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Hansen's comments on the authors' response to the Technical Note titled:

*A demonstration of the existence and importance of "hand-to-hand" vapor transport
as it relates to the effective vapor diffusion coefficient in low-density snow*

Symbols

D	diffusion coefficient
k	thermal conductivity
q	energy flux
u_{sg}	latent heat of sublimation of ice
v_v	water vapor diffusion velocity
x	macroscale coordinate
ξ	microscale coordinate
γ_v	density of vapor component
θ	absolute temperature
ϕ_α	volume fraction of constituent α

Superscript

cond	conduction
eff	effective

Subscript

i	ice constituent
ha	humid air constituent
v	vapor component within humid air
v-a	water vapor in air

The authors' defense to the entire Technical Note of Hansen is summarized in their concluding remarks given below.

"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. "

Let us dispense with the second comment first as it requires only a brief response. The authors wish to argue that the instant transport of water vapor through the ice is non-physical, yet, they do not dispute that hand-to-hand diffusion is a real mechanism associated with mass transfer.

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At the risk of repetitive arguments, mass transfer in a continuum is based on water vapor moving across a surface. The surface does not care where the molecules of water came from, either around an ice grain or through a hand-to-hand mechanism. All the surface knows is that vapor is moving across it. 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 mass transfer at the macroscale.

Now let us address in detail the first comment which is really the sole basis for their arguments of rejecting the conclusion of the Technical Note. The authors assert that I have produced an *invalid* derivation of the mass flux and that they have corrected the derivation of mass flux in Appendix C of Fourteau (2021). Their claim that my derivation is "invalid" is patently false. In what follows, I clearly lay out the differences in the approach I put forth as well as that of the authors. The results are fascinating and, further, clearly show the approach of Hansen leads to a far more appealing and correct final conclusion. I also identify where the authors' formulation has gone awry.

We address the competing views articulated in the Technical Note and the authors response to the note by studying mass transfer in a layered ice/humid air microstructure in the presence of a temperature gradient as shown in Figure 1. The layered microstructure is an ideal test case of the two approaches in that an analytical solution exists—*a solution based only on one-dimensional heat and mass transfer principles with a long history of supporting development.*
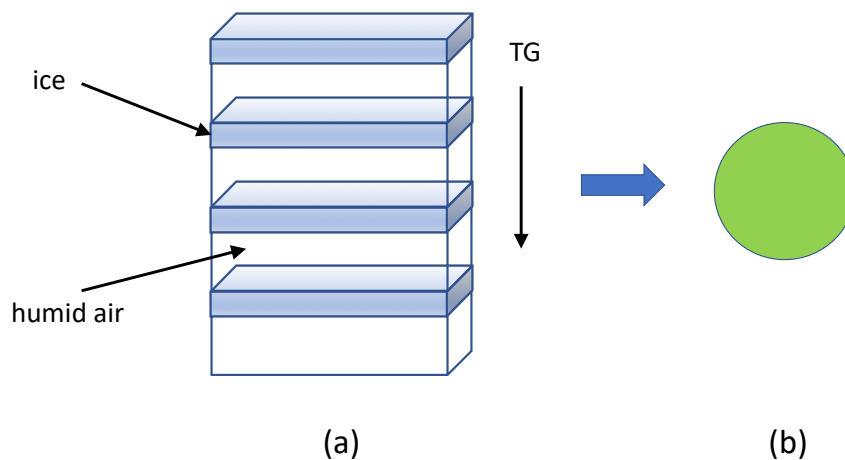

Figure 1. (a) Layered microstructure of ice and humid air in the presence of a vertical temperature gradient. (b) The homogenized continuum point possessing macroscale continuum properties representative of the ice/humid air mixture.

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We begin by making a statement that both the approach of Hansen and the authors agree on. The energy flux of the homogenized continuum is

$$q = -\left(\frac{k_i\left(k_{ha} + D_{v-a}\ u_{sg}\frac{d\gamma_v}{d\theta}\right)}{\phi_i\left(k_{ha} + u_{sg}D_{v-a}\left(\frac{d\,\gamma_v}{d\,\theta}\right)\right) + \phi_{ha}k_i} \right)\frac{\partial\theta}{\partial x} \tag{1}$$

A challenge is met immediately in that the above equation does not permit a clean decomposition into terms involving only thermal conductivity and only diffusion. Hansen and Foslien (2015) introduced definitions of thermal conductivity and the diffusion coefficient given by

$$k = \left(\frac{k_i k_{ha}}{\phi_i\left(k_{ha} + u_{sg}D_{v-a}\left(\frac{d\,\gamma_v}{d\,\theta}\right)\right) + \phi_{ha}k_i} \right), \tag{2}$$

and

$$D = \left(\frac{k_i D_{v-a}}{\phi_i\left(k_{ha} + u_{sg}D_{v-a}\left(\frac{d\,\gamma_v}{d\,\theta}\right)\right) + \phi_{ha}k_i} \right). \tag{3}$$

This approach essentially involves splitting the numerator of Eq. (1) into two terms and identifying the diffusion coefficient with the term involving u_{sg} in the numerator. Quoting Fourteau (2021):

"We however argue that Eq. (C.4) (Eq.1 above) is only one way among many to rewrite q_{lam} under the form $A\nabla T + L\,D_o\,\frac{dc_{sat}}{dT}\,B\nabla T$ and thus that the identification of the latent-heat flux with the second term of the decomposition is arbitrary."

Fourteau et al. (2021) then go on to propose their own decomposition of the thermal conductivity and diffusion coefficient. Let us follow both paths and distil the debate down to just a few basic equations, affording simple comparisons and concrete conclusions.

To begin, let us start with Equation (1) which is not in dispute and repeated below as:

$$q = -\left(\frac{k_i\left(k_{ha} + D_{v-a}\ u_{sg}\frac{d\gamma_v}{d\theta}\right)}{\phi_i\left(k_{ha} + u_{sg}D_{v-a}\left(\frac{d\,\gamma_v}{d\,\theta}\right)\right) + \phi_{ha}k_i} \right)\frac{\partial\theta}{\partial x} \tag{4}$$

Now rearrange Eq. (4) by dividing numerator and denominator by k_i, leading to:

$$q = -\left(\frac{\left(k_{ha} + D_{v-a}\ u_{sg}\frac{d\gamma_v}{d\theta}\right)}{\phi_i\left[\frac{k_{ha} + u_{sg}D_{v-a}\left(\frac{d\,\gamma_v}{d\,\theta}\right)}{k_i}\right] + \phi_{ha}} \right)\frac{\partial\theta}{\partial x}. \tag{5}$$

The value of the thermal conductivity of ice is on the order of 100 times that of the thermal conductivity of humid air. Therefore, neglecting the terms in square brackets in the above equation leads to

$$q = -\left(\left(\frac{k_{\text{ha}}}{\phi_{\text{ha}}}\right) + \left(\frac{D_{\text{v}-\text{a}}}{\phi_{\text{ha}}}\right) u_{\text{sg}} \frac{d\,\gamma_{\text{v}}}{d\,\theta}\right) \frac{\partial\theta}{\partial x} \quad . \tag{6}$$

The authors did not seem to want to touch on this simplification in the Technical Note, so let's make some comparisons of the normalized energy flux between the exact form in Eq. (1) and the approximate form of Eq. (6).

Let us use needed numerical values from Appendix A of the Technical Note. Further let's assume a temperature of 271.15 K (-2°C) as high temperatures are where diffusion is greatest (Hansen and Foslien, 2015, Fourteau, 2020). A brief table of differences between the exact and approximate forms of the normalized energy flux is shown below.

	Eq. 1 $\left\lvert q/\frac{\partial\theta}{\partial x}\right\rvert$	Eq.6 $\left\lvert q/\frac{\partial\theta}{\partial x}\right\rvert$
$\phi_{\text{i}} = 0.2$	0.056701	0.05699
$\phi_{\text{i}} = 0.3$	0.064567	0.065131

The results are remarkably close (errors less than 1 %) which should come as no surprise. Figure 2 shows a plot of the exact normalized energy flux of Eq. (1) and the approximate energy flux of Eq. (6). The results show the two are in excellent agreement for ice volume fractions below 0.7. In what follows, let us restrict the discussion to ice volume fractions below 0.7 as it opens a window of clarity on the competing views.

I'll briefly remark that restricting ice volume fractions to be below 0.7 is hardly a constraint when it comes to studying snow. Diffusion is only a meaningful form of heat transfer for low densities (typically less than 0.5 ice volume fraction) and high temperatures, a result confirmed by Hansen and Foslien (2015) and the present paper of Fourteau.

The macroscale energy flux for the continuum mixture may be written as

$$q = -\left(k^{\text{eff}} + D^{\text{eff}} u_{\text{sg}} \frac{d\gamma_{v}}{d\theta}\right) \frac{\partial\theta}{\partial x} \quad . \tag{7}$$

where k^{eff} and D^{eff} represent the effective thermal conductivity and the effective diffusion coefficient of the mixture.

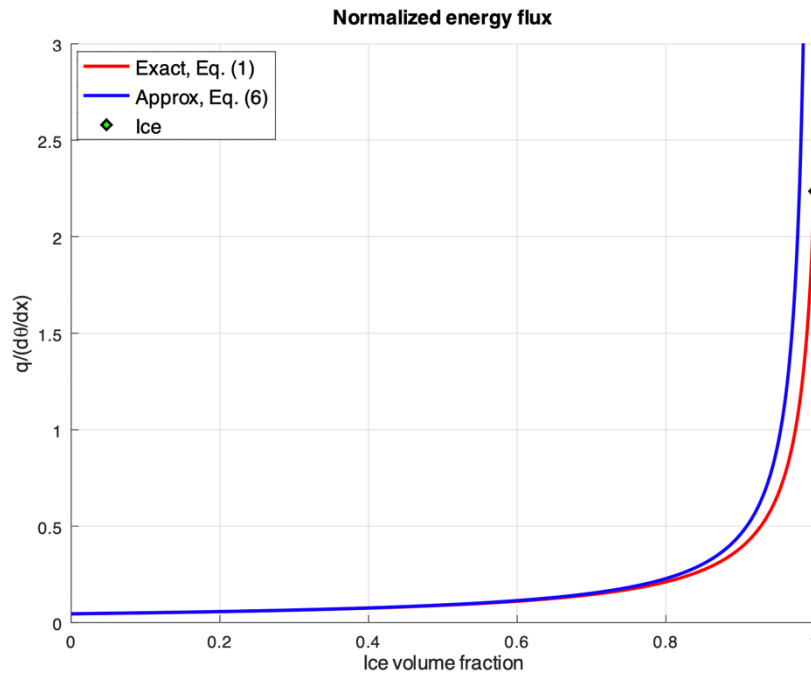Comparing Eqs. (6) and (7) leads to the relations

Figure 2. A comparison of the normalized energy flux of the layered microstructure using Eqs. (1 & 6), respectively. Note that the exact energy flux converges to the known energy flux for the case of solid ice.

$$k^{\text{eff}} = \left(\frac{k_{\text{ha}}}{\phi_{\text{ha}}}\right), \qquad\qquad (8)$$

and

$$\boxed{D^{\text{eff}} = \left(\frac{D_{\text{v}-\text{a}}}{\phi_{\text{ha}}}\right)}. \qquad\qquad (9)$$

The authors apparently wish to argue the above results are "invalid" owing to the claimed "arbitrary" nature of the decomposition of Eq. (1). However, by performing the order of magnitude analysis on Eq. (1) first, and restricting the discussion to ice volume fractions below 0.7, there is no ambiguity in the decomposition of thermal conductivity and the diffusion coefficient. The decomposition is rigorously correct and the identification of terms is crystal clear when comparing Eqs. (6 & 7). As for the word "arbitrary" used by the authors to describe the decomposition, we shall see that their approach is the one that appears to utilize an arbitrary decomposition to arrive at a non-physical result.

I'll dispense with some of the details of the authors' development and refer the reader to Appendix C in Fourteau et al. (2021). Begin by noting Eq. (C.4) of Fourteau (2021) is precisely Eq. (1) of this response. Next, a critical equation in their development arises in the statement below

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"The heat flux q^{cond} through the sole process of conduction is thus given by"

$$q^{cond} = (1 - \emptyset)k_i \nabla T_i + \emptyset k_a \nabla T_a \tag{10}$$

or written in terms of the notation used herein

$$q^{cond} = \phi_i k_i \nabla \theta_i + \phi_a k_{ha} \nabla \theta_a \qquad . \tag{11}$$

From this point on, it is merely an exercise in algebra for the authors to arrive at their results for thermal conductivity and the effective diffusion coefficient given by

$$k^{eff} = \left(\frac{k_i \left(k_{ha} + D_{v-a}\, \phi_i\, u_{sg}\, \frac{d\gamma_v}{d\theta} \right)}{\phi_i \left(k_{ha} + u_{sg} D_{v-a} \left(\frac{d\gamma_v}{d\theta} \right) \right) + \phi_{ha} k_i} \right) \qquad , \tag{12}$$

and

$$D^{eff} = \left(\frac{k_i \phi_{ha} D_{v-a}}{\phi_i \left(k_{ha} + u_{sg} D_{v-a} \left(\frac{d\gamma_v}{d\theta} \right) \right) + \phi_{ha} k_i} \right) \qquad . \tag{13}$$

The above results are consistent with Fourteau (2021), Appendix C, Eq. (C.7) and accompanying discussion.

In their present form, the complexity of Eqs. (12 & 13) and those of Foslien (1994) make comparisons difficult. However, we can proceed with the same order of magnitude analysis and follow up by restricting the discussion to ice volume fractions below 0.7.

Begin by dividing numerator and denominator of Eqs. (12 & 13) by k_i leading to

$$k^{eff} = \left(\frac{\left(k_{ha} + \phi_i D_{v-a}\, u_{sg}\, \frac{d\gamma_v}{d\theta} \right)}{\phi_i \left[\frac{k_{ha} + u_{sg} D_{v-a} \left(\frac{d\gamma_v}{d\theta} \right)}{k_i} \right] + \phi_{ha}} \right) \tag{14}$$

$$D^{eff} = \left(\frac{\phi_{ha}\, D_{v-a}}{\phi_i \left[\frac{k_{ha} + u_{sg} D_{v-a} \left(\frac{d\gamma_v}{d\theta} \right)}{k_i} \right] + \phi_{ha}} \right) \tag{15}$$

Neglecting the terms in square brackets by following the identical arguments used previously for the approach of Hansen leads to

$$k^{eff} = \left(\frac{\left(k_{ha} + \phi_i D_{v-a}\, u_{sg}\, \frac{d\gamma_v}{d\theta} \right)}{\phi_{ha}} \right) \tag{16}$$

$$D^{\text{eff}} = D_{\text{v-a}} \tag{17}$$

We are now in a position to bring remarkable clarity to comparisons of the two approaches. To begin, the energy flux of the macroscale mixture is given by Eq. (7) repeated below as:

$$q = -\left(k^{\text{eff}} + D^{\text{eff}} u_{\text{sg}} \frac{d\gamma_v}{d\theta}\right) \frac{\partial\theta}{\partial x} \ . \tag{18}$$

The energy flux of Foslien (1994) is given by Eq. (6), repeated below as:

$$q = -\left(\left(\frac{k_{\text{ha}}}{\phi_{\text{ha}}}\right) + \left(\frac{D_{\text{v-a}}}{\phi_{\text{ha}}}\right) u_{\text{sg}} \frac{d\gamma_v}{d\theta}\right) \frac{\partial\theta}{\partial x} \tag{19}$$

where $k^{\text{eff}} = \left(\frac{k_{\text{ha}}}{\phi_{\text{ha}}}\right)$ and $D^{\text{eff}} = \left(\frac{D_{\text{v-a}}}{\phi_{\text{ha}}}\right)$.

Hence, in this case, the thermal conductivity depends only on the humid air thermal conductivity and the diffusion coefficient depends only on the binary diffusion coefficient of water vapor in air.

The energy flux of Fourteau is given by

$$q = -\left\{\left(\frac{\left(k_{\text{ha}}+\phi_i D_{\text{v-a}} u_{\text{sg}} \frac{d\gamma_v}{d\theta}\right)}{\phi_{\text{ha}}}\right) + \phi_{\text{ha}} \left(\frac{D_{\text{v-a}}}{\phi_{\text{ha}}}\right) u_{\text{sg}} \frac{d\gamma_v}{d\theta}\right\} \frac{\partial\theta}{\partial x} \tag{20}$$

where Fourteau identifies

$$k^{\text{eff}} = \left(\frac{\left(k_{\text{ha}}+\phi_i D_{\text{v-a}} u_{\text{sg}} \frac{d\gamma_v}{d\theta}\right)}{\phi_{\text{ha}}}\right) \qquad \text{and} \qquad D^{\text{eff}} = D_{\text{v-a}} \ .$$

Note that, upon careful inspection, the energy fluxes of Hansen and Fourteau (Eqs. 19 & 20) are identical—a comforting result for both approaches. The difference in the two approaches is in the decomposition to thermal conductivity and the effective diffusion coefficient. In Foslien's model, the thermal conductivity and the diffusion coefficient separate cleanly, whereas the Fourteau approach apportions part of mass diffusion to the thermal conductivity and part of the mass diffusion to the diffusion coefficient. Without addressing the root cause just yet, I'll simply note the result of Fourteau is neither mathematically or physically appealing.

Now let us briefly revisit how these two results came about.

In the case of Foslien:

i.   First principles of heat and mass transfer were used to derive Eq. (1), a result that is actually used by Fourteau.

An important equation utilized in the derivation of the macroscale energy flux is the volume average of the constituent temperature gradients given by

$$\frac{\partial \theta}{\partial x} = \phi_i \left(\frac{\partial \theta}{\partial \xi} \right)_i + \phi_{ha} \left(\frac{\partial \theta}{\partial \xi} \right)_{ha} \qquad (21)$$

This equation is discussed in depth in the Technical Note and is derived by Özdemir et al. (2008)—a very important paper.

ii.   An order of magnitude analysis is used to simplify the exact form of Eq. (1). The authors may be tempted to challenge the order of magnitude analysis but I have clearly demonstrated the exact and approximate solutions are in excellent agreement for ice volume fractions below 0.7.

iii.   The result of (i and ii) is an energy flux equation with two terms and no room for an "arbitrary" interpretation of the decomposition to thermal conductivity and the diffusion coefficient.

In the case of Fourteau:

i.   Appendix C, Eq. (C.4) of Fourteau (2021) is Eq. (1) of this response—the same as developed by Foslien.

ii.   Fourteau then invokes the statement "The heat flux q^{cond} through the sole process of conduction is thus given by" (using my notation)

$$q^{cond} = \phi_i k_i \left(\frac{\partial \theta}{\partial \xi} \right)_i + \phi_{ha} k_{ha} \left(\frac{\partial \theta}{\partial \xi} \right)_{ha} \qquad (22)$$

The diffusion coefficient is then developed by recognizing the latent heat flux contribution is given by $q - q^{cond}$.

iii.   The remainder of the development is an algebra exercise.

At first blush, there seemingly appear to be two correct approaches, yielding different results for the diffusion coefficient. However, there is a fundamental problem with the approach of Fourteau. *In particular, the volume averaging technique of Eq. (22) is not valid for a diffusing mixture of ice and humid air—a profound statement.*

Let's develop the arguments in detail. Following Özdemir et al. (2008), the volume average heat flux, (for our problem it is the energy flux), defines the energy flux of the mixture as

$$q = \phi_i q_i + \phi_{ha} q_{ha}. \qquad (23)$$

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Now, if one wishes to determine only the thermal conductivity of the mixture, we set the diffusion terms to zero, or neglect them if that is your preference, and we invoke Eq. (23). This process leads identically to Eq. (22), under the condition that diffusion is neglected. For ice volume fractions less than 0.7, this process will produce a thermal conductivity given by

$$k^{\text{eff}} = \left(\frac{k_{\text{ha}}}{\phi_{\text{ha}}} \right) \quad . \tag{24}$$

Although such an approach is not necessary, I'll note that the approach outlined to achieve the thermal conductivity of snow is precisely that used by Calonne (2011) and Riche and Schneebeli (2013). For example, Calonne et al. (2011) state: "Neglecting convection, phase change and under steady state conditions, heat transfers at the microscopic scale are described by…" Riche and Schneebeli state: "No phase change is implemented in the model."

Next, we seek to add diffusion into the problem. Again, the correct statement for volume averaging of the mixture is that the volume averaged energy flux is given by Eq. (23). This equation is developed in detail by Özdemir et al. (2008) from thermodynamics and an entropy consistency between scales. There is no such similar justification for the use of Eq. (22) as done by the authors. By using Eq. (23), one is immediately led to the energy flux of the mixture given by Eq. (19), the same form produced by Foslien (1994).

Eq. (23) is firmly grounded in thermodynamic principles. In contrast, Eq. (22), while seemingly straight forward, is simply haphazardly postulated. As a result, the thermal conductivity developed by the authors depends on the diffusion coefficient. Indeed, the authors' decomposition, and not the one of Foslien, is the one that appears "arbitrary." I won't use the powerful term of "invalid" used by the authors, but I believe *non-physical* is an appropriate description of their result.

In summary, the entire Appendicx C of Fourteau (2021) is algebraically correct. However, the mathematics and physics are inherently flawed. ***The irony of the development of the authors is that, by using Eq. (22), the authors were attempting to isolate thermal conductivity terms in the effective thermal conductivity of the mixture and, in doing so, they introduced diffusion into the thermal conductivity.*** In contrast, Foslien's approach cleanly separates thermal conductivity and diffusion terms for ice volume fractions below 0.7 where the order of magnitude analysis is precise. Moreover, there is nothing arbitrary in the Foslien decomposition as any extraneous possibilities are eliminated in the order of magnitude analysis.

In closing, I'll make one final comment about Foslien's model. Instead of initially focusing on the layered microstructure, let's examine the diffusion of water vapor in humid air alone. The energy flux is given by (Bird, 1960)

$$q_{\text{ha}} = - \left(k_{\text{ha}} + D_{\text{v-a}} u_{\text{sg}} \frac{d\,\gamma_{\text{v}}}{d\,\theta} \right) \frac{\partial \theta}{\partial x} \quad . \tag{25}$$

After a detailed and precise analysis shown in Eqs. (1-9) of this response, we find that the energy flux for the ice/humid air mixture can be achieved by simply dividing the RHS of the above equation by ϕ_{ha}, leading to

$$q = -\left(\left(\frac{k_{ha}}{\phi_{ha}}\right) + \left(\frac{D_{v-a}}{\phi_{ha}}\right)u_{sg}\frac{d\,\gamma_v}{d\,\theta}\right)\frac{\partial\theta}{\partial x} \quad . \tag{26}$$

Note that in Eq. (26), the ice phase has the same influence on the thermal conductivity as it does on the latent heat. Intuition would certainly argue for this—they are both heat transfer terms. In brief, the solution put forth by Foslien is rigorous, elegant, clean, and in agreement with the expected physics. Moreover, there is nothing arbitrary in the decomposition under the constraint of ice volume fractions less than 0.7.

As an aside, Equation (26) generates a desirable trait in that the thermal conductivity and the diffusion coefficient decouple from one another. However, the additional coupling terms neglected in the order of magnitude analysis are important for extremely high ice volume fractions as they serve to keep the solution bounded and, further, generate self-consistent results for the known energy flux in the limiting case of solid ice, see Figure 2. The cacophony put forth by the authors about Eq. (1) having multiple ways to combine terms is a whole lot of nothing other than in regions of exceptionally high ice volume fractions where diffusion is virtually nonexistent—this particular argument is simply a non-issue, yet it is the defining feature of their objection to my work.

In closing, I would like to return to two quotes of the authors concerning their response to the Technical Note. In discussing the Note, they assert:

- "An invalid derivation of the mass flux in the layered structure by Hansen and Folsien (2015)."

    and

- … "the identification of the latent-heat flux with the second term
of the decomposition is arbitrary."

The authors have attempted to discredit the Technical Note by making cavalier statements such as the above with no justification. Their assertions are patently incorrect. If they wish to make these claims, they have ample opportunity to support them in this detailed response.

For the benefit of the reader, I would ask the authors to please:

i.   Point out explicitly where the solution outlined in Eqs. (1-9) is invalid, subject to the constraint of ice volume fractions below 0.7.

ii. Where is the arbitrary nature of the decomposition of Eq. (6)?

The fact that the authors are not meeting this challenge head-on is a clear indicator of the rigorous nature of the approach of Foslien. As I said previously, 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 mass transfer at the macroscale.

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Given the results produced in this response, the reader is encouraged to revisit the Technical Note. The Note provides remarkable clarity on the mass transfer problem in a layered ice/humid air microstructure, producing a unified theory where the analytical solution, volume averaged solution, and the known surface flux developed some 50 plus years ago (de Quervain, 1963) are all in perfect agreement. The authors are unable to match these consistent results with their formulation.

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