

Kowalski's comment presents a conflict between the conservation of momentum and Fick's 1<sup>st</sup> law of diffusion. The point is taken by a simple case of trace amount of argon diffusing in the mixture of dioxygen (O<sub>2</sub>) and triatomic ozone (O<sub>3</sub>). An example given is about the situation that the description of diffusion based on the mass fraction includes no argon gradient and thus no argon transport (in the inertial frame of co-ordinates i.e. with respect to stationary axes) whereas the description for the same situation, but based on the molar fraction, includes the gradient and thus diffusion.

The comment by Kowalski's is "shooting off". The system studied is the ternary system and it has been well-known since 1949 that for the ternary system one should not use Fick's 1<sup>st</sup> law. The very good approximation is Maxwell-Stefan equations (aka Stefan-Maxwell equations). They are (the following text is directly copied from Vilà-Guerau de Arellano et al. (Biogeosciences 22, 6327-6341, 2025)

### 2.3 Stefan-Maxwell equations

Stefan-Maxwell equations provide a framework to study the dependence of the flux as a function of the gradients of other of all other species. For a multicomponent system consisting of  $n$  chemical species, the relationship between the one-dimensional mole fraction gradient of species  $\alpha$  and the species flux densities is given by the Stefan-Maxwell equation (Curtiss and Hirschfelder, 1949; Kalkkinen et al., 1991; Bird et al., 2007):

$$\begin{aligned} \frac{\partial x_\alpha}{\partial l} &= \sum_{\beta=1, \beta \neq \alpha}^n \frac{x_\alpha F_\beta - x_\beta F_\alpha}{c D_{\alpha\beta}} = \sum_{\beta=1, \beta \neq \alpha}^n \frac{x_\alpha f_\beta - x_\beta f_\alpha}{c D_{\alpha\beta}} \\ &= \sum_{\beta=1, \beta \neq \alpha}^n \frac{x_\alpha x_\beta (u_\beta - u_\alpha)}{D_{\alpha\beta}}, \end{aligned} \quad (14)$$

where  $D_{\alpha\beta}$  is the binary diffusivity of species  $\alpha$  and  $\beta$ , and the summations are over all species  $\beta$  other than species  $\alpha$ .

Here  $c$  is the total molar concentration of the mixture.

For the ternary system in Kowalski's comment we obtain

$$\frac{\partial x_A}{\partial l} = \frac{x_A F_{O_2} - x_{O_2} F_A}{c D_{AO_2}} + \frac{x_A F_{O_3} - x_{O_3} F_A}{c D_{AO_3}}$$

where subscript A refers to argon, O<sub>2</sub> to dioxygen and O<sub>3</sub> to ozone. For the case of no argon transport we can set  $F_A = 0$  which leads to

$$\frac{\partial x_A}{\partial l} = \frac{x_A F_{O_2}}{c D_{AO_2}} + \frac{x_A F_{O_3}}{c D_{AO_3}}$$

Thus the situation that no argon is transported means that there exists the gradient of argon molar fraction. The net transport is still zero as the diffusive transport is counter-balanced by the Stefan flow (ternary effect) induced by dioxygen and ozone transports.

I ask Kowalski to read Sections 17.7 – 17.9 in Bird et al. (Transport Phenomena, 2007, John Wiley and Sons). In addition, instead of presenting controversial new theory for

diffusion and asking others to disprove it, it would be more fruitful to find a flaw in the derivation of the Maxwell-Stefan equations by Curtiss and Hirschfelder (J. Chem. Phys. 17, 550-555, 1949) and disprove it. I try to be constructive but I am sorry if I am too destructive.