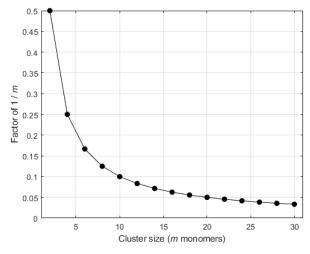
I'd like to thank the authors for the replies to mine and other reviewers' comments, and the clarifications made in the manuscript.

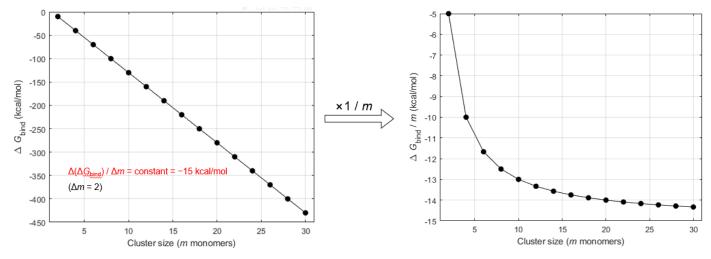
I would like to ask further clarifications related to the reply to my previous comment 2: The authors reply that they did mean that the discussed quantity in relation to Fig. 4b is the actual gradient $\Delta(\Delta G)$, which I suggested to use.

I apologize for being unclear; what I actually meant was to study the gradient $\Delta(\Delta G)$ without dividing the ΔG data by *m*. This is because the 1/m factor will always lead to a similar curve that first changes steeply and then levels off—approximately at *m* values of ca. 15-20 molecules—*regardless of the exact chemical properties*.

That is, when you scale the numbers in Fig. 4a by 1/m, the shape of the curve is determined by this factor, so you are simply bending the graphs in Fig. 4a:



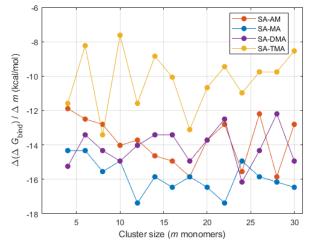
One can think of e.g. an example case of ΔG data where each energy contribution $\Delta(\Delta G)$ upon addition of monomer pair is approximately same, i.e. the gradient is $\Delta(\Delta G)$ constant, and there isn't any size-dependent transition. However, scaling this data by 1/m naturally results in the shape in the plot above:



It clearly doesn't sound reasonable to interpret this as a physical or chemical transition from clusters to particles. (Note that just to illustrate the risk in such interpretation, this example is qualitatively similar to Fig. 4.)

I'm unsure of the physical interpretation of the $\Delta G/m$ quantity. It is said to be "average binding free energy contribution in the clusters". Generally, the composition- and size-dependent contribution of monomers (or monomer pairs) to the binding free energy is described by the change upon addition of molecules, i.e. $\Delta(\Delta G)$; in contrast, the average $\Delta G/m$ doesn't contain any information of these

contributions. For the data in Fig. 4a, the actual size-dependent contributions $\Delta(\Delta G)/\Delta m$ look approximately like this:



This is quite different from the curves in Fig. 4b.

Therefore, I would ask the authors to at least add the following:

- 1. Clarify why diving the binding free energy by the total number of molecules is expected to give information on the cluster-to-particle transition, and what the physical meaning of this quantity is. Preferably give references, if possible, since I think that this might not be clear to all readers.
- 2. Please show and/or discuss how $\Delta G/m$ relates to the actual size-dependent monomer (pair) contribution $\Delta(\Delta G)/\Delta m$, and why $\Delta G/m$ should be used instead of e.g. $\Delta(\Delta G)/\Delta m$. I still recommend to show also $\Delta(\Delta G)$ (it's okay if it doesn't look as perfect as the smooth $\Delta G/m$ curves).
- 3. Explain that the general shape and leveling off of the $\Delta G/m$ curves is primarily not related to the chemical properties or composition, but simply follows from the 1/m factor which is same for all chemistries.