

## Response to tc-2022-83

### General Comments:

The author of the tc-2022-83 comment argues that macroscopic water vapor diffusion flux in snow (the vapor flux at a large scale, when snow is viewed as a homogeneous medium) can be enhanced, in opposition to conclusions advanced, notably, in Fourteau et al., (2021a). Here, “enhanced” means that the macroscopic diffusion flux of vapor in snow is larger than the diffusion flux that would be observed in pure air under the same vapor concentration gradient.

The tc-2022-83 comment suffers from major flaws: it goes against physics (such as mass conservation or the difference between advection and diffusion fluxes), mixes up sources of vapor and diffusion fluxes of vapor, misunderstands what a macroscopic description is, and unfairly mischaracterizes the work it is supposed to comment on.

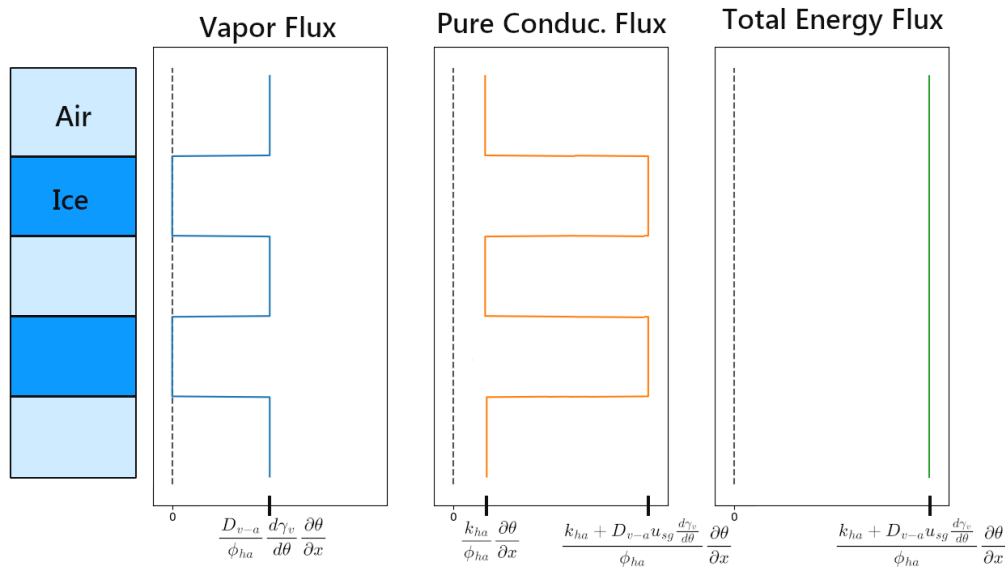
We have explained in Fourteau et al. (2021a), and in a public response to a comment to Fourteau et al. (2021b) (<https://tc.copernicus.org/preprints/tc-2020-317/tc-2020-317-AC1.pdf>), that several fundamental problems needed to be avoided when discussing macroscopic diffusion of vapor. These problems are however at the core of the tc-2022-83 comment. Specifically, two major ones can be pointed out:

#### **Problem 1** (explained in 3<sup>rd</sup> paragraph, Section 3 of Fourteau et al., 2021a):

The author of tc-2022-83 is trying to compute the macroscopic water vapor flux in an idealized layered structure of ice and air (see Figure 1 below). The combination of this specific layered structure and of deposition/sublimation at the ice/air interface leads to a non-trivial situation, which is not treated properly in tc-2022-83. To compute the macroscopic water vapor flux, the author computes the microscopic vapor flux crossing horizontal surfaces in the air phase, between two ice layers.

However, in a layered structure of ice and air, the microscopic water vapor fluxes (and the heat conduction fluxes) differ between the ice and air phases (see Figure 1 below for the values in each phase). Computing the vapor fluxes in the air only, and ignoring what is occurring in the ice, is arbitrary and yields incorrect results. This is shown with a simple reduction to absurdity: with the same reasoning, one could compute the water vapor fluxes crossing a horizontal surface in the ice phase (the ice being as much a part of snow as the air), and find that the macroscopic vapor flux is null. Thus, the same reasoning leads to different diffusion coefficients for the same snow microstructure, in contradiction with the notion that a given snow microstructure has a unique macroscopic description. In other words, trying to obtain the macroscopic flux as the flux through given horizontal surfaces is flawed.

What happens is that by focusing on just the air phase, the author is misinterpreting local microscopic fluctuations as the actual macroscopic description. At the macroscopic scale, these microscopic fluctuations average out into the volume-average. The removal of these fluctuations in effective transport coefficients is one of the basis of a macroscopic description (e.g. Quintard and Whitaker, 1990).



**Figure 1:** Mass, heat conduction, and total energy fluxes in a layered structure. The demonstrations in the tc-2022-83 comment essentially revolve around computing the fluxes in the air phase only (where vapor flux is maximal and conduction flux minimal), neglecting the fact that these fluxes are different in the ice.

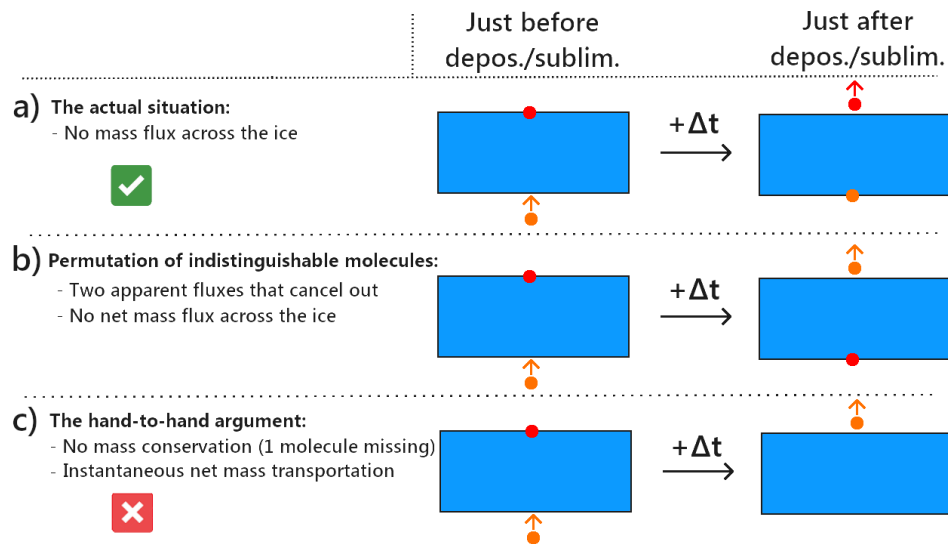
**Problem 2** (explained in 2<sup>nd</sup> and 3<sup>rd</sup> paragraphs, Section 2 of Fourteau et al., 2021a):

To support enhanced macroscopic water vapor fluxes, the tc-2022-83 comment is also invoking the “hand-to-hand” mechanism, in which the simultaneous deposition of a water molecule below an ice grain and sublimation at the top of the ice grain is counted as an actual mass transport, as if the water vapor molecule were instantaneously transferred across the ice.

However, the simultaneous deposition and sublimation of vapor simply does not produce any mass transport across the ice. No mass is spatially transported through the ice in the process, as molecules stay where they are during sublimation/deposition (see Figure 2a below and Figure 1 of Fourteau et al., 2021a).

The tc-2022-83 comment also points to the indistinguishability of water vapor molecules as supporting the hand-to-hand mechanism. The argument says that since water molecules are indistinguishable, everything really appears as if the vapor molecule instantaneously moved across the ice. However, indistinguishability does not change the situation: no mass is transferred during sublimation/deposition. Indeed, if one argues that it appears as if the vapor molecule teleported through the ice phase, then the same argument would also hold for the water molecule in the ice phase: the molecule at the top surface would re-appear at the bottom surface, canceling the hypothetical hand-to-hand flux of vapor across the ice. All in all, no net mass is spatially transferred during the simultaneous deposition/sublimation (see Figure 2b). The better (and simpler) physical description is of course not to invoke these apparent fluxes and the indistinguishability of molecules, and to only consider diffusion fluxes in the pores, as done in Fourteau et al., (2021a).

Finally, by arguing that the situation is equivalent to one where the depositing vapor molecule would have been instantaneously transferred through the ice, two physics principles are violated: (i) matter does not teleport and (ii) mass cannot simply disappear (1 molecule is missing in Figure 2c).



**Figure 2:** Three visions on what happens during the deposition/sublimation of vapor. a) The first one is the actual/real one (and there is no mass transfer). b) The second is what can be said by properly using the argument of indistinguishability (it is a weird view of things, but it is consistent with the actual physics). c) The third one is the hand-to-hand argument: it violates mass conservation and the notion that infinitely fast mass transport is impossible.

The tc-2022-83 comment essentially revolves around Problem 1: the mass and heat conduction fluxes are computed only in the air phase through various methods (from direct computation, from mass balance in a control volume having its surface boundaries in the air, from deposition at the ice/air interface, etc), and then wrongly attributed to the macroscopic scale, as if results obtained in the air applied to the whole structure. Problem 2 is also regularly invoked as a supporting interpretation of the results yielded by Problem 1.

To support its conclusion, the tc-2022-83 comment also introduced the “reservoir effect”: there is a vast amount of water molecules in the ice waiting to sublimate and diffuse in the air. However, this whole argument is based on a confusion between vapor sources (that do not move mass across space) and vapor diffusion fluxes (that actually move mass from one place to another). It is a variation of Problem 2.

Apart from that, we also want to stress that the tc-2022-83 comment mischaracterizes what is actually said in Fourteau et al. (2021a) (e.g. Specific Comments L16 and L25 below) and does not reply to the points raised in it (e.g. L364-372 below). Scientific standards would expect the author to properly describe the article they are commenting on. Failure to do so is detrimental to readers and referees, and deteriorates the quality of the scientific debate.

Finally, we note that the tc-2022-83 comment contradicts 30 years of well-established scientific literature on the subject of the macroscopic description of humid materials. For instance, Whitaker (1998) provides a demonstration that the macroscopic vapor diffusion flux is the volume average of the microscopic fluxes (right-hand-side of Equation 162). Section 5.1 of Moyne et al. (1988) provides an analysis of the exact layered problem that the tc-2022-83 comment is focusing on. The normalized diffusion coefficient is given in Equation 60 and the thermal conductivity in Equation 59. They are both equal to the expressions given in Appendix C of Fourteau et al. (2021a). Moyne et al. (1991) provides a general expression of the normalized diffusion coefficient based on the averaged temperature gradient in the air phase (Equation 12). This is consistent with what we obtained in Equation 5 of Fourteau et al. (2021a) and the exact same expression as in Equation 10 of Fourteau et al. (2021b).

By ignoring well-established techniques of homogenization, the tc-2022-83 represents a step back in the understanding of water vapor movement in porous media.

## Specific Comments:

Here, we quickly list some of the tc-2022-83 comment's biggest flaws, as an illustration of what was summarized in the General Comments.

**L16:** Claiming that we consider that “*the ice phase has no effect other than occupying volume where diffusion cannot occur*” is false. We actually spent most of our article discussing the influence of the coupling between the diffusion flux and the presence of ice through surface effects.

**L25:** We never argued that there is an actual “*counterflux of water vapor in the form of downward motion of ice*” and are well aware that this downward motion is only an appearance (as exemplified by the sentence “*for an observer focused on the ice everything **appears** as if the ice disappearing on the sublimation side reappeared on the depositing side*”).

We actually argued that the author's “hand-to-hand” mass flux is also only apparent, and that to be consistent they should consider both apparent fluxes and not just the one that suits their conclusion (“*the **apparent** flux of vapor skipping the ice is compensated by an equal counter-flux of water molecules in the ice phase*”). See Figure 2 of this response.

**L42:** The releasing of water vapor from the ice phase is a source, not a transport flux. It does not spatially move mass, it only changes its form (from ice to vapor). There is no mass transport other than when and where vapor diffuses in the pore.

**L79:** The indistinguishability of water molecules does not create a mass transport (Problem 2).

**L109:** Of course, gradient metamorphism can exist without molecules teleporting through the ice, and the author should be aware of that as it is explained in Fourteau et al. (2021a).

We clearly stated at the end of Section 2 of Fourteau et al. (2021a) that one should be careful to separate between the notions that (i) water molecules preferentially deposit below ice grains under temperature gradient (which is physically sound and visible in the simulations in Figures 3 and 5 of Fourteau et al., 2021a), and (ii) that such deposition patterns act as mass transport (which is Problem 2).

**L170:** Problem 1. The author computes the diffusive mass flux in the air phase (and the deposition/sublimation flux at the ice/air interface, which has an equal magnitude), but then inappropriately interprets it as the macroscopic diffusive flux.

**L258:** Problem 1. The author computes the diffusive flux in the air, but then wrongly interprets it as the macroscopic diffusive flux.

**L299-310:** It is normal that Eqs 16 and 17 do not reproduce the mass and conduction fluxes in the air, since they are supposed to give the macroscopic flux (that would be Problem 1 otherwise).

**L335-353:** Term (2) simply corresponds to the extra heat conduction in the ice phase (see the orange curve in Figure 1 of the response), scaled by the amount of ice that carries this extra conduction flux.

As this extra heat conduction in the ice phase accommodates the release/absorption of latent heat at the ice/air interface (in order to respect the continuity of the total energy flux at the ice/air interface), it is perfectly normal that it depends on latent heat and diffusion of vapor.

**L364-372:** Problem 2 (sublimation/deposition does not create a mass transfer). This whole discussion is also unnecessary when one realizes that term (2) is simply the extra conduction flux in

the ice due to microscopic latent heat/conduction coupling (see Section 2.2 and Section 3.1 of Fourteau et al., 2021b for a discussion on this physical effect).

The author should be aware of that, as that is exactly what we pointed out in Section 4.3 of Fourteau et al. (2021a) when discussing Hansen and Folsien (2015): “*However, during the identification of the latent-heat contribution to the total energy flux, some of the heat conduction contribution of the ice is attributed to the latent-heat transport*”.

**Section 4.2:** The author is trying to compute the macroscopic advecto-diffusive (with an advection and a diffusion component) water flux within a moving control volume. The control surfaces are placed in the air phase.

- Eq. 37 is false, the microscopic advection flux of vapor at the boundaries is missing ( $v_{i/c}\gamma_v$ ).
- First line of Eq. 38 is false, the averaged advection flux of vapor is missing ( $\phi_{ha}v_{i/c}\gamma_v$ ).
- With the advection of vapor properly included, the volume averaged advecto-diffusive flux does not equal the advecto-diffusive flux on the air surface boundaries. They differ by  $\phi_i v_{i/c}\gamma_v$ . This is normal since the advection and diffusion fluxes in the air are not representative of the whole microstructure (variation of Problem 1).
- This (wrongly) computed macroscopic advecto-diffusive water flux is finally inappropriately interpreted as the sole macroscopic diffusive flux.

To sumup Section 4.2: The macroscopic advecto-diffusion flux is wrongly computed by dropping off the advection of vapor. Then, the advection of ice is lumped with the diffusion of vapor and the ensemble is interpreted as a pure diffusion flux (neglecting the fact that extra advective water fluxes have been introduced through the motion of the reference frame).

To try a simple analogy, this is akin to saying that someone can run at 50km/hr because they are moving at this velocity in the reference frame of a moving car. To obtain the actual speed of the runner respective to the ground, we need to retract the advection term due to the movement of the reference frame. The same thing applies to obtain the diffusion flux of water in a moving reference frame.

If the author includes the advection of vapor in Eq. 38, and then removes the macroscopic advective flux of water ( $v_{i/c}\langle\gamma_{water}\rangle = v_{i/c}(\phi_{ha}\gamma_v + \phi_i\gamma_i)$ ), they would find that the remaining diffusive flux is given by the volume-averaged microscopic diffusion fluxes (as in Moyne et al., 1991, Whitaker et al., 1998, or Fourteau et al., 2021a).

**Section 4.3:** There is no conflict to resolve here: microscopic vapor fluxes depend on whether we are in the air or the ice. The difference between the two is accommodated through sublimation/deposition at the ice/air interface. If the lower and upper microscopic boundary fluxes differ, it means that there is a net accumulation/depletion in the control volume that accommodates the microscopic flux imbalance.

But sublimation and deposition are vapor sources/sinks, not diffusion fluxes of vapor. They do not transfer mass in space (Problem 2). The vapor that is released at the ice/air interface was not spatially moved during the process.

The reservoir effect is just a confusion between sources (that do not move mass) and diffusion fluxes (that do move mass).

**L699-706:** The lake analogy is wrong and at the heart of Problem 1. There is no local sources and accumulation/depletion of mass in a steady state lake: thus what goes in must go out, in every control volume. That is why one can find that all the microscopic mass fluxes integrated across a perpendicular surface spanning the lake are equal: all surfaces have a representative mass flux

across them. In this peculiar case, the “macroscopic” mass flux going through the lake can be obtained by simply considering the inflow or outflow fluxes.

In the layered ice structure, there is local accumulation/depletion of mass (at the ice/air interface): what goes in can accumulate and stay there. Thus, the flux is not constant through any perpendicular surfaces (see Figure 1 of the response), and one cannot simply look at mass fluxes through a simple surface (Problem 1).

**L714-720:** The author got it correct that point A just stay in the ice phase, potentially being released later, where it will start diffusing again from the very same point in space where it deposited. However, this incorporation of water in ice does not create any mass flux across the ice phase at the moment of deposition (this would be Problem 2), and there is no mass movement besides when and where vapor diffuses in the air.

**L723-L734:** The author is arguing that since a molecule in the ice might be eventually released, from the exact same place where it deposited and after sufficient time has elapsed, this molecule should be counted as having skipped across the ice phase during its deposition. This is absurd.

**Section 5.1:** This section is a convoluted way of falling into Problem 1.

First, the author computes the mass deposited below the ice during a time interval  $\tau$  as the product of the deposition rate and  $\tau$  (Eq. 48). They then find that this quantity of mass is larger than the deposition that would occur if the deposition flux were equal to the diffusion flux in pure air.

This result is trivially stating that the deposition flux in the layered structure is larger than the diffusion flux in pure air under a similar gradient, something we already knew and agreed upon (see Figure 1 of this response). But interpreting the elevated deposition flux as an elevated macroscopic flux is Problem 1 (confusion between local microscopic fluxes and the macroscopic one).

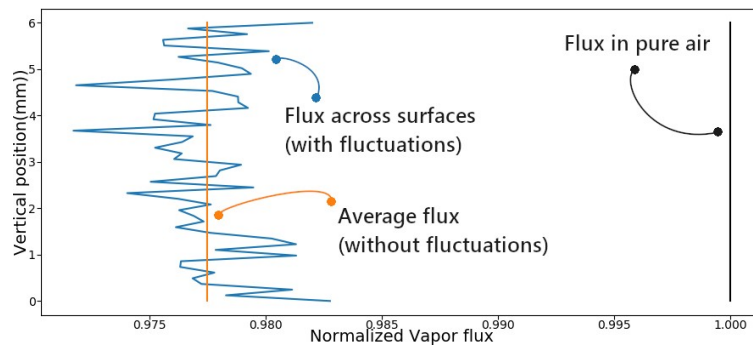
It might also seem that the demonstration relies on tracking mass movement over time, since the chosen time interval is the time it would take a “material point” to travel from a sublimation surface to a deposition one. However, the demonstration would be the same whatever the time interval  $\tau$  (this can easily be seen by keeping  $m$  as  $\gamma_v v_v \tau$  in the computation). Thus, the notion of tracking mass movement during a deposition/sublimation cycle is irrelevant to the demonstration.

**L888:** Most of the errors in the tc-2022-83 comment originate from the confusion between the macroscopic flux and local microscopic fluxes through horizontal surfaces, which strongly differ in the case of the layered microstructure.

However, for random heterogeneous media, such as snow, this problem disappears, as microstructural fluctuations across horizontal layers tend to statistically average out. In this case the mass fluxes through horizontal surfaces are equal to the volume average (as explained in our public response <https://tc.copernicus.org/preprints/tc-2020-317/tc-2020-317-AC1.pdf>).

We took the time to compute the mass fluxes crossing the various horizontal surfaces of a snow microstructure with a finite-element simulation (Figure 3 below): the surface fluxes are basically all the same (with small microscale fluctuations). They are below the flux that would be observed in the air under a similar macroscopic vapor gradient (i.e. not enhanced).

So, not only is the idea of computing fluxes through individual horizontal layers giving strongly erroneous results in the case of the layered structure, it would not even lead to enhanced vapor fluxes in the case of real snow.



**Figure 3:** Vapor fluxes across different horizontal surfaces in a snow sample (in blue). They slightly fluctuate around the average (in orange), and are inferior to the flux in pure air under a similar gradient (black). With a larger sample, the fluctuation would decrease further.

### References:

K. Fourteau, F. Domine, and P. Hagenmuller: Macroscopic water vapor diffusion is not enhanced in snow, *The Cryosphere*, 15, 389–406, doi:10.5194/tc-15-389-2021, 2021a

K. Fourteau, F. Domine, and P. Hagenmuller: Impact of water vapor diffusion and latent heat on the effective thermal conductivity of snow, *The Cryosphere*, 15, 2739–2755, doi:10.5194/tc-15-2739-2021, 2021b

A.C. Hansen and W.E. Foslien: A macroscale mixture theory analysis of deposition and sublimation rates during heat and mass transfer in dry snow, *The Cryosphere*, 9, 1857–1878, doi:10.5194/tc-9-1857-2015, 2015

C. Moyne, J.C. Batsale and A. Degiovanni: Approche expérimentale et théorique de la conductivité thermique des milieux poreux humides – II. Théorie, *Int. J. Heat Mass Transfer.*, 31, 11, 2319-2330, 1988, doi:10.1016/0017-9310(88)90163-9

C. Moyne and P. Perre: Processes related to drying: Part I, Theoretical model. *Drying Tech.*, 9, 5, 1135-1152, 1991, doi:10.1080/07373939108916746

M. Quintard and S. Whitaker: Two-phase flow in heterogeneous porous media I: The influence of large spatial and temporal gradients, *Transp. Porous Media*, 5, 341-379, 1990

S. Whitaker: Coupled transport in multiphase systems: A theory of drying, *Adv. Heat Transfer.*, 31, 1998, url: [https://www.researchgate.net/publication/241065896\\_Coupled\\_Transport\\_in\\_Multiphase\\_Systems\\_A\\_Theory\\_of\\_Drying](https://www.researchgate.net/publication/241065896_Coupled_Transport_in_Multiphase_Systems_A_Theory_of_Drying) (last access: 03/06/2022)