Report on acp-2010-732 Glycine in Aerosol Water Droplets: A Critical Assessment of Köhler Theory by Predicting Surface Tension from Molecular Dynamics Simulations by X. Li *et al*

The submitted contains a molecular dynamics investigation of the curvature dependence of surface tension of aerosols with glycine and using Köhler's theory. In Köhler's theory the nucleation is governed by two (driving) terms, a Kelvin (surface tension) term and a bulk Raoult therm.

The main reason why I can not recommend the work for publication is that the authors own data demonstrate that one can not use Köhler's theory for an aerosol with Glycine. The reason is simply that the mixture do not follow Raoult's (ideal mixture) law but instead of Henry's. This can be seen by compare the density profiles in Figure 3 (Glycine) with the profiles of (pure) water in Figure 1 (The author have omitted the profile of water with Glycine!). I do not know whether this crucial shortcoming can be healed.

I have some other critical comments: From where comes Eq. 4? It is not a trivial problem to handle the effect of long range forces in gas-liquid interfaces. See e.g. a resent published article J. Chem. Phys. 130, 054703 (2009), but there are many other, which show that although the forces are weak- the net effect for a semi infinite continuum is (very) important. Well you might argue that you indeed have taken this effect into account by Eq.4, but it looks fundamentally wrong: The (effective) energy parameter ϵ_{ij} between different specie is not an arithmetic mean, but a geometric ("Lorentz-Berthelot",due to that it is a mean of Boltzmann factors)?

The interpretation of the stress profiles (Figure 2 and Figure 4) and the conclusion drawn from them is not "well founded". Only the net effect of the difference of the stress is well defined and give the thermodynamic effect. The problem is discuss in Proc. Roy. Soc. A 379, 231 (1982).

In summary I can not recommend the article for publication.