Author Comments (Manuscript ID: acp-2010-732)

We thank Anonymous Referee #2 for the valuable and supportive comments. We here respond to the comments and revise our manuscript accordingly.

Referee #2

The authors perform molecular dynamics (MD) simulations of water and water-glycine droplets to determine the curvature dependence of surface tension. The obtained surface tension is then used in Köhler theory to calculate critical supersaturations and the results are compared to recent measurements. Such a study is justified because the amino acids may act as cloud condensation nuclei in marine areas. This work parallels to a previous work by the authors (Li et al., 2010) where cis-pinonic acid-water clusters were studied. In the present paper the same simulation methods are used but the rigid SPC/E water model is replaced by a semi-flexible SPC/E model. I find the paper well written, interesting, and relevant to atmospheric sciences. The methods and argumentation are scientifically sound. I can recommend publication after the authors consider the following items and correct some misprints.

1. Comment: *If surface tension is needed in a thermodynamic theory (like Köhler theory), it is preferable to use a thermodynamically defined surface tension. Unfortunately, surface tension for spherical interfaces is not a uniquely defined quantity. A good choice is the surface tension related to the surface of tension. Then the relevant thermodynamic machinery, that is Laplace equation, Gibbs adsorption equation, etc., is valid. Equation 9 would give this surface tension, if a)* R_e *were the radius of the surface of tension* R_s *and b) W corresponded to the true thermodynamic free energy barrier to nucleation. In eq. 9 neither case is true.* R_e *is the radius of the equimolar surface and W is calculated using the Irving-Kirkwood pressure tensor, which results in wrong energy barrier (see ten Wolde and Frenkel, J. Chem. Phys. 109, 9901 (1998)). Unfortunately, it is very difficult to obtain* R_s *and the correct energy barrier from MD simulations, and therefore the approximations in the paper are acceptable, especially as the clusters are quite large (N>750). The authors should nevertheless highlight the fact that only an approximation of a thermodynamic cluster surface tension is obtained.*

Our response: We thank the referee for pointing out this. We agree with the fact that R_e is not exactly equal to R_s and that the work of formation calculated from the Irving-Kirkwood pressure tensor is not exactly equal to the energy barrier of nucleation. However, as the referee suggested, for large clusters such errors are expected to be sufficiently small. We have added statements in the manuscript emphasizing the approximation we used in computing the surface tension.

2. Comment: *The curvature-corrected surface tension brings the critical supersaturations a bit* *closer to the experimental values. Nevertheless, I think it would be best to mention in the conclusions or in the end of section 4 that there are probably other factors causing the discrepancy in addition of surface tension (Kristensson et al., 2010).*

Our response: We thank the referee for this point. As mentioned by Kristensson et al. in their paper (J. Phys. Chem. A, 114, 379–386, 2010), the discrepancy between the experimental results and the Köhler model arises from a combination of factors including surface tension, nonideality, and uncertainties in the density of glycine. In our study, the surface tension correction to the critical supersaturation is calculated to be small, therefore other factors cannot be ruled out. We have added statements in the manuscript highlighting the importance of other factors.

3. Comment: *On page 4 there are discussion on previous simulation studies of droplets. The authors might want to add the following recent papers to the references list: B. J. Block et al., J. Chem. Phys. 133, 154702 (2010) and J. Julin et al, J. Chem. Phys. 133, 044704 (2010).*

Our response: We thank the referee for this. We have added these two papers in the reference list.

4. Comment: *The density profiles from planar and spherical simulations are fitted to hyperbolic tangent function. Some authors prefer error function. Have the authors checked if fitting to an error function form would change the results? I find it conceivable that the location of equimolar surface might be affected.*

Our response: We thank the referee for pointing out this. We have checked the results by fitting to the error function and updated Table 2, Table 3 and Table 4 with supplemented data. It is found that the influences on R_e and σ are no more than 0.01 nm and 0.1 mJ m^{-2}, respectively. The use of the error function only introduces a very slight change in the fitted densities. Therefore we conclude that the difference between the results obtained from the hyperbolic tangent function and the error function is negligible.

Table 2. Calculated liquid density, radius for equimolar dividing surface, work of formation, and surface tension for spherical interfaces in systems 1–6. Shown in parentheses are the data calculated by fitting to the error function.

System	ρ_α / nm^{-3}	R_e / nm	W / 10^{-19} J	σ / mJ m^{-2}
1	34.636 (34.589)	1.73 (1.73)	7.61	60.78 (60.72)
2	34.614 (34.569)	1.90 (1.90)	9.74	64.16 (64.09)
3	34.469 (34.430)	2.18 (2.18)	13.73	68.83 (68.77)
4	34.357 (34.328)	2.40 (2.41)	16.91	69.84 (69.79)
5	34.267 (34.238)	2.75 (2.76)	22.55	70.96 (70.90)
6	34.187 (34.161)	3.27 (3.27)	31.38	70.13 (70.08)

Table 3. Calculated liquid density, radius for equimolar dividing surface, work of formation, and surface tension for spherical interfaces in systems 7–10. Shown in parentheses are the data calculated by fitting to the error function.

System	ρ_α / nm^{-3}	R_e / nm	W / 10^{-19} J	σ / mJ m^{-2}
7	33.566 (33.531)	1.93 (1.93)	9.46	60.66 (60.61)
8	33.506 (33.478)	2.43 (2.43)	16.33	65.88 (65.83)
9	33.377 (33.352)	2.79 (2.79)	22.66	69.60 (69.55)
10	33.445 (33.423)	3.30 (3.30)	33.02	72.24 (72.19)

Table 4. Calculated liquid density, fitting parameter ξ, original part and dispersion correction of surface tension for planar interfaces in systems 11–12. Shown in parentheses are the densities obtained by fitting to the error function.

System	ρ_α / nm^{-3}	ξ / nm	σ_o / mJ m^{-2}	σ_d / mJ m^{-2}
11	33.674 (33.631)	0.141	60.01	5.62
12	33.251 (33.211)	0.137	60.15	6.02

5. Comment: *Technical corrections*

i) The caption of Table 4 is erroneous.

ii) Page 23176, equation 9: the exponent of R_e *is 2, not 3.*

iii) Page 23178, line 24: radius number densities of water -> radial number densities of water.

iv) Page 23182, the beginning of line 9: are -> where.

v) In several places in the text: Alejandre, not Alejandrea. Irving, not Ivring (also in the captions of figs. 2 and 4). Zakharov, not Zakharova (also in the references list).

Our response: We thank the referee for pointing out these technical corrections. We have corrected all these typos in the manuscript.