We would like to thank the anonymous reviewer for their helpful feedback on this manuscript. 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.

Scientific points from referee #1:

1. Line 90 gives a constraint on the value of ϕ, but is there any independent method of estimating ϕ independent of comparison with experimental values of $\sigma\alpha\beta$, or is this essentially a purely empirical factor? Some additional discussion of this parameter would he helpful.

The following text has been added following Eq. 3 on lines 93-101 of the revised manuscript to better define and describe the role of ϕ:

The value of ϕ can be defined in the case of binary systems with molecules 1 and 2 of approximately equal size and spherical shape in a hexagonal arrangement as

$$\phi = \frac{d_{1,1}d_{2,2}}{d_{1,2}^2},\tag{1}$$

where d is the equilibrium distance between the two molecules denoted in the subscript (Girifalco and Good, 1957). In the case of binary systems with molecules of unequal sizes or shapes, the value of ϕ could be approximated by the following equation:

$$\phi = \frac{4\mathscr{V}_1^{1/3}\mathscr{V}_2^{1/3}}{\left(\mathscr{V}_1^{1/3} + \mathscr{V}_2^{1/3}\right)^2}.\tag{2}$$

where $\mathscr{V}$ is the molar volume of 1 or 2 respectively (Girifalco and Good, 1957). Qualitatively, ϕ is described as being lower in systems where the the primary molecular interactions are different, for example dispersion forces in phase β and hydrogen bonding in phase α (Girifalco and Good, 1957).

2. What a Guggenheim interface is, which is mentioned on lines 124 and 205, should be briefly explained.

The following text has been added to line 143-149 of the revised manuscript to better define a Guggenheim interface:

Guggenheim interfaces are an alternative approach to the classical Gibbsian treatment of interfaces, the latter treating them as infinitely thin two-dimensional surfaces located at the point where the excess concentration of some species in the system, typically the solvent, is zero (Gibbs, 1906). Guggenheim interfaces instead treat the surface of a solution as a thermodynamically distinct region from the bulk liquid phase beneath it and the gas phase above it (Guggenheim, 1940). Such a definition allows for more flexibility in defining the location of the interface, its extent and composition, as well as accounting for the feedback of bulk–surface partitioning on the bulk composition in small droplets.

Minor presentation points from referee #1:

1. The first paragraph of the results section, lines 287-297 do not really seem to contain results, but rather some definitions and background information that would benefit the reader to see earlier in the manuscript. I would suggest that this section/information be moved to the introduction section.

The described paragraph as been shifted to the final paragraph of the introduction section.

2. In the paragraph starting on Line 320, the authors switch back and forth between referring to the different approaches by name and by equation number. For the sake of the reader, it would be helpful to stick to one consistent naming throughout the manuscript, rather than switching back and forth. Related, the shorthand 'Butler equation' is used on line 338 for 'Butler equation with geometric mean activity coefficients' (line 323), but the figure just uses 'Geom. Mean. Act. Coeffs.' Consistency would improve clarity.

Equation references have been updated throughout the text to consistently use the common name of the equation followed by the equation number in parenthesis.

References

40   Gibbs, J. W.: Thermodynamics, vol. 1, Longmans, Green and Company, 1906.

Girifalco, L. A. and Good, R. J.: A Theory for the Estimation of Surface and Interfacial Energies. I. Derivation and Application to Interfacial Tension, The Journal of Physical Chemistry, 61, 904–909, https://doi.org/10.1021/j150553a013, 1957.

Guggenheim, E.: The thermodynamics of interfaces in systems of several components, Transactions of the Faraday Society, 35, 397–412, https://doi.org/https://doi.org/10.1039/TF9403500397, 1940.