase) in order to obtain a flux expressed in kg/m^3 per s per m^2 of snow material.
- For periodic structures, the mass fluxes crossing various surfaces cutting the REV might differ from one another (for instance in the layered structure discussed by Andrew Hansen, the vapor flux is maximal in the air and zero in the ice). This means that variability exists at a scale below that of the REV. As in a macroscopic description this sub-REV variability should not appear, and following the work of Pinzer et al. (2012), we compute the average vapor flux crossing the various planes across the entire REV in order to yield a uniquely defined macroscopic vapor flux (as expected when viewing the sample as a homogeneous equivalent medium). In the case of random heterogeneous structures, this average value is equal to the flux crossing any given plane. Finally,

taking the average vapor fluxes crossing the planes of a given microstructure is equivalent to performing a volume average over the entire microstructure (the mass flux being zero in the ice).

The text of comments of Andrew Hansen has been copied below in blue. Our responses to the specific points are given in black, and mostly reiterate the points made in this introduction.

Best Regards,
Kévin Fourteau on behalf of all co-authors.

I believe the authors have a fundamental error in their development of the effective diffusion coefficient for snow found in Eq. (10). The difficulty is that their formulation for D^{eff}_{fast} does not account for the diffusion mechanism of hand-to-hand vapor transport as described by Yosida (1955).

The formulation of the macroscopic vapor flux as the spatial average does indeed not include the hand-to-hand mechanism as proposed by Yosida et al. (1955) and recently supported by Hansen and Folsien (2015) and Hansen (2019). As sublimation and condensation do not transport mass, we consider that this mechanism is nonphysical, and should not be included as a mass transport mechanism.

Controversy abounds regarding the value of the effective diffusion coefficient for snow???a material property essential for studying macroscale heat and mass transfer in a snowpack. Let D_{v-a} represent the binary diffusion coefficient of water vapor in air. In the case of snow, the effective diffusion coefficient is influenced by several competing features of mass diffusion that may either enhance or impede mass diffusion compared to vapor diffusion in humid air only. While there is little debate about some factors affecting vapor diffusion, there are notably differing views on the influence of “hand to hand” diffusion as described by Yosida (1955). To be clear, there is no question as to the existence of hand-to-hand diffusion of water vapor in snow. Rather, the debate centers on the relation of this mass transfer mechanism to the effective diffusion coefficient of snow needed for the governing field equations of the macroscale snowpack.

There is no debate on the notion that in the case of fast sublimation and deposition of water molecules, the net movement of vapor in the pores tends to be from one ice grain to another, in a “hand-to-hand” fashion. There is still ample controversy regarding whether the deposition and sublimation of water molecules count as a proper mass transfer mechanism. As explained above, we argue that since no mass is spatially moved during sublimation and deposition, it does not count as a mass transfer mechanism and only diffusion in the pore space should be considered to compute the total mass flux.

The Supplement to this comment provides a Technical Note that addresses the physical significance of “hand-to-hand” vapor transport as it applies to its influence on the effective vapor diffusion coefficient of a layered ice/humid air microstructure in the presence of a temperature gradient. The layered microstructure shows enhanced water vapor diffusion at all ice volume fractions compared to diffusion in humid air alone (see Eq. 16). Moreover, the hand-to-hand vapor transport mechanism is an important mechanism contributing to this enhanced diffusion. I’ll note the history of this topic dates as far back as de Quervain (1963).

As detailed in Appendix C of Fourteau et al (2021), the way Eq. 16 of the technical document is derived is invalid. The problem lies in the identification of the terms composing Eqs. 13 and 14 to obtain Eq. 16. As the decomposition of q in Eq. 13 under the form (A + B L dcsat/dT) grad(T) is not unique, one cannot identify A and B directly with Keff and Deff in Eq. 14.

We also want to point out that if one accepts that the total heat flux is given by the addition of a pure conduction heat flux and of a latent heat flux equal to F L, where F is the macroscopic vapor flux, then it follows that F is given by the average of the microscopic vapor flux field, as proposed in Fourteau et al. (2021). This is shown by Eqs 6 and 7 of the paper under discussion.

A good deal of the confusion in computing the diffusion coefficient of water vapor in an ice/humid air mixture stems from calculations made using volume averaging techniques for a Representative Volume Element (RVE). The Technical Note shows that, when the hand-to-hand diffusion mechanism is accounted for, volume averaging leads to the known analytical solution for the diffusion coefficient in a layered ice/humid air microstructure. In contrast, if one ignores hand-to-hand diffusion, as the authors have done, volume averaging leads to an inconsistency with the known analytical solution for mass transfer.

As explained above we disagree we the effective diffusion coefficient of the layered microstructure given in the technical document, and it is thus normal that our computation does not yield the same result.

The mass flux, j, is computed as the product of the density times the velocity of a continuum point as it crosses a surface, dS. When the continuum is viewed as a mixture of ice and humid air undergoing diffusion, the density of interest in the mass flux is the density of water vapor in humid air. There is no question as to the value of this parameter. A much more vexing aspect of the mass flux is an understanding of the velocity of the humid air from a macroscale continuum perspective. I will refer to this continuum velocity as the apparent velocity of the water vapor. A lack of a clear interpretation of the apparent velocity of water vapor is the source of some 60 years of confusion surrounding the effective vapor diffusion coefficient in snow. I believe the Supplement clearly illuminates the importance of properly accounting for the macroscale apparent diffusion velocity in a calculation of the effective diffusion coefficient.

Is is true that in the case of a specie diffusing in a homogeneous medium (for instance water vapor diffusing in free air), the diffusion mass flux at a given point can be expressed as the product of the specie density and of the average molecule velocity in the vicinity. Note that this average velocity corresponds to the velocity of the mass center of the nearby molecules, and does not corresponds to the real velocities of the individual molecules. Moreover, during diffusion molecules do not share a common velocity (contrary to advection).

One can extend this to the case of diffusion through a porous medium. It seems that the computations provided as Position A in the technical document perform this task well (although we did not perform independent computations to verrify this point).

The inconsistency of the predicted diffusion coefficient developed by the authors with the analytical solution is enough to invalidate the diffusion perspective they advocate for.

As stated above the derivation of Eq. 16 in the technical document in invalid, and therefore cannot be used to invalidate our results.

However, Fourteau et al. (2020) made some arguments regarding hand-to-hand diffusion on pages 3-4 that deserve some attention. Quoting the authors:

“Our understanding is however that the second part of the mechanism proposed by Yosida et al. (1955) is not physically sound, and that the continuous condensation and sublimation of molecules cannot be used to explain their experimental results. A schematic illustration of the experiment is given in Figure 1, with only two cans for simplicity. The hand-to-hand delivery of water molecules is represented by the orange and red dots, condensing on the lower side and sublimating on the upper side of the ice grain at the interface between the two cans. For this mechanism to explain the experimental observations, the continuous condensation and sublimation should produce a real mass flux from one can to the other, as if the condensing molecule reappeared as the sublimating one. However, what actually happens is that the condensing molecule (represented as an orange dot in Figure 1) remains incorporated at the bottom of the ice grain , thus remaining in the first can. Similarly, the sublimating molecule (represented as a red dot in Figure 1) was already present in the second can. The synchronous sublimation and condensation therefore do not lead to a mass transfer between the two cans. This is different from the molecules traversing the boundary in the air phase (represented as green dots in Figure 1), that actually lead to a mass transfer by depleting the first can in favor of the second one. We therefore argue that the hand-to-hand mechanism, as proposed by Yosida et al. (1955), is not physically sound.”

The above arguments, while technically true in content, have nothing to do with the concept of mass diffusion and the effective diffusion coefficient!

The quoted arguments discuss what processes count as mass transfer mechanisms across a given boundary and are therefore relevant to how the macroscopic vapor flux should be computed.

A study of fundamental continuum mechanics reveals the diffusive flux is a surface phenomenon, defined entirely by mass transfer across a differential surface, dS. Indeed, the mass transfer across a surface is given by j dot n, where n is the unit normal and j is the mass flux. In the context of an ice grain (or ice layer), under a steady temperature gradient, water vapor molecules condense on, say, the lower boundary of ice as they sublimate from the upper boundary, a point easily recognized in the layered microstructure. The authors appear to want to argue that mass diffusion is not enhanced across an ice grain (or ice layer) because there is no net mass transfer in an out of the ice. On the contrary, mass diffusion is occurring, as mass transfer is defined across a surface, any surface, either surface, but not both surfaces above and below the ice.

We argue that there is no mass transfer within the ice phase, and not that “there is no net mass transfer in an out of the ice”.

As explained in the introduction, in the case of some specific periodic microstructures (such as the layered structure), one cannot compute the macroscopic mass flux by computing the mass flux through any surface. It would mean that the same REV could be attributed different effective diffusion coefficients, depending on the chosen surface.

As an analogy, think of steady state heat conduction through an ice layer. Do you wish to argue that there is no heat transfer because the heat entering one side of the ice is countered by the heat leaving the other side of the ice?

Contrary to mass diffusion, there is an actual heat flux within the ice phase as ice is thermal conductor. There is however no diffusive mass flux in the ice phase.

In brief, the heat flux
vector and the mass flux vector are surface phenomena applied to a single differential surface, dS. The authors also wish to make arguments related to mass conservation over a control volume. In particular they note: “Because of mass conservation during the diffusion of water sublimation/condensation process, the apparent flux of vapor skipping the ice phase is compensated by an equal counter-flux of water molecule in the ice phase. Therefore, the mass transfer from one control volume to another is solely governed by the diffusion of water molecules in the air phase (green dots in Figure 1).” The error in logic here is identical to that articulated previously. Mass flux is strictly a surface phenomenon. For instance, if you want to know the mass flux through a layered microstructure, you need only look at the rate of accumulation of mass on the warmer surface of the ice. What is happening on the other side of the ice is irrelevant.

Our arguments precisely discuss what mechanisms count to transfer mass across a given surface (the surface between two control volumes). If one argues that since water molecules are indistinguishable a condensing molecule can be permuted with a sublimating molecule elsewhere so that it seems the condensing water molecules instantly traveled through the ice phase, one needs to be consistent and recognize that the permutation also makes the sublimating molecule instantly travel in the other direction, such that in the end no net mass transport as occurred.

In the case of a layered structure, the mass flux through horizontal planes is not constant throughout the microstructure (it is zero in the ice phase, and quite high in the pore space). There are no valid reasons to assume that only the planes in the pores are relevant. The mass flux should be computed taking into account the whole microstructure, and not simply the zones where is it maximal.
As pointed out by Pinzer et al. (2012), this mistake leads to a largely overestimated mass flux in Colbeck (1993) for instance.

Mechanics also does not care if the medium is ice or air. Apply the authors arguments to the humid air between adjacent ice layers. One layer is sublimating off an ice surface and the same amount of mass is condensing on the next ice surface. By the authors same argument for ice, the mass flux through the humid air would be zero. Of course, this is clearly a physically unreasonable position.

We do not see how one can understand that our arguments lead to zero mass flux in the humid air.

Finally, the authors wish to draw on support from Giddings and LaChapelle who state; "The hand-to-hand transfer does not contribute to the flux because this transfer does not shift water molecules across a plane fixed in the solid network". I’ll respond to this comment with a nuanced, but precise, set of arguments that have already been touched on.

To begin, the governing equations of macroscale heat and mass transfer, see Hansen and Foslien (2015) are based on an Eulerian description of the motion. In an Eularian description, all field variables are defined in terms of their current position and time, i.e., the description focusses on a region in space and not on the path of individual molecules. An example of this description of motion is flow over an air foil. The focus is on the flow at the air foil, not where the air molecules came from. The molecules may be from a kilometer away or a thousand kilometers awayâĂŤit simply does not matter. Nor does it matter whether or not we even know where the molecules came.

The controversy is not a problem of Eulerian versus Lagrangian description. What matters for the macroscopic mass flux is to know if there is a net transfer of matter across a given boundary. It therefore matters that water molecules diffusing in the pores actually cross the boundary (one side is mass-depleted at the expense of the other) while sublimating and depositing molecules does not cross any boundary (while they change phases, they do not leave one side for the other).

Now consider the mass flux vector across a differential surface, dS, at the macroscale. The mass flux vector is the product of the vapor density times the diffusion velocity. The density of water vapor is not in dispute. All that remains is a precise definition of the macroscale continuum diffusion velocity. Focusing on the layered microstructure for the moment, we have two distinct possibilities: A) The motion of water vapor through the ice is zero, and B) the motion of water vapor through the ice is effectively infinite through the source/sink phenomenon. Position A yields a volume averaged diffusion coefficient that contradicts the known analytical solution and, further, contradicts the known surface flux in the humid air. In contrast, position B yields a volume averaged diffusion coefficient that is in perfect agreement with the known analytical solution and the known surface flux. The volume averaged solution is critical as that is what the authors rely on to generate the macroscale diffusion coefficient. Finally, it cannot be overstated that the analytical solution requires no such interpretation of apparent diffusion velocity and represents an independent calculation of macroscale mass diffusion in the layered microstructure.

As explained above the derivation of Eq 16 of the technical document is invalid and therefore does not invalidate position A or our results.

Now, about the experiments of Yosida. In one sense, I would agree with the authors that Yosida’s experimental results are flawed but for entirely different reasons. de Quervain (1963) argued the layered microstructure is an upper bound on mass transfer, a point clearly demonstrated in Hansen (2019). Yosida’s experiments blow through the upper bound of the layered microstructure, thereby invalidating the results.
As an aside, what happens when mass transfer across the lower boundary of an RVE is different from that across the upper boundary. This is when the “mass supply” term in the macroscale governing equation of Hansen and Foslien (2015) kicks in and either net deposition or sublimation may occur within the RVE. That said, it has nothing to do with the surface phenomenon of computing a surface mass flux. Moreover, one can have a huge mass flux across a surface with no change in mass within the RVEâĂŤthe flux in simply equals the flux out.

Yes contrary to the heat diffusion, the vapor flux across various planes of an RVE can vary. This is a microscopic scale variability, that should not be appear at the macroscopic scale. Otherwise, it would mean that a given RVE is not characterized by a single macroscopic flux, and therefore also not by a single effective diffusion coefficient. Averaging over the various planes composing a microstructure removes this microscale variability and yields a uniquely defined mass flux and a uniquely defined effective diffusion coefficient.

I am sure the authors will want to fervently defend their position but their arguments are simply not true. Hand-to-hand mass transfer is an integral part of properly predicting the mass diffusion coefficient in snowâĂŤthe physics of heat and mass transfer demands it, on graphic display in the layered microstructure. It defies logic to acknowledge hand-to-hand diffusion as an important mass transfer mechanism and then neglect this mechanism in the accompanying mathematical development of the diffusion coefficient governing mass transfer at the macroscale.

One final interesting point–the authors are arguing for a value of the diffusion coefficient between 0.8 and 1. On the other hand Hansen (2019) argued for a diffusion coefficient value between 1 and 1.3–they are not that far apart but the physics are vastly different. Lastly, doubters may be tempted to dismiss the Supplement presentation on the grounds that a layered ice/humid air microstructure is not representative of snow. Although the microstructure is very simple in form, it provides amazing insight into the heat and mass transfer in ice/humid air mixtures in general and, remarkably, provides energy flux results that are strikingly similar to those found in very low-density snow. The comparisons with low density snow, supported by interesting physical arguments, have been completed but have been removed from the present Supplement for publication purposes.

We agree that the layered structure is an interesting case that should not be dismissed. However, as written in Appendix C of Fourteau et al. (2021), the proper decomposition of the total heat flux into a pure conduction flux and a latent heat flux yields an effective diffusion coefficient lower than that of the air and consistent with computing the macroscopic mass flux as a spatial average.

I would also like to comment on Figures 8 and 9 of the present work showing aspects of thermal conductivity. In Figure 8, the authors compare an empirical curve they developed against similar (not identical conditions) curves from Calonne et al. (2011) and Riche and Schneebeli (2013). I think a much more interesting comparison is to compare these curves with the theoretical thermal conductivity model of Foslien (1994) where both conduction and heat transfer due diffusion are included.

Figure 1 shows the regression curves for thermal conductivity of snow developed by Riche and Schneebeli (2013), Calonne et al. (2011), and Fourteau et al. (2020b), along with the theoretical model of Foslien (1994). The curves are rather remarkable in that Foslien's model, developed nearly 25 years earlier than the present work, tracks the three regression curves extremely well. In the comparison with Fourteau, the two curves are nearly indistinguishable for ice volume fractions up to 0.2. Additionally, we note that the exact formulation of Foslien is self-consistent, meaning the model predicts the known effective thermal conductivity of ice for an ice volume fraction of one. The empirical finite element solutions do not achieve this consistency. The correlation of Foslien's model with the finite element predictions provides important credibility to the analytical energy flux model of Foslien (1994).

We agree that Folsien (1994) thermal conductivity model provides reasonable thermal conductivity values. We however disagree on how the effective diffusion coefficient is derived from this thermal conductivity formulation.

Our FEM simulations are only performed with rather low density samples, and it is therefore normal that our parametrization does not provide valid results outside of the range of studied samples.

A better point of comparison will be the recent work of Calonne et al. (2019) that performed FEM simulations for the entire range of possible densities. Even though the computations are performed assuming slow surface kinetics, the numerical should remain more than reasonable at high density.

Figure 2 shows the temperature dependence of the effective thermal conductivity as predicted by Foslien's (1994) model. Consistent with results shown in Figure 9 of Fourteau (2020b), the curves in Figure 2 show a flattening of thermal conductivity with increasing temperature. The similarities between Fourteau et al (2020b) and Foslien's 1994 model are striking. Note that the curves generated by Foslien's model are all analytical in nature and are generated from a single equation (Eq. B.2) with no adjustable parameters, demonstrating the veracity of Foslien's theory.

Again, we are fine with the thermal conductivity formulation of Folsien (1994) which on this aspect is perfectly consistent with our paper.

As a final worthy comparison, I would suggest the authors compare results for the contributions of vapor transport to thermal conductivity from Figure 8 of Hansen and Foslien (2015) with the low and high density, fast kinetics results shown in Figure 5 of Fourteau (2020b). Given Foslien's analytical results contain no adjustable coefficients, the accuracy of the vapor contribution to the energy flux is, again, truly remarkable.
I'll close with a bit of a prediction. Foslien's 1994 model of heat and mass transfer is a unified development, meaning the diffusion coefficient and the thermal conductivity are derived from a single energy flux equation. Moreover, present research done by the author shows the layered microstructure closely models the energy flux at low densities and the pore microstructure (de Quervain, 1963; Hansen and Foslien, 2015) closely models snow at very high densities. Foslien's analytical model correctly accounts for the behaviors at either extreme of very low or very high ice volume fractions. Finally, the Foslien model is based on precise formulations of heat and mass transfer applied to simplified microstructures. The results are then cleverly combined in an attempt to emulate snow. My experience is that volume averaging to obtain linear properties is very forgiving when it comes to specific microstructures–in other words, the microstructural representation need not be precise. Substantial research on generating the elastic modulus of composite materials will bear this out, as does the heat transfer work on snow of Christon (1994). For these reasons, I have every reason to believe Foslien's diffusion coefficient model is accurate.

It is not because the thermal conductivity formulation is accurate, that the derivation of the effective diffusion coefficient of the technical document from it is valid.

A first order correction to the authors results that includes hand-to-hand diffusion can be obtained by dividing their diffusion coefficient by the humid air volume fraction. I emphasize that this is not rigorously correct but it is an easy approximation. A very quick glance at Figure 7 of Fourteau (2020b) suggests the corrected data will, indeed, very much align with Foslien (1994).
In a follow-on paper I suggest a numerical modeling path forward to include the influence of hand-to-hand vapor diffusion in snow at all densities, using a precise and novel microscale analysis on RVE's of true snow samples. The authors would be well positioned to pursue these ideas if interested. I wish them well. They clearly have put in a great effort on this subject.

Note: All references in these comments are provided in the Supplement

In conclusion:
The arguments put forward by Andrew Hansen rely on two main errors:

- An invalid derivation of the mass flux in the layered structure by Hansen and Folsien (2015).
- The reliance on the hand-to-hand mechanism that instantly transports water molecules through the ice phase, which is not a real physical phenomenon.

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