Interactive comment on Atmos. Chem. Phys. Discuss., 11, 6957, 2011.
Reply to anonymous referee # 2
2011-05-02

1. In the abstract, the term 'Humic-like substances of larger systems' is unclear and should be rephrased.

To clarify we exchange it with "[…] Humic-like substances in systems containing 10000 water molecules [...]"

2. Details on computational cost of the simulations would be of interest (typical total CPU time per one trajectory).

This information is surely of interest. The time for each simulation, done in steps of 1 ns, varies quite a lot, so the answer will be in form of an interval. The following sentence is inserted at the end of section 3.

The typical simulation time required for 1 ns trajectory is 50±10 hours with two AMD quad-core CPUs, run on the Ekman supercomputer at the center for high performance computing (PDC), Royal Institute of Technology (KTH) in Sweden.

3. MD simulations were performed for systems containing 3 different HULIS species, however, results of only two of them (CPA and PAL) are discussed in section 4.1 (Aggregate formation). Results for PAD should also be provided and discussed, unless they are very similar to one of those shown (in which case it should be stated).

We found three types of behavior that concerns all 3 different HULIS species. As suggested we add the following paragraph to make this clear to the reader.

The two types of aggregate formation for CPA and PAL discussed are also applicable to PAD, which showed similar behavior. Either there is a) no aggregate formation, b) aggregate formation in the interior of the cluster or c) aggregate formation near the surface. This is valid for all three HULIS compounds.

4. In the radial number density (RND) plots, RND of water should be included to show the position of the water/vapor interface of the cluster and to provide a frame of reference regarding the location of the various features occurring on the radial density profile of the HULIS species.

The following two RND plots have replaced Fig. 3a and 4a respectively. Since the RND for HULIS is less than for water, a green line show the values for HULIS multiplied with a factor of 10 to make it easier to compare HULIS and water RND. This is pointed out in the text to each figure.

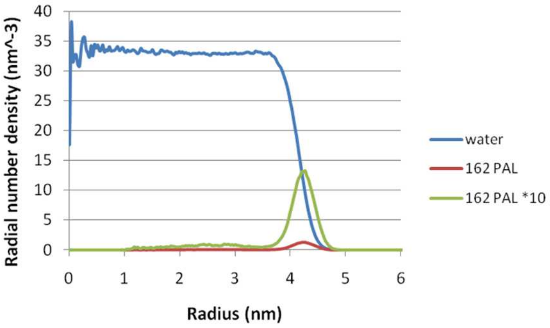

Fig. 3a. 162 PAL system with aggregate near inner surface showing a large peak for radial number density at 4.2 nm. The green line is the RND for the 162 PAL system multiplied with a factor of 10.

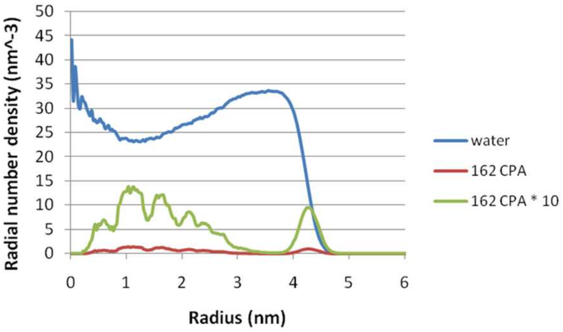

Fig. 4a. Radial number density showing two main peaks at 1.1 and 4.2 nm. The green line is the RND for the 162 CPA system multiplied with a factor of 10.

5. Regarding the surface tension calculations (section 4.2), in the paragraph below Table 2, please provide the experimental value and also the value obtained for the extended liquid/vapor interface using the SPC/E model explicitly. Please discuss possible effects of a deviation of the surface tension of a water cluster of 10 000 water molecules from that of an extended liquid/vapor interface.

A referens to experimental data is made. Regarding the planar interface for surface tension, we had unfortunately missed to include this information. The section is rewritten and reads as follows:

Because each water molecule near the surface interacts with fewer neighboring water molecules, surface tension is created. The effect is less pronounced in a sphere than in a planar interface. The smaller the sphere, the less prominent the effect would be. For pure water, the surface tension for liquid/gas interface calculated using the SPC/E water model is 57.0 mN/m for a sphere with radius of 4.3 nm and 65.0 mN/m for a planar interface (Bahadur and Russel, 2008). Experimental data give a higher value for the planar liquid/gas interface; 71.97 mN/m for at 298 K. It is common for water models to give surface tensions different from the experimental values, as few force field model could reproduce the experimental surface tensions accurately (Chen and Smith, 2007). The general behavior of the system, however, is not affected. Among the water models available, the SPC/E model is able to give the surface tensions which are closest to the experimental values. The deviation from a spherical system of 10000 water molecules from that of a planar interface is not crucial, since results for surface tension measurements in spherical clusters show a good resemblance of the trend with experimental data even for a system of half the size, 5000 water molecules (Li et. al, 2010).