

RESPONSE TO QUIRINE KROL

We are thankful to Quirine Krol for her detailed and constructive review of our manuscript.

We in general agree with the insightful remarks of the reviewer and will modify the manuscript accordingly (see our responses to the general and specific comments). There are however a few points where we share a slightly different point of view, and we believe that there are worth discussing before addressing the comments.

Our main goal with this article is to provide the broader snow community with a simple understanding on why :

- The macroscopic vapor flux in snow cannot be larger than in free air
- Surface kinetics plays a determinant role in the macroscopic vapor flux (and should not be overlooked)
- Previous theoretical and modeling studies erred when they reported a macroscopic vapor flux larger than in free air

Accordingly and in an effort of simplicity and clarity, we have emphasized physical reasoning and narrowed our focus to the magnitude of the macroscopic vapor flux, rather than using a proper upscaling method and deriving upscaled equations.

On the other hand, we understand from the review that the reviewer considers that the article could benefit from:

- The use of a proper upscaling method (such as the asymptotic-scale expansion or the volume averaging method).
- A restructuring of the article, with a shorter discussion of the hand-to-hand mechanism
- A more more in details study of non-linear kinetics

Our point of view on these points is that:

- We were not able to derive an upscaled equation of the vapor diffusion equation under arbitrary kinetics. From our understanding this is an active topic of research and methods that apply to snow under arbitrary kinetics are not yet readily available. We are currently (with other colleagues) trying to extend the asymptotic-scale expansion method to arbitrary kinetics in snow, but cannot guarantee when and if it will succeed. Therefore, as proposed in our discussion section we see this study as a preliminary work justifying the need for a rigorously upscaled model under arbitrary kinetics. We will modify Section 3 to better explain that our only goal is to quantify the macroscopic flux, and not to derive upscaled equations.

- We believe that it is necessary to keep a detailed discussion of the hand-to-hand mechanism, as it is a commonly accepted concept in the broader snow community. If the hand-to-hand mechanism were not addressed up front, one could potentially attack the rest of our results by invoking the absence of the hand-to-hand mechanism in our computations.

Similarly a discussion on what is the macroscopic flux and how it relates to microscopic fluxes in the pores is necessary, as it explains most of the discrepancies with previous theoretical studies. We will emphasize that the definition of the macroscopic flux used in this article is physically sound and matches with the definitions used in previously published studies.

- When we introduced the notion of non-linear kinetics, we had in mind to only present a purely illustrative example of the potential impact of non-linear kinetics, and to show that it does not

induce a vapor flux larger than in free air. While we fully agree that an in-depth study of the effects of non-linear kinetics would be more than welcome, we do not think that our article is the right place for it. Indeed, we fear that (i) it will blur our main goals (a clear rebuttal of the hand-to-hand mechanism and of the notion of macroscopically enhanced diffusion in snow) and (ii) at this point we could only offer a superficial look at the matter. A dedicated study, starting on simple structures to decipher the patterns of surface vapor saturation and extending to more complex snow microstructures could be done in the future. One would also need a more detailed treatment of the ice crystals' surface physics (presence of vicinal surfaces, potential non-symmetry of the evaporation and deposition process) to be able to offer quantitative conclusions.

We have copied the comments of the reviewer below in light blue, and provided our answers in black below them. Modifications to the text of the manuscript are proposed in green.

Finally, note that we have discovered two numerical errors in the previous version of the manuscript that we intend to correct in the new version:

- **L343**: The average gradient in the air is 79.00 K/m (and not 77.57 K/m as previously stated).
- **Table 1** and **Fig 7**: The effective diffusion coefficients for the Melt Forms sampled have been underestimated by 33%.

GENERAL COMMENTS :

1 – The chosen upscaling method of ‘volumetric averaging’ over ‘cross-section averaging’ (L120-L132) is based on the argument that microscopic scale variations are not accessible by area averaging. To my understanding this is an issue related to the Representative Element Volume (REV). Snow microstructures are measured with μ CT, large enough such that the volume is representative and homogeneous in a volumetric manner. If cross-sections are used this might not be satisfied anymore as rightly addressed by Pinzer et al. [2012] and volumetric averaging can be chosen. It is therefore not the intrinsically preferred method, but one that is dictated by the specific microstructure.

We agree that computing the macroscopic flux by volume averaging instead of cross-section averaging is related to the specificity of the microstructure. As snow is a random porous medium we believe that cross-section averaging should yield adequate results as long as one select a “large enough” surface to be somehow representative of the overall microstructure. In the case of idealized and periodic microstructure however, the existence of such representative surface is not guaranteed. For instance in a disconnected sphere structure, if one selects an infinitely large cross-section between two planes of spheres, this surface would not be representative and would over-estimate the vapor flux. For this we believe that volume averaging is the safest choice, as it works in situations where cross-section averaging might fail.

We will propose to rewrite the paragraph to indicate that while surface averaging might sometimes be appropriate, volume averaging is a safer choice in general. We will add **L124**:

“Yet, this method of computing the macroscopic vapor flux can be problematic. Indeed, as pointed out by Pinzer et al. (2012) the water vapor fluxes through different horizontal planes of a microstructure are not necessarily all equal. Thus, depending on the chosen plane, the same snow sample could be assigned different macroscopic fluxes, contrary to the notion that the snow sample is homogeneous from the macroscopic point of view. To avoid this issue, the macroscopic flux should therefore be computed as the volume average microscopic vapor flux over the entire representative volume of the microstructure (Shertzer and Adams, 2018), which is equivalent to averaging the fluxes through various horizontal planes (Pinzer et al., 2012).”

2 – The chosen upscaling method is important especially if we couple the effective diffusion to the macroscopic mass and heat transport Calonne et al. [2014]. This study should explicitly relate its results to this study, and how these equations should be adapted.

We do not use a proper upscaling method per se. Contrary to Calonne et al. (2014), we therefore do not attempt to derive the equation governing water vapor at the macroscopic scale.

We will put clearly in the text that we do not upscale to a macroscopic equation and that our goal is only to quantify the macroscopic vapor flux and its dependence on surface kinetics. Therefore, contrary to Calonne et al. (2014) we do not provide a macroscopic vapor equation. We will add a paragraph **L144**:

“Note that the goal of this work is only to quantify the macroscopic water vapor flux in snow and its associated effective diffusion coefficient. Contrary to Calonne et al. (2014) we do not attempt to derive the macroscopic equations governing water vapor at the layer scale.”

We also propose to rename Section 4 of the article from “**Quantifying the macroscopic vapor flux in snow**” to “**Bounding the effective diffusion coefficient of water vapor in snow**”

The asymptotic scale derivation of Calonne et al (2014) assumes a small influence of vapor sinks and sources at the ice/pore interface. There is no guarantee that their upscaled equation applies for large α , when sublimation/deposition effect become significant. As a matter of fact, the macroscopic vapor flux that we derive for large α does not correspond to the upscaled model of Calonne et al (2014), as they predict that the vapor flux should be the same as in an inert medium.

3 – The accommodation coefficient, including its name, should be introduced in the introduction including experimental observations such as Libbrecht [2005], Harrington et al. [2019] and possibly other studies. The choice of values for the simulations should be linked and/or motivated by deficiencies of these studies.

We will rename α the sticking coefficient for the entire article.

We will discuss the potential influence of kinetics in the introduction. However, we do not think the sticking coefficient should be explicitly discussed in the introduction. Indeed, experimental measurement of α are mainly limited to the problem of deposition of facets, and it is not a given that such measurements apply for the entirety of ice surfaces in snow, which includes sublimation and non-faceted surfaces. Moreover, this is in line with our methodology to cover a broad range of sticking coefficient values.

We will rewrite the paragraph starting **L18**:

“The physics at play in the pores is generally agreed upon, even though questions about the precise kinetics of the sublimation and deposition of water molecules onto ice surfaces in snow remains open (Legagneux and Domine, 2005, Pinzer et al., 2012, Calonne et al., 2014, Krol and Löwe, 2016). However, even for investigators assuming the same physics at the microscopic scale, the transition from the microscopic to the macroscopic scale remains a point of contention in the snow community (Giddings and Lachapelle 1962, Colbeck 1993, Pinzer et al., 2012, Hansen and Folsien, 2015, Shertzer and Adams 2018).”

We will also rewrite the end of the introduction to better introduce our work on the accommodation coefficient, starting from **L39**:

“The aim of this paper is to clarify the origin of these discrepancies and to quantify the macroscopic vapor flux based on theoretical and numerical modeling. As the kinetics of sublimation and deposition of water molecules on the ice surfaces in snow is not well constrained, we decided to

explore a broad range of possible kinetics in our study. We start by considering in Section 2 whether the hand-to-hand mechanism, as originally proposed by Yosida et al. (1955), can indeed explain the large macroscopic vapor fluxes observed in snow. Then in Section 3, we recall how the macroscopic vapor flux can be obtained from the microscopic vapor flux occurring at the pore scale. In Section 4 we present theoretical work to bound the macroscopic vapor flux in snow, by treating two limiting cases of surface kinetics.”

4 – Although symbols in equations are generally well described and it is clear from the context what they mean, it might be helpful to the reader to introduce systematic notation to distinguish between upscaled quantities and local quantities, e.g.

$$F = \frac{1}{V} \int_V f dx^3$$

in other words, how are F and C related to their microscopic quantities?

We will add in the text **L132** the equation giving the macroscopic water vapor flux from the microscopic water vapor flux.

“Again, the averaging needs to be performed over the total volume, including the ice space, and the macroscopic vapor flux F is thus given by

$$F = \frac{1}{V} \int_{V_a} f dV$$

where V and V_a respectively represent the total volume of the snow sample, and the pore volume.”

5 – In case of volume averaging, gradients of microscopic fluxes are influenced by sources and sinks at internal ice-air interfaces Whitaker [1998], Krol and Löwe [2018], i.e.

$$\langle \nabla f \rangle = \nabla \langle f \rangle + \int f d\mathbf{n}$$

In case of the idealized spheres the second term vanishes because of symmetry, but for your snow samples it might not be the case, and should be shown, either by estimating the order of magnitude of the gradient of your sources and sinks, or by analysis of the simulations that this term is rightfully neglected. Here it matters how the macroscopic quantities are related to their microscopic counterparts. Note that in your simulations you average over both phases, vapor and ice, but you neglect the sinks and sources. I believe with these microstructures it is probably alright, but it should be estimated/shown that you can do so.

We do not use the spatial averaging theorem (SAT) in our treatment, and directly compute the left-hand side of the equation, integrating the microscopic vapor fluxes over the microstructure. The effects of sinks and sources at the ice/pore interface are taken into account directly at the microscopic scale, through Robin or Dirichlet boundary conditions.

We will however include a new appendix using the SAT to demonstrate that the macroscopic flux is maximal in the infinitely fast kinetics case. This new appendix is attached at the end of this document.

SPECIFIC COMMENTS:

110: Naming of the coefficient α . This coefficient is often related to the phase-change it represents i.e. deposition, sublimation, or sticking parameter.

We will use replace “accommodation coefficient” with “sticking coefficient” throughout the manuscript.

111: There is no evidence or discussion in the paper that suggests that convection is one of the candidates responsible for the experimentally observed mass deficiency.

We will remove the mention of convection from the abstract, and rewrite the discussion **L437**: “Indeed, the importance of convective mass transport in subarctic snowpacks has notably been pointed out by Trabant and Benson (1972) and Sturm and Johnson (1991), and thus appears as a good candidate to explain the high vapor movement in subarctic snowpacks.”

146: Suggestion to shorten this paragraph and move to the discussion. The notion of hand-to-hand diffusion should be discarded on the fact this is simply no physical transport of water molecules.

We think it is important to keep this full paragraph. Indeed, the notion of water molecules short-cutting the ice space is commonly held in the snow science community, and we think a detailed rebuttal is welcome. As a concrete example, when we initiated this study we thought that the hand to hand mechanism as described by Yosida et al. (1955) was a valid mechanism.

1120-132: Please be very specific about your methods of upscaling. See general comments 1, 2, and 5.

As mentioned in the general comments 2 and 5, we do not use any method of upscaling and simply directly compute the volume average of the microscopic vapor flux. Because of it, we cannot provide an upscaled equation. We instead rely on a phenomenological approach.

We however added precision that in our numerical and theoretical computations we assumed that the macroscopic gradient of water vapor equals the macroscopic gradient of saturated vapor.

For this we added **L103**:

“Let us consider a volume of snow (Figure 2a), subjected to vertical macroscopic temperature and vapor gradients at its boundaries. For this study we consider that the macroscopic water vapor gradient equals the macroscopic gradient of saturated vapor, and is therefore driven by the macroscopic temperature gradient (as in Yosida et al.,1955, Colbeck, 1993, Sokratov and Maneo, 2000, Pinzer et al., 2012).”

and **L141**:

“However, one should keep in mind that the effective diffusion coefficients computed in this work might depend on the applied vapor and thermal gradients, and are therefore not necessarily intrinsic. Moreover the proposed numerical values may also not apply in the case where the macroscopic concentration gradient is decoupled from the macroscopic thermal gradient.”

We will also add a discussion on this assumption **L435**:

“Furthermore, we assumed in this study that the macroscopic water vapor gradient is equal to the macroscopic gradient of saturated vapor, driven by the macroscopic thermal gradient. This assumption has been regularly made in the snow science community (Yosida et al.,1955, Colbeck, 1993, Sokratov and Maneo, 2000, Pinzer et al., 2012), and is supported by the idea that the ice in

the snowpack tends to impose water vapor saturation at the macroscopic scale. It however remains possible that the macroscopic water concentration deviates from saturation, notably if the deposition and sublimation kinetics is slow. A rigorous upscaling method yielding the equations governing macroscopic water concentration would therefore also help quantifying if such a situation of non-saturation at the macroscopic scale is likely to occurs in real snowpacks, and indicate how the macroscopic vapor flux should be computed in such a case.”

L133: Here I would expect a mathematical definition, including upscaling methods, see comment 5.

As explained above, we directly compute the macroscopic vapor flux as the volume average of the microscopic vapor flux (and justify this choice with physical consideration). We will add reference to previously published study of Schertzer and Adams (2018) that uses the same formula for the computation for the macroscopic flux. We will write the equation relating the macroscopic water vapor to the microscopic one (see our proposed text modification in the response to the general comment 4).

L137: Semantic comment: What does ‘ideally’ mean in this context? Maybe include that intrinsic, in this context, means that D_{eff} is independent of the external temperature gradient. When D_{eff} is dependent on the external gradient, one could say that the response of the material is non-linear. Does this break the definition of the effective diffusion coefficient, meaning the coefficient that quantifies the vapor flux as a linear response to an applied concentration gradient?

In general, in the snow community, when one uses an effective diffusion coefficient one expects it to be independent of the applied concentration gradient and thus to be a true proportionality constant between flux and gradient. While standard upscaling techniques such as the asymptotic-scale expansion or the volume averaging method prove that the obtained diffusion coefficients are independent of the applied gradient (under certain conditions of course), our phenomenological approach does not prove it. We therefore want to stress that what we defined as the effective diffusion coefficient might not be intrinsic (even-though it appears to be the case with linear kinetics based on our simulations).

In our article, we call the ratio of the flux over the gradient the effective diffusion coefficient, even in the case where it is not constant with respect to the gradient. While we understand that some might prefer to use a different term in this case, we choose this approach as it allow us to easily compare it to the diffusion coefficient in air and is easily understood by the broader snow science community. We will rephrase **L137** with:

“In the snow science community the effective diffusion coefficient D_{eff} is usually expected to be independent of the applied thermal and vapor gradients (e.g. Yosida et al., 1955, Colbeck, 1993). In this case, it is possible to treat the problem of macroscopic vapor transport in snow with a generalized Fick's law, where D_{eff} is independent of the applied boundary conditions and only depends on the snow microstructure. Such an effective diffusion coefficient does not depend on the external conditions, and is then said to be intrinsic (Auriault et al., 2010). However, one should keep in mind that the effective diffusion coefficients computed in this work might depend on the applied vapor and thermal gradients, and are therefore not necessarily intrinsic.”

L150: In this paragraph I suspect at least an expression for the the macroscopic vapor flux as suggested by the title.

We will explain in the text that solving the microscopic equations yield the microscopic water vapor fluxes and that volume averaging the microscopic fluxes yields the macroscopic water vapor flux: “Solving Equation 2 we obtain the microscopic vapor fluxes inside the whole microstructure. Using Equation 1 then yields the water vapor flux at the macroscopic scale F .”

L165 and 308: How infinite can αv_{kin} be? v_{kin} is finite $\sim 10^2$, and $0 < \alpha < 1$. In principle it should be compared to the actual interface velocity v_n in the Robin b.c. as stated in Kaempfer and Plapp [2009]. A discussion on α and its values would be appreciated Libbrecht [2005], Saito [1996], Legagneux and Dominé [2005].

The notion of infinitely fast kinetics is purely theoretical, and should be seen as a limiting case. We use this notion to treat the case where the kinetics is fast enough to impose vapor saturation at the interface. In practice, αv_{kin} cannot indeed be greater than v_{kin} . We will clarify in the text, that treating αv_{kin} as infinite is a simplifying assumption. The validity of this assumption to model macroscopic vapor diffusion in snow is treated in the discussion, and remains an open question. We will add **L165**:

“While the infinitely fast kinetics case is strictly theoretical, as αv_{kin} is less than or equal to v_{kin} , it helps apprehending the macroscopic vapor flux when surfaces kinetics processes are much faster than diffusion in the air space”

To know if the infinitely fast kinetics is appropriate for macroscopic flux modeling, we believe that one should compute the value of the second Damkohler number (defined as $Da = \alpha * v_{\text{kin}} * l / D_0$), where l is a length characterizing the microscopic scale. Such an approach is notably used in the diffusion-reaction community (e.g. Munnichi and Icardi, 2020 and Bourbatache et al., 2020). This number essentially compares the characteristic times of the surface processes and of diffusion in the air space, and characterize how the oversaturation at the ice/pore interface compares to the concentration gradient in the pores.

We do not believe that the interface velocity in itself is a good quantify to differentiate the fast and slow kinetics regime of the macroscopic vapor flux. Indeed, increasing the thermal gradient at constant α increases the interface velocity, but do not necessarily modify the effective diffusion coefficient, as seen in our Figures 4 and 6 for example.

L263: This paragraph includes an important realization, how does it relate to the expression for the macroscopic heat transport provided by Calonne et al. [2014]. This could be treated in the discussion.

The study of Calonne et al. (2014) is based on the assumption of slow kinetics. On the other hand, Hansen and Folsien (2015) assume that water vapor is constantly saturated at the microscopic scale, which corresponds to infinitely fast surface kinetics. Therefore, while Calonne et al. (2014) find that heat conduction in snow occurs similarly as in an inert medium, Hansen and Folsien (2015) find that water vapor becomes an integral part of heat transport with fast kinetics.

The degree of coupling between heat transfer depends on the kinetics of sublimation and deposition. We are currently working on a manuscript investigating the different behavior between very slow and very fast kinetics (thus not investigating the intermediate cases and the transition between slow to fast kinetics). Our preliminary results are in line with the idea that with very fast kinetics, vapor transport becomes an integral part of heat transfer (as proposed by Yosida et al., 1955 or Moyne et al., 1988) and appears in the effective thermal conductivity of snow.

We will add in the text **L264** that:

“In their model, water vapor is at constant saturation in the pores (thus corresponding to the case of infinitely fast kinetics), and acts as an integral part of heat transfer by transporting latent heat between sublimation and deposition surfaces (as notably proposed by Yosida et al., 1955)”

L282: Some more details on the technicalities of the simulation should be provided, are T and c computed simultaneously? or is c computed given T ? How is it parallelized, and how long does it take? What are the meshing requirements, how many points etc.

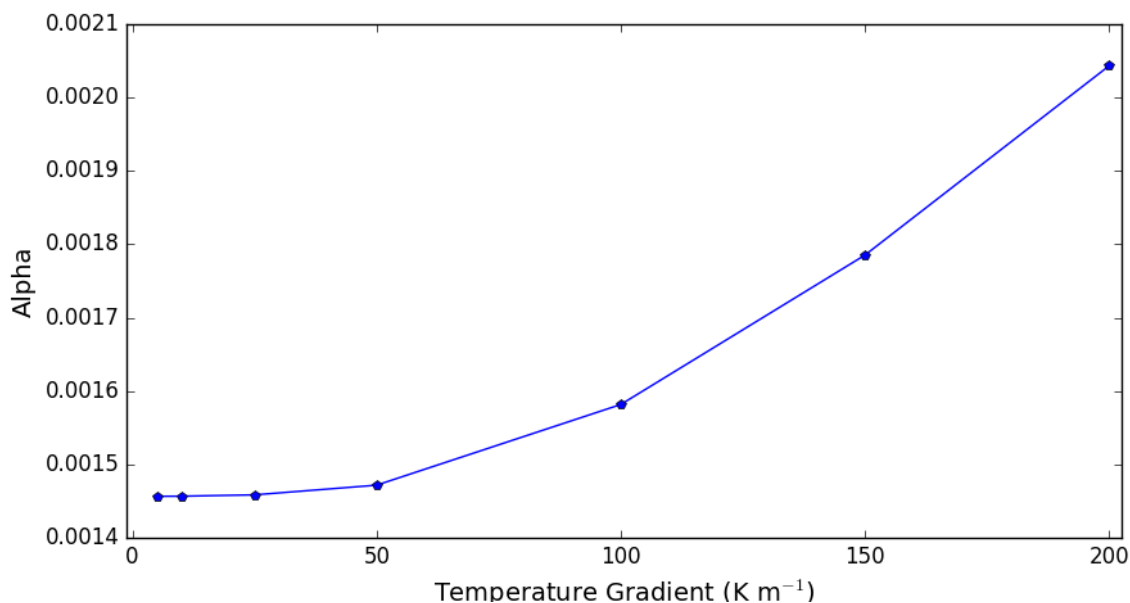
We will extend the paragraph **L330** to add more details on how the simulations are performed.

“The heat and diffusion equations are solved using the finite element method with the open-source software ElmerFEM (Malinen and Raback, 2013). We use the readily available ElmerFEM modules dedicated to the heat and diffusion equations, which are solved with iterative methods. We first solve the steady-state heat equation in order to obtain the temperature field in the entire microstructure. The steady-state vapor diffusion equation is then solved using the saturation concentration at the ice/pore interface resulting from the previously computed temperature field. In the case of simulations performed on measured snow microstructures, the tetrahedral meshes have been derived from Xray computed microtomography images using the CGAL meshing library. The meshes have been refined to capture the ice/pore interface, and contains between 18 and 50 millions elements, depending on the snow sample. Moreover, in the case of snow samples the meshes have been partitioned into 20 sub-meshes and the computation are performed using the parallel computing abilities of ElmerFEM. Under such conditions, a simulation typically takes a bit less than an hour to run. Finally, the outputs of the simulations are processed using the ParaView software to compute the volume averages.”

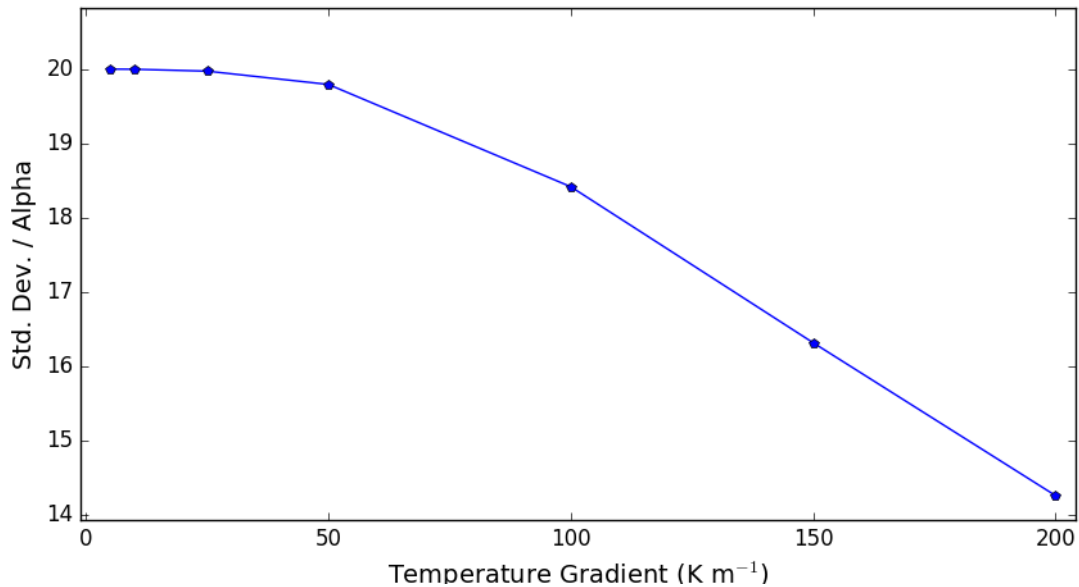
L367: For the non-linear kinetics results it might be useful to state the surface averaged simulated α and its variance.

We computed the surface averaged α as well as the associated standard deviation, for the DF snow sample. We find that the average value increases with temperature gradient, from 0.00145 at 5 K/m to 0.00204 at 200 K/m. Note that these value do not align with the constant α yielding equivalent macroscopic vapor fluxes.

Moreover, the variance remains high in all cases with a value around 0.029. Expressed relatively to the average value, the relative standard deviation drops from 20 at 5 K/m to 14 at 200 K/m. This high variability is consistent with observations that values of α cover almost the entire 0 to 1 interval within the microstructure.



We have included below two Figures showing α and the relative standard deviation as a function of thermal gradient.



Our understanding is that these results indicate that in the case of non-linear kinetics, the macroscopic flux is driven by very localized effects, not easily accessible through average quantities. Clearly this is an interesting topic of research, but as explained in the introduction of our response we believe that a dedicated and more thorough study would be more appropriate to discuss these points in details. Moreover, as we are not confident in the validity of the non-linear law chosen in this article, we should be careful not to over interpret these preliminary results.

L367: How sensitive is your result to the value σ_0 ? Since it might differ for different crystallographic surfaces.

We did not performed simulations with varying σ_0 parameters. Our understanding is that increasing σ_0 should shift the green curves of Figure 4 and 6 towards the right, as greater saturations, and thus greater macroscopic concentration gradients are needed to have high α .

However, as we are not sure that the used formulation of α as a function of saturation applies for the entirety of ice surfaces in snow, our results on non-linear kinetics should only be viewed as quantitative at this point.

Fig 4: I suggest to split this plot into two figures. One for linear simulations $D_{\text{eff}}^{\text{norm}}$ vs α and the other for non-linear dynamics $D_{\text{eff}}^{\text{norm}}$ vs ∇T including colorbar for surface averaged α . This suggestion is given to observe the type of transition between purely tortuous diffusion and phase transition enhanced diffusion. The data on the non-linear dynamics seems to rapidly depart from the tortuous diffusion case: is there a reason for this? We would expect also here a smoother transition between the two limiting cases, such as in Fig.6. The results for small temperature gradients puzzle me. A discussion on the results in this regime might be helpful.

We do not think our results for non-linear kinetics should be further interpreted in this paper. As explained before, we are not confident that the chosen law applies for the entirety of the snow sample.

We do not know why there is a faster transition from the tortuous diffusion to the phase transition enhanced diffusion for the idealized microstructure than for the measured microstructure. To answer this question a potential method would be to compare the results obtained on various simple

idealized microstructure, to decipher the influence of porosity and tortuosity, and quantify where the zones of deposition and sublimation appear in the microstructure.

L402: Moreover? Is there a reason not to compute the non-linear cases? In my opinion it is interesting and worth it to quantify the different non-linear responses of the 6 different snow types.

As we are not sure that the chosen formulation for non-linear kinetics applies to snow, our goal is only to provide an illustration of the effect of non-linear kinetics. We illustrate that it does not produce greater than in air vapor fluxes, but cannot produce more quantitative results. While it is certainly an interesting topic, it is out of our scope for now and we are not able to reach robust conclusions concerning the effects of non-linear kinetics. We will add to the text L355:

“[...] and might not properly apply for the entirety of ice surfaces in snowpacks. Indeed, this law has been derived using deposition measurement, and might not apply for sublimation surfaces (Beckmann and Lacmann, 1982). Moreover, we cannot rule out the presence of vicinal surfaces in the snowpack, where the proposed law does not apply (Legagneux and Domine, 2005). Therefore, the point of using such a law is to qualitatively study the potential impact of a dependence of α to the local vapor saturation, rather than to produce quantitative results.”

We will also explain why we chose not to perform non-linear kinetics simulations on all our sample set:

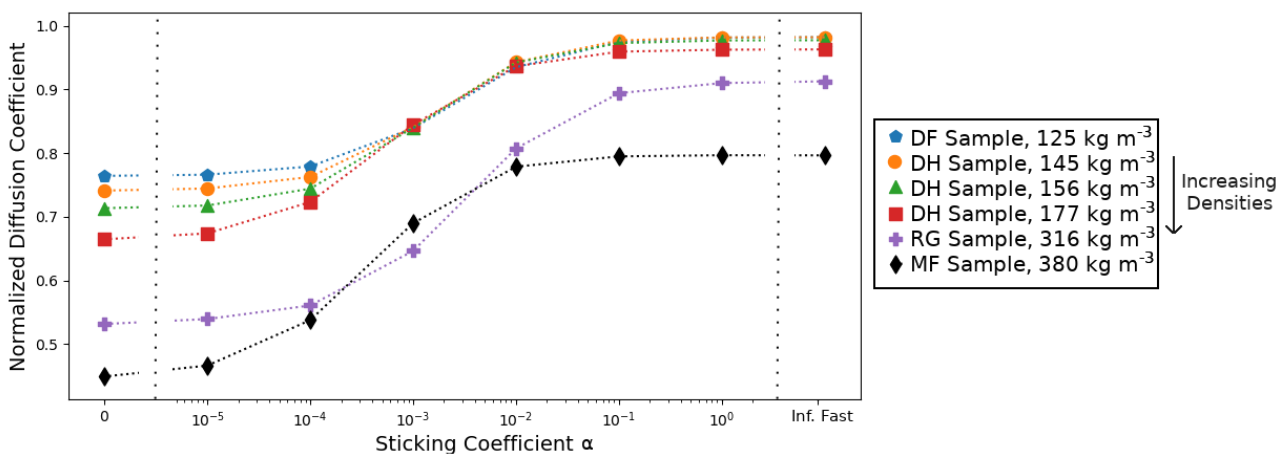
“We also did not compute $D_{\text{eff}}^{\text{norm}}$ with non-linear surface kinetics (i.e. when α is not constant), as we are not confident in the validity of the chosen non-linear law for snow modeling.”

Fig 7 : The Figure and Table have approximately the same information. Consider plotting again D_{eff} vs α and colorbar on density. Alternative, plot D_{eff}/φ and discuss the remaining influence of SSA. If SSA is presented in either a table or a plot, then a note in the discussion on its influence is desirable.

Our reasoning behind putting both the Figure and the Table is that the Figure helps apprehending the influence of density and α , while the Table provides the exact data point.

We have redone the Figure to plot D_{eff} as a function of α (inserted below in our response). This new figure clearly highlights that there are two regimes for the effective diffusion coefficient, with a transition for constant α coefficients around $1e-3$.

We will rewrite the sentence L404 to:



“Figure 7 highlights that the normalized effective diffusion coefficient exhibits two different regimes depending on the value of α . The transition between the fast and slow surface kinetics regimes occurs for values of α around 10^{-3} .”

It is hard to decipher the potential influence of SSA, as in our sample set SSA is well correlated with density. We will emphasize this point in the manuscript **L405**:

“We observe that the effective diffusion coefficient is well correlated with density, and show an almost systematic decrease of $D_{\text{eff}}^{\text{norm}}$ with increasing density, for all values of α . The correlation between $D_{\text{eff}}^{\text{norm}}$ and the specific surface area is not so well marked, notably for the RG sample that shows a large value of specific surface area without any clear impact on $D_{\text{eff}}^{\text{norm}}$. That being said, our sample set is only composed of six samples and for which density and specific surface area are correlated. A detailed study of the influence of microstructural parameters on the effective diffusion coefficient would require a larger sample set, notably to be able to decipher the independent influence of specific surface area and density.”

L440: A list of the general causes to why vapor flux was considered to be enhanced in the past is expected in the discussion.

We will add a new sentence at the end of the first paragraph of the discussion **L411**:

“We have shown that most of the previous theoretical studies reporting macroscopic vapor flux enhanced compared to free air used faulty computations of the macroscopic vapor flux, which resulted in systematic overestimation.”

L400: A reasonable explanation for why convection could be the cause of the experimentally observed mass deficit could go here.

We will reformulate the discussion on convection to **L437**:

“Indeed, the importance of convective mass transport in subarctic snowpacks has notably been pointed out by Trabant and Benson (1972) and Sturm and Johnson (1991), and thus appears as a good candidate to explain the high vapor movement in subarctic snowpacks.”

L448: ‘Disagree’, is an understatement. You show with numerical simulation that this concept is ill-defined. Suggestion: We show with numerical simulations that increased vapor flux by the hand-to-hand mechanism is not present.

We do not think our simulations can be used to show that the concept of hand-to-hand diffusion is ill-defined, as they by default do not include the hand-to-hand mechanism. As stated in the introduction to the review we believe that Section 2 and specific physical reasoning is necessary to refute the hand-to-hand mechanism.

L458: Avoid ‘intuitive’. Suggestion: consistent with actual water vapor transport.

We will rephrase to “We argue that the method used in this article, i.e. volume averaging over an entire microstructure including the ice, is the only one consistent with the actual nature of the macroscopic water vapor flux.”

L492: incorrect use of ‘inferior’, use ‘less than’.

We will systematically replace “superior” by “greater than” and “inferior” by “less than”.

TECHNICAL CORRECTIONS:

L59: The use of pore phase, throughout the manuscript is incorrect. Please use pore space, or vapor/gas phase. Also air phase is not commonly used.

We will rewrite the manuscript using the terms “pore space” and “gas phase”.

Overall The use of colons is not consistent, e.g. before equations introduced by ‘given by’ it is not very common to use them. Use of colons is generally restricted to lists or ‘may’ be used between independent clauses when the second sentence explains, illustrates, paraphrases, or expands on the first sentence. Equations are part of sentences and therefore colons should not appear more often before an equation than in other parts of your text.

We will remove the colon before the introduction of the equations

1.296 Outer brackets in the exponent should be larger, (use `\left(` and `\right)` commands).

We will rewrite the equation with larger brackets.

1.304 Condensation is reserved for the gas-liquid phase-transition, use deposition (or desublimation) also at other places throughout the manuscript.

We will replace condensation with deposition throughout the manuscript.

1.336 Goes → go.

We will correct the typo.

1.437 ? citation missing.

Yes a citation referring to to Sturm and Benson 1997 was missing. We will fix it in the new manuscript.

1.454 Similar → Equivalent.

We will rephrase as proposed.

Demonstration that the macroscopic vapor flux is maximal under infinitely fast kinetics

The aim of this appendix is to demonstrate that the macroscopic vapor flux is maximal in the case of infinitely fast kinetics. For this we start by applying the spatial averaging theorem whitaker1999method to the vapor concentration in the pores c

$$\langle \nabla c \rangle = \nabla \langle c \rangle + \frac{1}{V} \int_{\Gamma} c \mathbf{n} dS \quad (1)$$

where $\langle \bullet \rangle$ is an operator defined as $\langle \bullet \rangle = \frac{1}{V} \int_{V_a} \bullet dV$, and the concentration c in the surface integral is the vapor concentration at the ice/pore interface. Multiplying by D_0 , and using the notation introduced in this article for the macroscopic vapor flux \mathbf{F} , we have

$$\mathbf{F} = -D_0 \nabla \langle c \rangle - \frac{D_0}{V} \int_{\Gamma} c \mathbf{n} dS \quad (2)$$

Moreover, using the Hertz-Knudsen equation we have that the concentration at the interface is

$$c = c_{\text{sat}} - \frac{D_0}{\alpha v_{\text{kin}}} \nabla c \cdot \mathbf{n} \quad (3)$$

Equation 2 can thus be written as

$$\mathbf{F} = -D_0 \nabla \langle c \rangle - \frac{D_0}{V} \int_{\Gamma} c_{\text{sat}} \mathbf{n} dS + \frac{D_0^2}{V \alpha v_{\text{kin}}} \int_{\Gamma} (\nabla c \cdot \mathbf{n}) \mathbf{n} dS \quad (4)$$

Applying the same spatial averaging theorem to the saturation concentration c_{sat} , we have

$$\frac{1}{V} \int_{\Gamma} c_{\text{sat}} \mathbf{n} dS = \langle \nabla c_{\text{sat}} \rangle - \nabla \langle c_{\text{sat}} \rangle \quad (5)$$

Injecting Equation 5 in Equation 4 thus yields

$$\mathbf{F} = -D_0 \nabla \langle c \rangle - D_0 \langle \nabla c_{\text{sat}} \rangle + D_0 \nabla \langle c_{\text{sat}} \rangle + \frac{D_0^2}{V \alpha v_{\text{kin}}} \int_{\Gamma} (\nabla c \cdot \mathbf{n}) \mathbf{n} dS \quad (6)$$

As we assume that the macroscopic vapor concentration equals the macroscopic saturation concentration gradient (as in [5, 1, 3, 2]), we have that $\nabla \langle c \rangle = \nabla \langle c_{\text{sat}} \rangle$. Thus

$$\mathbf{F} = -D_0 \langle \nabla c_{\text{sat}} \rangle + \frac{D_0^2}{V \alpha v_{\text{kin}}} \int_{\Gamma} (\nabla c \cdot \mathbf{n}) \mathbf{n} dS \quad (7)$$

Let us now assume, without loss of generality, that the macroscopic vapor and thermal gradients are orientated downward. As seen in Figure 1, surfaces that are characterized by a normal vector pointing upward are deposition surfaces. The product $\nabla c \cdot \mathbf{n}$ is therefore negative, and $(\nabla c \cdot \mathbf{n}) \mathbf{n}$ is a vector pointing downward. Similarly, surfaces that are characterized by a normal vector pointing downward are sublimation surfaces. The product $\nabla c \cdot \mathbf{n}$ is thus positive, and

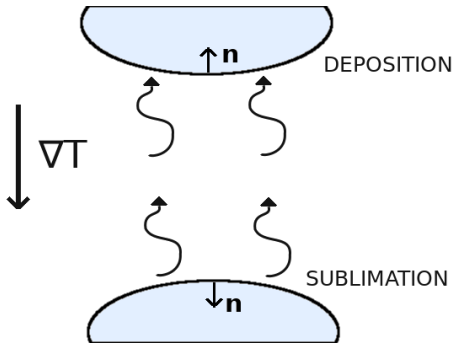


Figure 1: Schematic showing the normal vector \mathbf{n} of deposition and sublimation surfaces. Ice crystals are represented in blue, and the thermal and vapor gradients are assumed to point downward.

the vector $(\nabla c \cdot \mathbf{n}) \mathbf{n}$ is pointing downward. Therefore, for both type of surfaces $(\nabla c \cdot \mathbf{n}) \mathbf{n}$ is pointing downward. The surface integral term in Equation 7 thus acts in opposition of $-\langle D_0 \nabla c_{\text{sat}} \rangle$, and tends to reduce the macroscopic vapor flux. We thus have the inequality

$$|\mathbf{F}| \leq |\langle D_0 \nabla c_{\text{sat}} \rangle| \quad (8)$$

We will now show that this upper bound is reached in the infinitely fast kinetics case. Indeed, under the infinitely fast kinetics hypothesis the product αv_{kin} can be treated as going to infinity. At the same time, the surface integral of Equation 7 remains bounded, as the concentration gradient in the vicinity of the interface does not diverge. The surface integral thus vanishes, and the norm of the vapor flux is given by

$$|\mathbf{F}| = |\langle D_0 \nabla c_{\text{sat}} \rangle| \quad (9)$$

that is to say that the upper bound of the macroscopic vapor flux is reached under infinitely fast kinetics. Moreover, note that we re-derived that in the infinitely fast kinetics case, the macroscopic vapor flux is given by the spatial average of the saturation vapor concentration in the pore space.

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