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Title: Technical note: An assessment of the relative contribution of the Soret effect to open water evaporation

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Response to RC5 (Anon. Reviewer 5, 23 Sep 2024)

Review comments in italics.

Author Response in bold.

1. *This short technical note deals with quantifying the relative contribution of the Soret effect (i.e. thermodiffusion) compared to regular “Fickian” diffusion in the evaporation of open water bodies. The motivation behind the paper is to justify the standard practice of neglecting the Sorret effect for evaporation. For this purpose, it is based on the gas kinetic theory of Chapman and his colleagues (completed with experimental determination of the Sorret effect in binary mixtures) and on a recent dataset of evaporation rates under controlled conditions.*

I think the paper is of general interest for people working of water vapor transport (even beyond the sole problem of open water evaporation) and is well suited for HESS. There is however one main point of concern (General Comment 1) that I would like the authors to clarify.

We thank the reviewer for the comments.

2. *General Comments 1 – It is stated at the very start of the paper that the evaporation rate of an open water body is controlled by the ability of water vapor to diffuse in the air. From what I understand this is clearly the case when the air above the water is still (in this case, diffusion in the limiting factor, effectively setting its rate for the evaporation). However, as soon as wind and turbulence is involved, I have issues understanding to what extent the evaporation rate remains controlled by the molecular diffusion in the air. I’m no boundary-layer scientist, but from what I understand, the diffusion flux of Eq. 4 applies within the micro layer (following the wording of Roland B. Stull’s “An introduction to Boundary Layer Meteorology”), i.e. the zone just above the surface where molecular diffusion dominates. Thus, if it is to be translated into Eq. 5 I would say that the gradients of concentration and temperature are to be taken across this micro-layer. And I’m not sure that the concentration and temperature at the top of this micro-layer can be taken as x_a and T_a (as they are influenced by the surface). Otherwise, if x_a and T_a are taken to define the gradients, I think the diffusivity should rather be some “effective” diffusivity (including turbulent effects) and thus does not match the molecular diffusivity anymore. And in this case, it is not clear to me that one can upscale the Sorret effect to*

the whole boundary layer in a similar fashion (i.e. that there is an effective Sorret flux, including turbulence, that has the same form and the same thermal diffusivity ratio as in the purely molecular case). In other words, I think the problem boils down to the difficulty of reconstructing the surface concentration and temperature gradients based on the “air” values, which I assume can be significantly different from what happen in the micro-layer. I would thus like the authors to clarify this point. Especially, references to pre-existing literature treating this problem and relating molecular diffusion in the micro-layer to the air temperature/concentration would be appreciated.

We thank the reviewer for this comment. The reviewer is correct in that we are referring to the molecular boundary layer – what they term the “micro-layer”. Experiments (see Doe, 1967, Measurement of a mass transfer boundary layer. Nature 216, 1101–1103, doi:10.1038/2161101a0) show that that vapour concentration across this layer changes from the surface value (saturated, x_s) to the free stream value (x_a) as has been assumed in our formulation. Indeed, our formulation follows the standard description of the so-called threshold model that has been in use in hydrology, agriculture and climate for the last century. The details are fully described by Fig. 1 in the cited reference (Lim et al, 2012) which is reproduced below:

W.H. Lim et al. / Agricultural and Fc

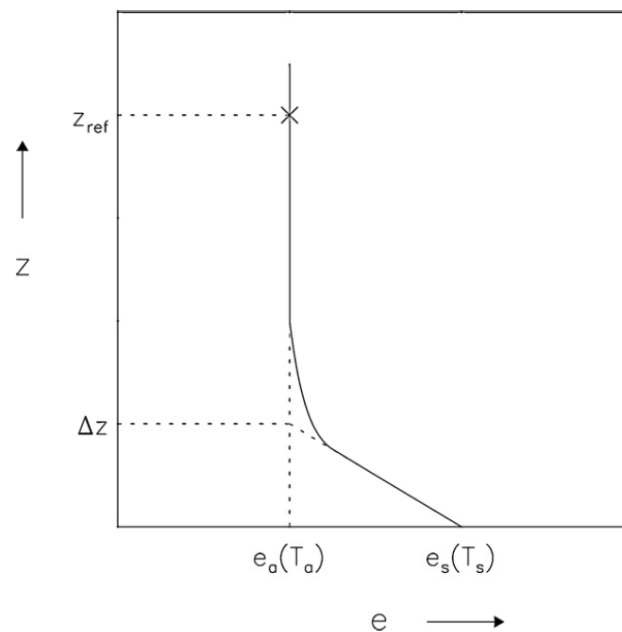


Fig. 1. A schematic diagram of the “threshold model” adopted here for the variation in vapour pressure (e) with height (z) above the evaporating surface. After Leighly (1937) and Machin (1964, 1970).

Fig. R1 Reproduced from Lim et al (2012)

The approach is to replace the actual profile (full line in Fig. R1) with an assumed “threshold-type” model (dotted lines in Fig. R1).

3. 2 – From what I understand the motivation behind this close look at the Sorret effect stems from the recent study of Griffani et al. (2024), that states that thermo-diffusion can

be an effective mechanism of water vapor transport and should not always be neglected. However, this motivation only appears in the discussion. I think it could be quite beneficial to include this in the introduction, as it relates to the state-of-the-art on the subject.

We accept the point and we can add a reference to Griffani et al as well as the necessary text in the introduction.

Specific and technical comments

4. *Abstract – I would systematically say “evaporation from open water” rather than simply “evaporation”.*

On L113 we can modify the text from “... by assuming evaporation follows ...” To read “... by assuming evaporation from open water follows ...”.

5. *L15 – As mentioned in the General Comment 1, references to pre-existing literature would be beneficial here.*

We can add a reference to the classical Monteith and Unsworth 2008 textbook, e.g., gradient (Fick’s Law) (Monteith and Unsworth, 2008) as requested.

6. *L21 – The mention of the Duffour effect is a bit off to me, especially as it is no longer mentioned in the text. It could potentially be discussed a bit more in the Discussion and Conclusions section, notably mentioning that the Onsager reciprocal relations allows one to estimate the Duffour effect from Sorret.*

We agree that the Dufour effect is left hanging on it’s own. We just wanted to make the point that the Onsager-based coupling leads to other effects. We would prefer to leave the text as it is there to highlight that additional flux which we do not study.

7. *L76 and L84 - I would say “limiting conditions” rather than “boundary conditions”.*

Ok, we can do that.

8. *L82 – If I’m not wrong the quadratic form was proposed in the work of Chapman (and certainly others). You could refer to them to justify this specific functional form*

Yes, Chapman did propose that. We can add the reference as requested.

9. *Eq 5 - It might be just me, but I’m not fond of mixing alphabetical and digits in Equations, as it obscure physical variables from actual math constant (and I find it harder to read and interpret). I would keep α_T rather than 0.05.*

We accept the point that this is a matter of “taste”. We also prefer using symbols but in this case we have supplied the actual numeric value that is assumed unchanging (see L131).

10. L136 – It relates to General comment 1. Could you elaborate on the physical significance of Delta z? Is it the thickness of the micro-layer (where diffusion dominates)?

Yes, it is the thickness you mentioned. We can modify the text leading to Eqn 5 (on existing lines 131-132) by stating that we follow the classical threshold model where the temperature and water vapour follow a linear profile from the (saturated) surface to the free stream value in the air over the distance Δz . (Also see response to point 2 above.)

11. Eqs 6a and 6b – Same as Eq. 5. I would go straight to the point and say that Eq 5 using some standards values yields a 99.6%/0.4% partition for the Fickian and Sorret fluxes.

Again a matter of taste. We think that having the numbers written out makes it very clear that the Soret effect will be small (by at least two orders) regardless of the boundary layer model chosen.

12. L149 – For me, “vanishingly small” implies that the Sorret contribution strictly goes to zero when x_s equals x_a . However, I do not think it’s the case as small Sorret contribution remains non zero (if x_s is different from 0 or 1, and T_s different from T_a).

Good point. We can modify the text accordingly.

13. L155 – If it’s the air temperature, please use T_a rather than T .

Good point. It is the air temperature and we can modify the text to read; “... air T range from 15”.

14. Figure 2 – I find it hard to determine the relative contribution of the Sorret flux in panel c near the origin of the graph. Perhaps add a second y-scale the relative contribution of the Sorret flux as a scatter of the total evaporation flux.

We agree that the Soret flux (denoted E_T) is hard to precisely determine near the origin but the main point here is not the exact numerical value, but rather that it is very small.

15. L194 – I think the wording could be improved. From what I understand, the issue is that Griffani et al.’s work is based on Landau’s derivation which assumes that (i) the water vapor molecule are much lighter than the dry air and (ii) the collisions are elastic; both assumption not applying to the actual mixture of water and dry air. The current wording rather suggests that the issues are (i) Griffani et al. is based on Landau ($M=1$ and elastic

collisions) and (ii) the collisions are assumed elastic; in which the same issue actual appears twice.

We can try and improve the wording as suggested. We note that the interpretation of the reviewer is correct.

16. L209 – Taking α_t as 0.05 rather than 0.5 effectively reduces the Sorret flux by an order of magnitude, and the 30% contribution of Sorret in Griffani et al. now becomes a 3% contribution, which is deemed negligible. That being said, I would still leave the door open that in almost all natural conditions, the Sorret effect appears negligible, but that pathological cases (i.e. very high temperature gradients with little concentration gradients) can still exist (and it links with the paragraph below).

Reviewer 3 raised a similar sentiment (see point 3 in the response to Reviewer 3). We repeat that response here:

Adopting a value of 0.05 for the thermal diffusion factor (α_T) we can pose an answer to the reviewers question about when the Soret effect might be important. The obvious condition would be where the absolute magnitude of the surface to air T difference was much larger. We assume the same data as in Table 3 (air at 298 K and 60% relative humidity, atmospheric pressure of 1 bar) but now we increase the surface temperature to 372 K (just under the boiling point of liquid water). At face value the relative magnitude of the Soret effect would be larger but in fact it is smaller than our original calculation (now 0.2 % of the total flux instead of 0.4%, based on Eqn 6a). The reason is that the surface is assumed saturated as appropriate for open water. Hence when the surface-air T difference is larger so is the gradient in mole fraction. The only way we could envisage a significant Soret effect is for the open water to be replaced by a highly concentrated brine-type solution where the saturated vapour pressure at the surface is close to zero and does not increase (much) with change in T . Again we could manufacture such a scenario but the absolute magnitude of the evaporative flux would be small. We can mention this important point in a future revision of the manuscript.