

We would like to thank the anonymous reviewer for their helpful feedback and comments. The following sections will address their comments in more detail. Author responses are in italics and updates to the manuscript text are marked in red.

1. On line 130 or elsewhere, it may be good to briefly describe how the partial molar area of each component is determined in the studied systems.

5 *The text has been updated on line 159-164 of the revised manuscript to include the following definition of partial molar area:*

For a surface phase represented by a concentric shell on a spherical droplet core, \mathcal{A}_i is defined as:

$$\mathcal{A}_i = \left(\frac{\partial A}{\partial n_i^s} \right)_{T,P,n_j^s,\sigma} = \gamma_i^s \frac{2r_p}{2\delta^s r_p - \delta^{s2}},$$

10 where A is the outer surface phase area, n_i^s and denote the molar amount and molar volume of i , respectively, in the finite-depth surface phase. r_p is the particle radius, i.e. the outer radius of the surface phase and δ^s is the radial thickness of the surface. For a derivation of Eq. (9), see Schmedding and Zuend (2023).

and on line 187:

Note that for a 2-D LL interface $\mathcal{A}_i^{\alpha\beta} = 2\gamma_i^\alpha / r^\alpha$.

- 15 2. On lines 243-247, it could be better to have these equations as displayed, numbered equations on separate lines, as it could be difficult for a general reader to easily parse these in-line equations.

The text has been updated on lines 275-280 of the revised manuscript as follows:

... it is assumed that both phases contribute material proportionally to the total amount of surface molecules, n^s , such that

$$n_j^s = q_j^\alpha \varepsilon_j n_j^{\text{tot}} + (1 - q_j^\alpha) \varepsilon_j n_j^{\text{tot}}$$

20 where q_j^α is the fraction of neutral component j in phase α , $(1 - q_j^\alpha)$ is the fraction of j in phase β , ε_j is the ratio of j in the surface phase to the total amount of j in the particle phase or n_j^s / n_j^{tot} , and n_j^{tot} is the total amount of species j in the liquid phases plus the surface phase while excluding any amount of species j in the gas phase. Similarly, for electrolyte component el , the contributions from phases α and β can be written as follows:

$$n_{el}^s = q_{el}^\alpha \varepsilon_{el} n_{el}^{\text{tot}} + (q_{\text{lim},el}^\alpha - q_{el}^\alpha) \varepsilon_{el} n_{el}^{\text{tot}},$$

25 where $q_{\text{lim},el}^\alpha \dots$

3. In Fig 3, Equation 15 could be referred to in the caption for the reader to be reminded of how $\sigma_{s,*}$ is defined.

Lines 264 - 265 of the revised manuscript have been updated to clarify the definition of σ^{s} :*

$$\sigma_i^{s*} = \frac{RT}{\mathcal{A}_i^s} \ln \left(\frac{a_i^s}{a_i^b} \right) + \sigma_i^o + \sigma^{ls} \frac{\mathcal{A}_i^b}{\mathcal{A}_i^s}.$$

Note that σ^s is now defined as $\sigma^s = \sigma^{s*} - \sigma^{ls}$; when σ^{ls} is negligible, $\sigma_i^{s*} = \sigma_i^s$.

30 *And the caption of Figure 3 has been updated to the following:*

Predicted values of σ^{s*} , which represent the measurable effect of expanding a finite depth surface phase as defined in Eq. (23); σ^s ; $\sigma^{\alpha\beta}$; and σ^{ls} (see legend)...

References

- 35 Schmedding, R. and Zuend, A.: A thermodynamic framework for bulk–surface partitioning in finite-volume mixed organic–inorganic aerosol particles and cloud droplets, *EGUsphere*, 2023, 1–42, <https://doi.org/10.5194/egusphere-2023-336>, 2023.