Second revision

[Comment from Anonymous Referee 1] The authors have addressed most of my concerns in this revision. However, I still find that proper credit is not given to the authors of the surface tension data that underlies this work. If data was used in this study, the original authors deserve a citation of their paper.

[Our answers] We thank the anonymous reviewer for their second revision. We have addressed their points as described in the following.

E.g. 1 - Line 269, 306, and Fig 3 still seem to imply the underlying surface tension data is reported by El Harber et al 2024, without giving credit to the authors who actually collected and reported the underlying data.

In order to create Fig. 3, a total number of 149 different sources were used. We think that it is more convenient to cite the review where all this data has been compiled instead of citing all these papers individually. To better emphasize that El Haber et al. 2024 did not do the measurements but just reviewed the data, we adjusted the sentences as follows:

Besides its functional groups, the surfactant model compound should be representative in its surface-active behaviour. As described in Sect. 2.1, surfactants can be characterized by their separation factor in water S_{1i} and their pure component surface tension σ_i. By fitting the binary Eberhart model (Eq. 3) to experimental surface tension data ~~reported in~~ compiled by El Haber et al. (2024), S_{1i} and σ_i were determined for 76 organic substances, which are shown in Fig. 3. The fitting procedure is described in supplement Sect. S6 and the data underlying Fig. 3 are provided in tabular form in supplement Sect. S6 and as a csv file (see code and data availability). In addition to these 76 organic compounds, S_{1i} and σ_i of atmospheric samples taken at five different locations were considered. Ekström et al. (2010) and Gérard et al. (2016) measured surface tension isotherms

The pure component surface tension σ_i and the binary separation factor in water S_{1i} of all substances used in this study are summarized in Table 2 (see supplement Sect. S8 for underlying experimental data and model fits). For water, $\sigma_1 = 72.0\,\mathrm{mN\,m^{-1}}$ was used throughout the study. For the six model substances (propionic acid, glutaric acid, valeric acid, pinonic acid, SDS, and oleic acid) used in the surfactant category (2), σ_2 and S_{12} are taken from the fits underlying Fig. 3. The pure liquid surface tension of glucose at room temperature is not known, since glucose crystallizes at this temperature. The binary aqueous solution data from Aumann et al. (2010), Lee and Hildemann (2013), and Romero and Albis (2010) compiled by El Haber et al. (2024) is only available in a narrow concentration range. An extrapolation to supersaturated concentrations with the Eberhart model yields a very high value ($\sigma_3 > 10000\,\mathrm{mN\,m^{-1}}$) with a large uncertainty (90 % confidence interval:

We also adjusted the caption of Figure 3 as follows:

Figure 3. Separation factor in water S_{1i} and pure component surface tension σ_i of organic substances (stars and 1–70: based on data ~~from~~ compiled by El Haber et al. (2024)) and of atmospheric samples taken at 5 different locations (71–74: Ekström et al. (2010), a–k: Gérard et al. (2016), see also supplement Sect. S6 – S8). Substances with black stars as markers are used as model compounds in this study. S_{1i} was determined by fitting the binary Eberhart model (Eq. 3) to experimental surface tension data. If σ_i was not reported in El Haber et al. (2024), it was considered an additional fitting parameter. For the atmospheric samples, σ_i was taken as the lowest measured value. Uncertainty bars show the 95 % confidence intervals of the fit parameters. Substance names and categories are the same as in El Haber et al. (2024), i.e., the category "alcohols" also contains ketones and aldehydes. Grey lines and labels of approximate regions of weak, intermediate, and strong surfactants show the suggested categorization following the results of this study.

E.g. 2 - Fig S10 now includes references to the original datasets where the underlying surface tension data was found, but those references are not included in the reference list in the SI. If

the dataset is used in this work, the manuscript from which it was taken needs to be properly referenced in the bibliography. A reader of this manuscript should not have to go to a different paper to find the references for the data shown in the figures.

We thank the reviewer for pointing that out. We have now added these references to the reference list in the SI.

Other additional small comments:
1. line 132 - a kinetic partitioning model has also recently been validated against microscopic data (https://doi.org/10.1021/acsearthspacechem.4c00199)

We thank the reviewer for pointing to this very recent study. Knowing about it, we can no longer write that the Monolayer model is the only model validated with experimental surface tension data, and so we have adjusted the paragraph as:

In large liquid volumes, the bulk composition x_i^{bulk} can be assumed to be equal to the total composition x_i^{tot} (i.e., $x_i = x_i^{\mathrm{bulk}} \approx x_i^{\mathrm{tot}}$) in surface tension isotherms (e.g., Eberhart model). Small droplets, however, have a large surface-to-volume ratio and therefore the partitioning of substances to the surface of the droplet can lead to their depletion in the droplet bulk. To take
130 this effect into account, a partitioning model is required that introduces mass conservation and allows to quantify the bulk depletion based on physical and geometrical assumptions. ~~Among the partitioning models described in literature,~~ Here, we choose the Monolayer model (Malila and Prisle, 2018) ~~is the only one that~~ for modelling bulk–surface partitioning, which has been validated with microscopic surface tension data ~~(Bzdek et al., 2020; Bain et al., 2023). Therefore, in this study, this model is chosen for bulk–surface partitioning.~~ in several studies (Bzdek et al., 2020; Bain et al., 2023, 2024).

2. check 'Monolayer model' is consistently capitalized

We thank the reviewer for spotting this. We have double-checked the spelling of ‘Monolayer model’.