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

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**Abstract.** It is standard practice to assume that evaporation depends on the gradient in water vapor concentration as per Fick's law. However, Fick's law is only true in an isothermal system. In general, we anticipate an additional mass flux due to the temperature gradient (in a non-isothermal system) and this is known as Soret diffusion or the Soret effect. Here we

10  evaluate the relative magnitude of the Soret effect and find that under typical environmental conditions it is at least two orders of magnitude smaller than classical concentration-dependent mass ('Fickian') diffusion. This result justifies the standard practice of ignoring the effect of the temperature gradient by assuming evaporation follows the gradient in water vapor concentration.

## 1 Introduction

15  Evaporation is usually described as mass transfer down a concentration gradient (i.e., Fick's Law). However, strictly speaking, Fick's law is only true under isothermal conditions (Bejan, 2016, p. 639). In many hydrologic applications the surface and adjacent air temperatures are substantially different and the near-surface evaporative environment is generally not isothermal. The temperature gradient will contribute to the mass transfer via what is known as the Soret effect that acts independently of the concentration gradient (Bejan, 2016, p. 639). The same Onsager-based framework leads to an

20  analogous conclusion that a concentration gradient will contribute to the sensible heat transfer and this is widely known as known as the Dufour effect (Bejan, 2016, p. 639). In applications (e.g., Hydrology, Agriculture, Ecology, Climate, etc.) it is a near universal practice to ignore the Onsager-based flux-coupling. Instead it is usually assumed that sensible heat transfer only depends on the temperature gradient and evaporation only depends on the concentration gradient (e.g., Monteith and Unsworth, 2008). From an open water surface the sensible heat transfer is usually small relative to the latent heat flux (i.e.,

25  the evaporative flux) but we are not aware of any estimates of the magnitude of the Soret effect under typical environmental conditions. This raises an important question of whether an approach that assumes evaporation is solely described by Fick's law is sufficiently accurate for applications in hydrology, agriculture, ecology and more generally for weather/climate studies. The aim of this technical note is to answer that question.

30   Ludwig (in 1856) first made a brief report noting the formation of a concentration gradient in the presence of a steady state temperature gradient in a liquid mixture. This initial observation was subsequently investigated in more detail using liquid solutions by Soret (in 1879). In liquids the phenomenon is now usually known as the Soret effect (or sometimes the Ludwig-Soret effect). In gas mixtures, the same thermodynamic phenomenon has often been called thermodiffusion and was predicted theoretically using the kinetic theory of gases (Chapman, 1916; Enskog, 1911) before the first experimental

35   confirmation (Chapman and Dootson, 1917). In essence, in the presence of a steady state temperature gradient there is a preferential sorting of the molecules with lighter molecules "diffusing" towards the hotter end of the gradient in the gas mixture. In air, which can be thought of as a mixture of dry air (equivalent molecular mass ≈ 29 g mol$^{-1}$) and water vapor (molecular mass ≈ 18 g mol$^{-1}$), the (lighter) water vapor molecules will tend to "diffuse" from colder to hotter regions. Hence when an evaporating surface is colder than the adjacent air we anticipate an additional mass flux due to the temperature

40   gradient. (Similarly, when the evaporating surface is hotter than the adjacent air we anticipate a reduction in the mass flux via the same phenomenon.) For the sake of brevity we refer to this thermal phenomenon as the Soret effect.

In the first half of the 20$^{th}$ century there was extensive interest in the Soret effect in gases because a comparison of the experimental results with theoretical calculations was routinely used to investigate the nature of the molecular collisions. A

45   comprehensive foundation text on the topic is available (Grew and Ibbs, 1952). In this paper we make use of experimental measurements of the Soret effect in gas mixtures to evaluate the magnitude of this thermal effect on evaporation relative to the traditional concentration-based ('Fickian') approach.

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| **Variable** | **Units** | **Description** |
|---|---|---|
| $J$ | mol m$^{-2}$ s$^{-1}$ | Flux density |
| $\rho$ | mol m$^{-3}$ | Molar density of the mixture. |
| $D$ | m$^2$ s$^{-1}$ | Ordinary mass diffusion coefficient |
| $D_T$ | m$^2$ s$^{-1}$ | Coefficient of thermal diffusion. |
| $x$ | mol mol$^{-1}$ | Mole fraction of target species (e.g., water vapor) in the mixture. |
| $T$ | K | Temperature of the mixture. |
| $k_T$ | (-) | Thermal diffusion ratio (dimensionless). |
| $\alpha_T$ | (-) | Thermal diffusion factor (dimensionless). |
| $E$ | mol m$^{-2}$ s$^{-1}$ | Evaporation from water body |

55 (row: $x$)

60   **Table 1   List of key variables**

## 2 Theory

In a binary gas mixture the diffusive flux $J$ (kg m$^{-2}$ s$^{-1}$) of the target species is given by (Grew and Ibbs, 1952),

$$J = \underbrace{[-\rho \ D \ \nabla x]}_{Fick's \ Law} - \underbrace{[\rho \ D_T \ \nabla(\ln T)]}_{Soret \ effect} \qquad , \qquad (1)$$

with $\rho$ (mol m$^{-3}$) the density of the mixture, $D$ (m$^2$ s$^{-1}$) the ordinary ('Fickian') mass diffusion coefficient, $x$ (mol mol$^{-1}$) the

65  mole fraction of the species of interest. $D_T$ (m$^2$ s$^{-1}$) is known as the coefficient of thermal diffusion with $T$ (K) the mixture temperature. Note that on this formulation the molar- and thermal-based diffusion coefficients have the same (classical) units for diffusivity of m$^2$ s$^{-1}$. We further note that in the recent literature it is common to define the "driving force" of thermodiffusion using $\nabla T$ (e.g., Ortiz de Zárate, 2019; Platten, 2006; Rahman and Saghir, 2014) instead of $\nabla(\ln T)$ as used in the original theory. As a consequence we note that in the above-cited modern literature the thermodiffusion coefficient has a

70  different physical meaning (and different units) from that in the original theory. Here we follow the original literature as per Eqn 1 (Chapman and Cowling, 1939; Grew and Ibbs, 1952) because nearly all of the key experimental work was completed before 1960 and was based on the classical formulation that uses $\nabla(\ln T)$ as the "driving force". On this approach we can make direct use of extensive (translated Russian) thermodynamic tables based on pre-1960 experiments (Vargaftik, 1983) as well as many useful graphical and tabulated summaries in the foundation textbook on the topic (Grew and Ibbs, 1952).

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Thermodiffusion depends primarily on the bulk composition of the mixture. For example, the obvious boundary conditions are that $D_T$ must equal zero when $x$ equals either zero or one since there can be no identifiable thermodiffusion in a pure substance. The classical approach is to define a dimensionless thermal diffusion ratio $k_T$ (Grew and Ibbs, 1952),

$$k_T = \frac{D_T}{D} \qquad . \qquad (2)$$

80  The dependence on bulk composition is incorporated by expressing $k_T$ as,

$$k_T = \alpha_T \ x \ (1 - x) \qquad , \qquad (3)$$

with $\alpha_T$ the dimensionless thermal diffusion factor. The (quadratic in $x$) form of Eqn. (3) is the simplest that captures the requisite boundary conditions ($k_T = 0$ when $x = 0$ or 1). Combining Eqns 1-3 we have,

85  $$J = \underbrace{[-\rho D \nabla x]}_{Fick's \ Law} - \underbrace{[\rho D \ \alpha_T \ x \ (1 - x) \frac{\nabla T}{T}]}_{Soret \ effect} \qquad . \qquad (4)$$

## 3 Empirical estimate of the thermal diffusion factor $\alpha_T$ for a H$_2$O-Dry air mixture

The most extensive database on thermodiffusion in gases that we have been able to locate includes some 12 pages of summarised experimental data in a set of (translated Russian) thermodynamic tables (Vargaftik, 1983, p. 654-665) that document experimental estimates of $k_T$ and $x$ for numerous binary gas mixtures. In addition there are useful experimental

90    data ($\alpha_T$) for a subset of the same binary gas mixtures in the foundation textbook on the topic (Grew and Ibbs, 1952). Surprisingly neither of these extensive data sources list a single experiment involving water vapor and we have not been able to locate an experiment involving water vapor elsewhere in the scientific literature. Instead, as described below, we use experimental data for gas mixtures that have very similar macroscopic properties to infer the appropriate value of $\alpha_T$ for the $H_2O$-Dry air mixture of primary interest.

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Foundation work by Chapman established that the molecular masses of the mixture components only influence the thermal diffusion ratio/factor by their ratios (Chapman, 1940). The two macroscopic (dimensionless) variables used to collate the various theoretical/numerical results are known as the proportionate mass difference $M$ ($= (m_1 - m_2) / (m_1 + m_2)$, with $m_1$ the molecular mass of the heavier component) and the proportionate diameter difference $\Sigma$ ($= (\sigma_1 - \sigma_2) / (\sigma_1 + \sigma_2)$, with $\sigma_1$ the
100   diameter of the heavier component) defined by the collision diameters ($\sigma$) of the mixture components (Chapman, 1940; Grew and Ibbs, 1952). In general, the thermal diffusion factor $\alpha_T$ increases with $M$ and $\Sigma$, but is much more sensitive to $M$ than to $\Sigma$ (Grew and Ibbs, 1952, Fig. 7, p. 29). The values of $M$, $\Sigma$ for three binary gas mixtures ($N_2$-$CO_2$, $N_2$-$N_2O$, $H_2$-$CO_2$) for which we have extensive experimental data (Grew and Ibbs, 1952; Vargaftik, 1983) are listed in Table 2 along with relevant values for a $H_2O$-Dry air mixture for which we have no experimental data. We note that the values of $M$, $\Sigma$ for the $H_2O$-Dry
105   air mixture are more or less the same as those for the two $N_2$-based mixtures. The experimental data for the three binary gas mixtures are shown in Fig. 1.

| Gas Mixture | $M$ | $\Sigma$ |
|---|---|---|
| $N_2$-$CO_2$ | 0.22 | 0.08 |
| $N_2$-$N_2O$ | 0.22 | 0.08 |
| $H_2$-$CO_2$ | 0.91 | 0.46 |
| $H_2O$-Dry air | 0.23 | 0.07 |

**Table 2: Dimensionless variables ($M$, $\Sigma$; see main text) for four binary gas mixtures. Calculations for $\Sigma$ use the following values for the collision diameter: $\sigma(N_2) = 3.7 \times 10^{-10}$ m, $\sigma(CO_2) = 4.3 \times 10^{-10}$ m, $\sigma(N_2O) = 4.3 \times 10^{-10}$ m, $\sigma(H_2) = 1.6 \times 10^{-10}$ m, $\sigma(Dry air) = 3.7 \times$**
110   **$10^{-10}$ m that have been calculated using an empirical formula dependent on the molecular mass (Wang and Frenklach, 1994, see their Eqn 6).**

The experimental results highlight the strong dependence of $k_T$ on the bulk mixture composition (Fig. 1a). With data for $k_T$ and the mole fraction (of $N_2$ and $H_2$) available in the tables we calculate the dimensionless thermal diffusion factor $\alpha_T$ for all
115   three gas mixtures (Fig. 1b). The results show that the experimental values of $\alpha_T$ for the two $N_2$-based gas mixtures are more or less the same (Mean $\approx 0.05$) (Fig. 1b). The experimental values of $\alpha_T$ summarised by Grew and Ibbs (1952, p. 130) for the $N_2$-based mixtures are consistent with those given here (Fig. 1b). In contrast, the experimental values of $\alpha_T$ for the $H_2$-$CO_2$ mixture are much larger as expected based on the large difference in molecular mass between the components in that mixture

(Grew and Ibbs, 1952, see their Fig. 8 on p. 30). In the absence of any contrary evidence, we assume that the experimental

120 data for the two $N_2$-based gas mixtures can be used for the $H_2O$-Dry air mixture. We know by experiment that $\alpha_T$ usually

increases slightly with $T$ and the empirical equation of Youssef et al. (1965, their Eqn 7) for the $N_2$-$CO_2$ mixture implies that

the value of $\alpha_T$ would be ≈ 0.048 if adjusted back to a mean $T$ of 300 K. In the context of the magnitude of the Soret effect

relative to concentration-dependent diffusion we show below that it is not necessary to consider the $T$ dependence because

the Soret effect is relatively small. With that we adopt $\alpha_T = 0.05$ for the $H_2O$-Dry air mixture and use that value in the next

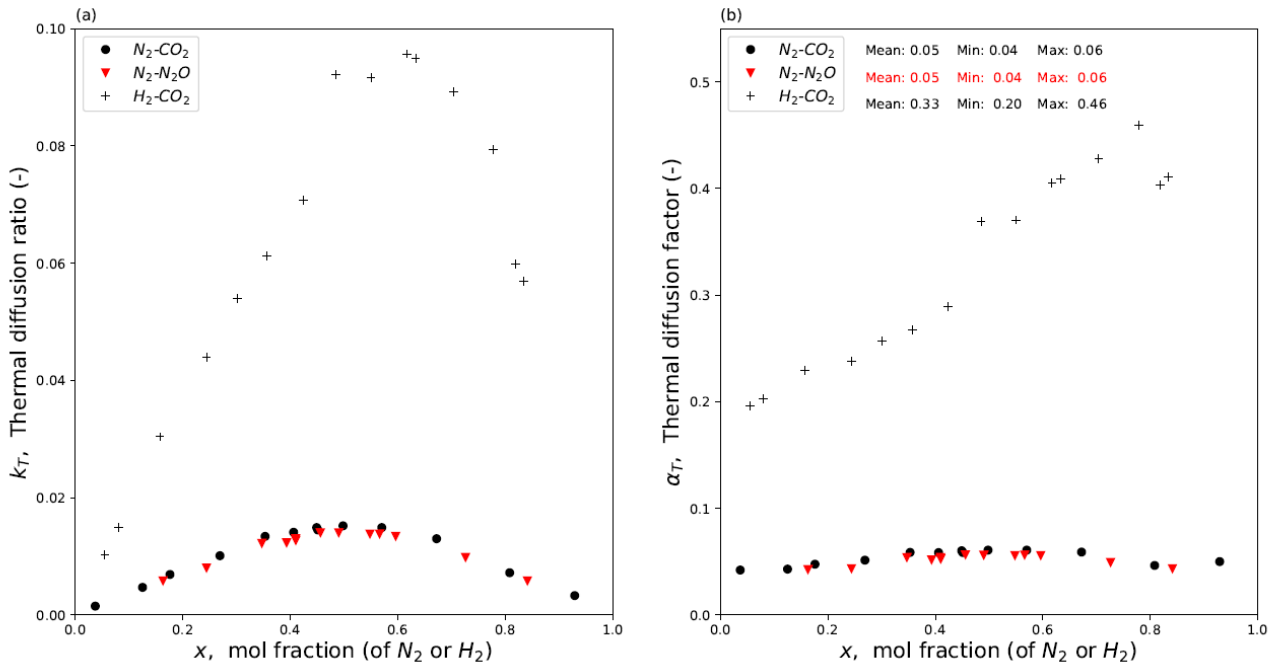
125 section to evaluate the magnitude of the Soret effect on evaporation relative to the effect of concentration-dependent

diffusion.

**Figure 1: Experimental data for three binary gas mixtures at a mean $T$ of 328 K (Vargaftik, 1983), p. 661). Composition dependence of the (a) thermal diffusion ratio ($k_T$) and (b) the thermal diffusion factor ($\alpha_T$, calculated per Eqn 3).**

130 **4 Sensitivity of evaporation to the Soret effect**

We begin by rewriting Eqn 4 with $\alpha_T = 0.05$ and we use the typical hydrologic sign convention (evaporation is positive

upwards from the liquid surface) with the evaporation rate ($E$) given by,

$$E = \frac{\rho\,D}{\Delta z}\left[(x_s(T_s) - x_a) - 0.05\,\bar{x}\,(1 - \bar{x})\,\frac{(T_s - T_a)}{\bar{T}}\right] \qquad , \qquad (5)$$

with $x_s(T_s)$ the (saturated) mole fraction of water vapor at the evaporating surface of temperature $T_s$, $x_a$ the mole fraction of

135 water vapor in air having temperature $T_a$, $\bar{x}$ is the mean mole fraction ($= (x_s(T_x) + x_a)/2$) of water vapor over the diffusive

pathway of thickness $\Delta z$ and $\bar{T}$ ($= (T_s + T_a)/2$) the mean $T$ over the same pathway. Note that we will have a positive Soret effect when $T_s < T_a$.

To take a typical numerical example, assume standard air at 298 K and relative humidity of 60% with total air pressure of 1 bar where the surface $T$ is initially 5 K cooler than the air. With those data (Table 3) we have,

$$E = \frac{\rho\,D}{\Delta z}\left[(0.0232 - 0.0189) - \left((0.05)(0.0211)(0.9789)\left(\frac{293-298}{295.5}\right)\right)\right] \quad , \tag{6a}$$

which equals,

$$E = \frac{\rho\,D}{\Delta z}[0.0043 + 0.000017] \quad . \tag{6b}$$

In this numerical example, concentration-dependent mass diffusion contributes 99.6% of the total flux with the Soret effect contributing 0.4% to the total. We could enhance the percentage due to the Soret effect by holding the air properties constant while decreasing the surface $T$ until, for example, $x_s(T_s)$ equals $x_a$. In that limit the mole fraction gradient would become zero and Soret effect would then be 100% of the total flux. However, that total flux would be vanishingly small as we show below.

| Variable | Value | Units |
|---|---|---|
| $x_s(T_s)$ | 0.0232 | mol mol$^{-1}$ |
| $x_a$ | 0.0189 | mol mol$^{-1}$ |
| $\bar{x}$ | 0.0211 | mol mol$^{-1}$ |
| $T_s$ | 293 | K |
| $T_a$ | 298 | K |
| $\bar{T}$ | 295.5 | K |

**Table 3 Data for numerical example: standard air at 298 K and rel. humidity = 60%, $T_s$ = 293 K**

To make a more comprehensive assessment of the magnitude of the Soret effect we use a recently published experimental database on evaporation (Roderick et al., 2023). The database includes 70 individual evaporation experiments made under carefully controlled laboratory conditions with $E$ measured using an accurate balance. The experiments encompass a broad range of typical environmental conditions ($T$ range from 15 to 45°C, relative humidity range from 20 to 78%, wind speed ($U$) range from 0.5 to 4 m s$^{-1}$). One important feature of the experiments is that the air properties ($T$, humidity, wind) were held constant during each evaporation experiment which meant that the water bath from which evaporation occurred was a very good approximation to the classical theoretical wet bulb thermometer. As a consequence the water bath was generally colder than the adjacent air which would tend to maximise the (positive) contribution of the Soret effect.

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To analyse each experiment, the approach was to use Eqn 5 to first calculate the value of $\Delta z$ that gave the observed evaporation with the mass diffusion coefficient for water vapor in air calculated at the mean temperature. With that calculated value of $\Delta z$ we were then able to separate the observed evaporation into two terms as follows,

$$E = E_M + E_T \qquad . \qquad (7)$$

165 The first term ($E_M$) is the component of the evaporation due to classical concentration-dependent mass diffusion and the second term ($E_T$) is due to the Soret effect. The results are converted to a mass basis of expression and shown in Fig. 2.
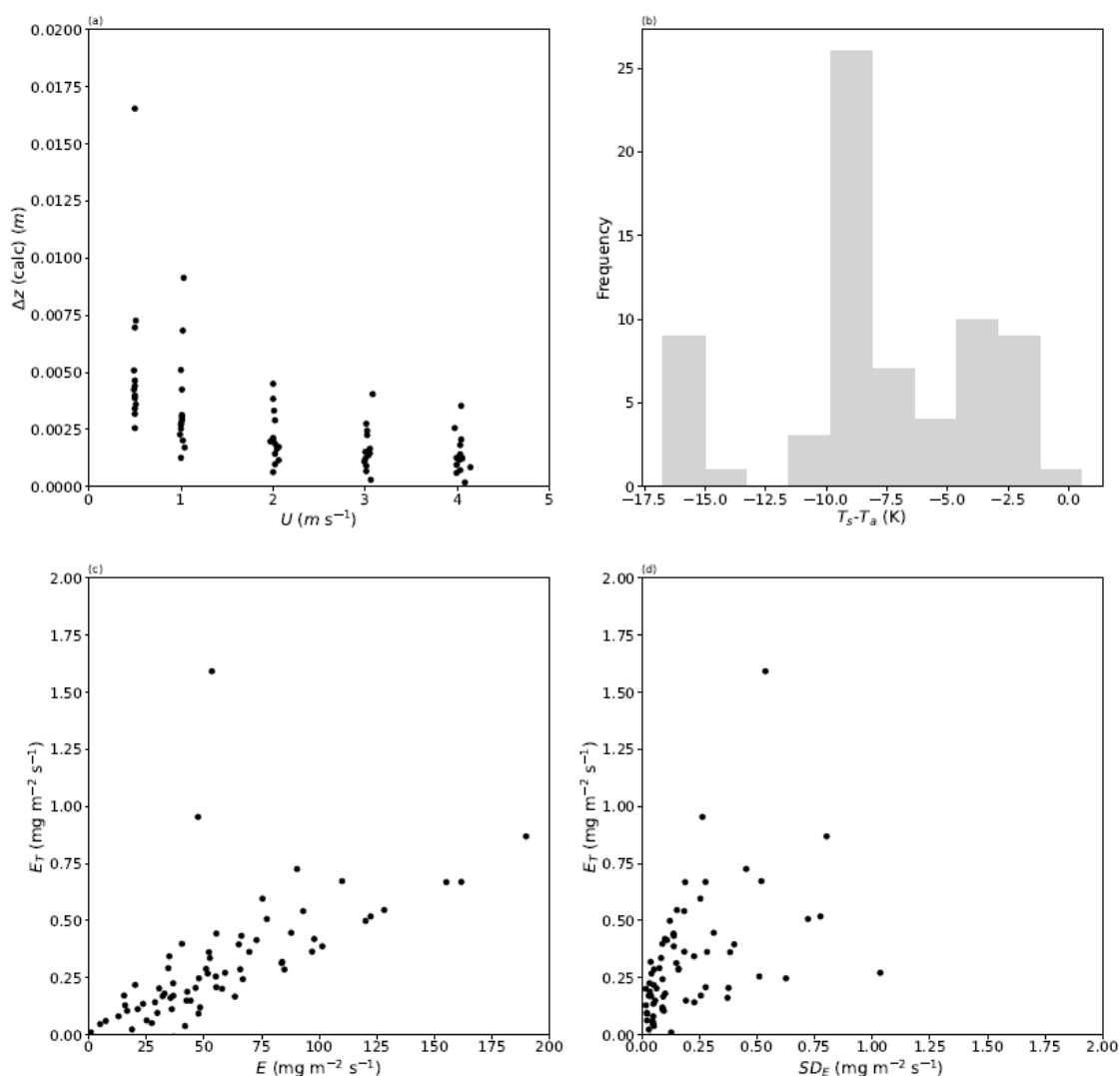
**Figure 2 Relative magnitude of the Soret effect on evaporation in 70 individual laboratory-controlled evaporation experiments. (a) Calculated value of the boundary layer thickness (see main text) as a function of windspeed and (b) histogram showing the**

170  **variation in $T_s$-$T_a$, over the 70 individual evaporation experiments. Component of the evaporative flux due to thermodiffusion ($E_T$) vs (c) the total evaporative flux ($E$) and vs (d) the experimental error in the total evaporative flux ($SD_E$).**

The (calculated) values of the boundary layer thickness (Fig. 2a) decline with wind speed as expected (Lim et al., 2012) and in all but one of the 70 experiments, the evaporating liquid water surface was colder than the adjacent air; by up to ~ 17°C in a few instances (Fig. 2b). We reiterate that these conditions maximise the (positive) contribution of the Soret effect to the

175  overall evaporative flux. Despite that, we find the component of the evaporative flux due to the Soret effect is typically two orders of magnitude smaller than the ordinary mass diffusion component (Fig. 2c). The evaporation was measured using an accurate balance under carefully controlled laboratory conditions with very small errors (up to a maximum error of 1 mg m$^{-2}$ s$^{-1}$) compared with many field-based evaporation measurements. Despite that, the thermodiffusive flux is about the same magnitude as the measurement error (Fig. 2d) implying that the Soret effect would not be measurable under typical field

180  conditions. Note that the maximum measured evaporative flux in the experimental program was ~ 190 mg m$^{-2}$ s$^{-1}$ (Fig. 2c) (~ 456 W m$^{-2}$ in climate science units and ~ 10 mmol m$^{-2}$ s$^{-1}$ in plant physiology units) while the maximum measurement error in $E$ was ~ 1 mg m$^{-2}$ s$^{-1}$ (Fig. 2d) (~ 2.4 W m$^{-2}$ in climate science units , ~ 0.056 mmol m$^{-2}$ s$^{-1}$ in plant physiology units).

## 4 Discussion and Conclusions

185  The Soret effect is a real phenomenon and played a prominent role in the overall development of the kinetic gas theory (Chapman and Cowling, 1939; Grew and Ibbs, 1952). However, we were unable to locate a single experiment involving water vapor and instead we assumed that the thermal diffusion factor for a $H_2O$-Dry air mixture was the same as for other well studied binary gas mixtures ($N_2$-$CO_2$, $N_2$-$N_2O$) (Fig. 1) having similar relative differences in molecular mass between the components (Table 2). Our experimentally-based result was a numerical value of 0.05 for the thermal diffusion factor in

190  a $H_2O$-Dry air mixture. Recent work has also examined the impact of the Soret effect on evaporation but used a numerical value of 0.5 for the (same) thermal diffusion factor (Griffani et al., 2024). This order of magnitude difference in the thermal diffusion factor requires an explanation.

The Griffani et al. (2024) result is incorrect for two reasons. First, it was based on Landau's original theoretical derivation

195  which assumed a binary gas mixture having perfect elastic collisions where the molecular mass of the heavier molecule was assumed to be substantially larger than that of the lighter molecule (Lifshitz and Pitaevskii, 1981, p. 36). The latter assumption implies a value for the proportionate mass difference $M$ of 1 which is much larger than the relevant value for a $H_2O$-Dry air mixture ($M$ = 0.23, Table 2). Secondly, real gas mixtures generally have a substantially lower thermal diffusion factor than that calculated using the perfect elastic collision assumption (Grew and Ibbs, 1952). In fact, Grew and Ibbs

200  (1952, p. 128) note that the experimental thermal diffusion factor for the $H_2$-$CO_2$ binary gas mixture considered here (Fig. 1, Table 2) is around 47% of the value computed using the perfect elastic collision assumption. Given that the porportionate

mass difference for the $H_2$-$CO_2$ binary gas mixture is close to 1 (Table 2, $M$ = 0.91), then a reasonable semi-empirical estimate for the thermal diffusion factor for the $H_2$-$CO_2$ binary gas mixture would be (= 0.47 × 0.5 = ) 0.24. At a low concentration of the lighter gas ($H_2$) the actual experimental results for the $H_2$-$CO_2$ binary gas mixture give the thermal

205 diffusion factor of around 0.2 (Fig. 1b) that is very close to the semi-empirical value calculated above. Hence because of the underlying assumptions the Griffani et al. (2024) estimate for the thermal diffusion factor is not relevant to a $H_2O$-Dry air mixture.

With that, our results using a thermal diffusion factor of 0.05 show that under typical environmental conditions the Soret

210 effect is at least two orders smaller than the classical concentration-dependent mass diffusion (Fig. 2c) and can be safely ignored in evaporation studies. This result justifies the long term hydrologic practice of simply assuming that evaporation follows Fick's law.

Under typical environmental conditions the Soret effect is also small in liquids. Despite that, there is current interest in

215 evaluating whether the Soret effect can be used to desalinate water (Xu et al., 2024). That approach requires clever engineering by recycling the treated stream multiple times to eventually separate the salt from the fresh water (Xu et al., 2024). Hence despite the fact that the Soret effect is small under typical environmental conditions it may still have important future engineering applications.

**Author Contribution**

220 MLR conceived the overall project. MLR and CJS undertook the analysis. MLR prepared the manuscript with contributions from CJS.

**Data Availability**

The evaporation experiment data is available at https://doi.org/10.5281/zenodo.8381685.

**Competing Interests**

225 The authors declare that they have no competing interests.

**Acknowledgements**

We thank Pierre Rognon and Graham Farquhar for discussions on the Soret effect. The research was supported by the Australian Research Council (DP190100791).

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