Supplementary Information for:

Multiscale molecular dynamics simulations of micelles: Coarse-grain for self assembly and atomic resolution for finer details

Pilar Brocos,[1] *Paola Mendoza-Espinosa,*[2] *Rolando Castillo,*[3] *Jaime Mas-Oliva,*[2] *Ángel Piñeiro*[1,*]

[1] Departamento de Física Aplicada, Facultad de Física, Universidad de Santiago de Compostela, Campus Vida, E-15782 Santiago de Compostela, Spain

[2] Instituto de Fisiología Celular (P.O. Box 70-243) and [3] Instituto de Física (P.O. Box 20-364), Universidad Nacional Autónoma de México, 04510 México D.F., Mexico

[*]To whom correspondence should be addressed. E-mail: Angel.Pineiro@usc.es

Contents: Figures S1-S9, Tables S1-S2

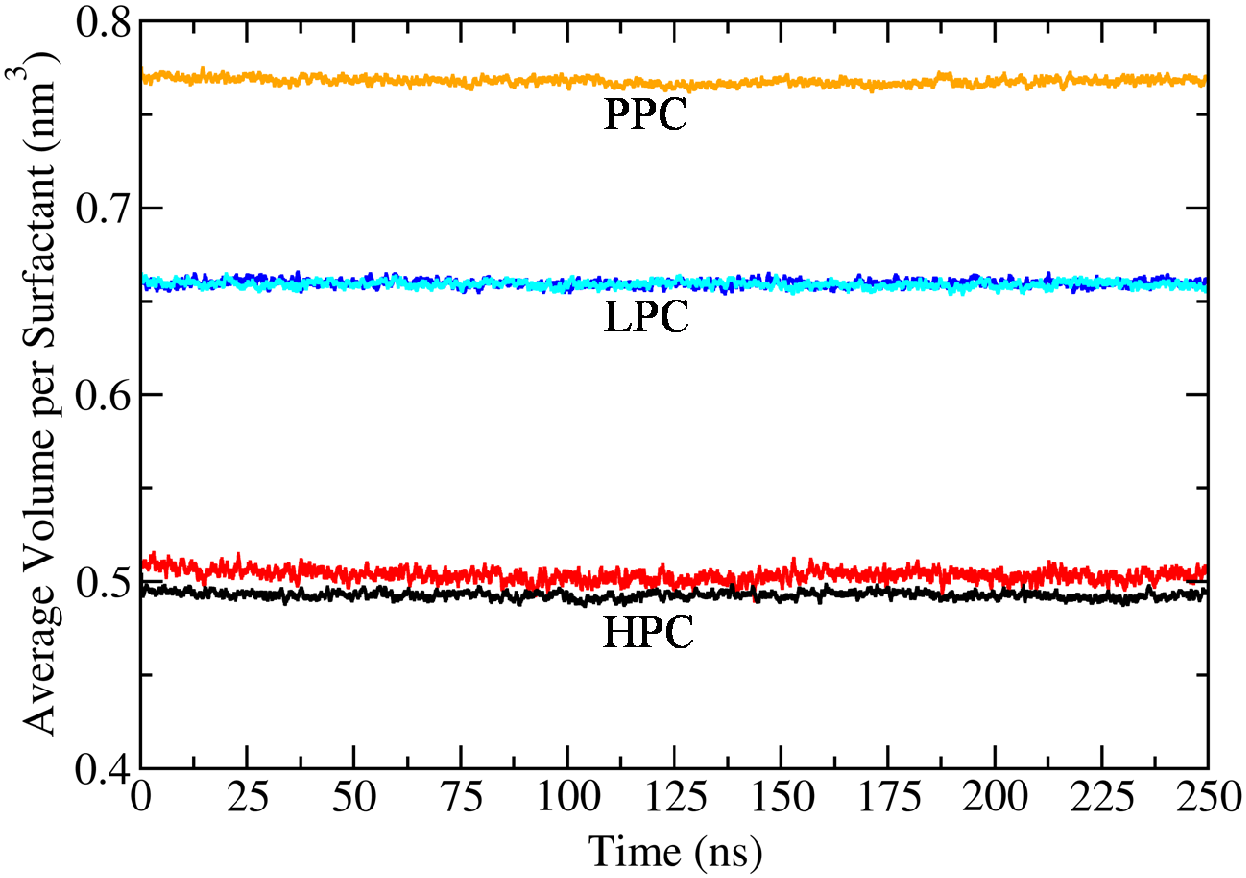


Figure S1.- Average volume per surfactant molecule (V_s) as defined in eq. 1 of the main text, obtained from the MD simulation of the systems consisting of 33 HPC (red), 67 HPC (black), 67 LPC (blue), 84 LPC (cyan), and 84 PPC (orange) molecules as a function of time.

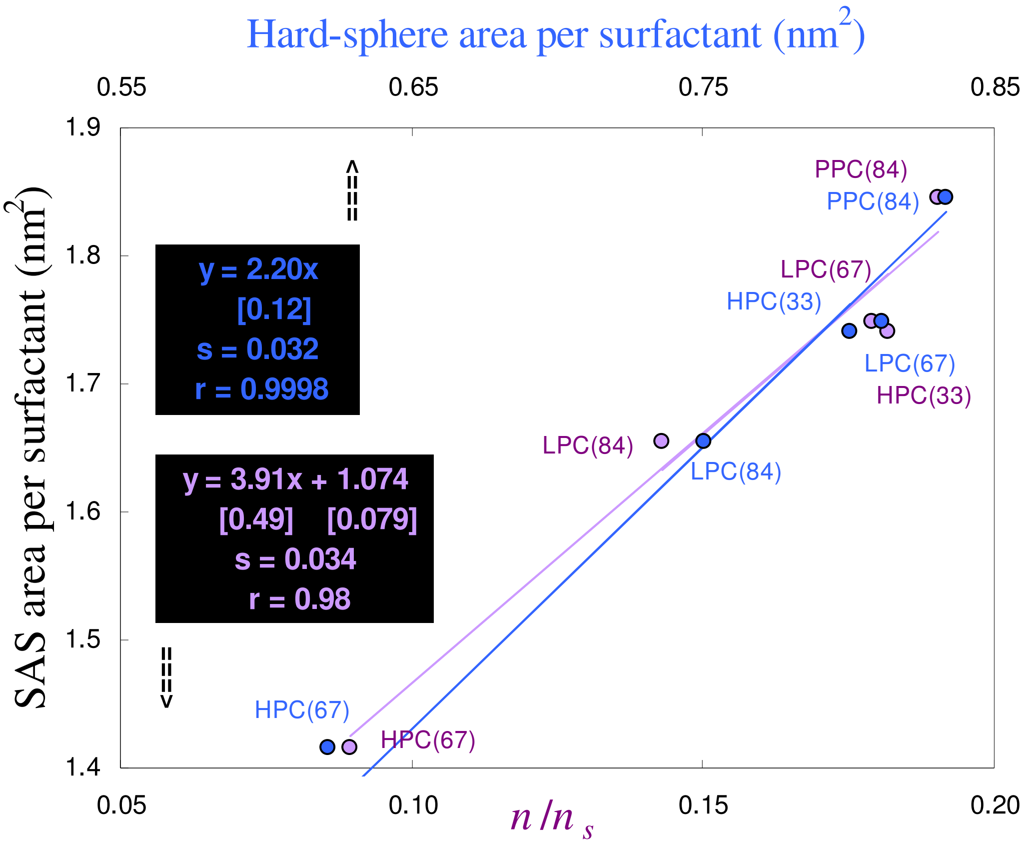


Figure S2.- Empirical linear correlations between the SAS area per surfactant and selected quantities for lyso-C$_n$PC micelles: (in purple) the ratio between the number of carbon atoms in the tail group and the number of micellized molecules, n/n_s, and (in blue) the hard-sphere area per surfactant, taking the average N-c.o.m. distance as the geometric radius. The standard deviation of the latter fit increases significantly (from 0.032 to 0.048) when the average P-c.o.m. distance is considered instead, affording a slope of 2.42 ± 0.13 (data not shown).

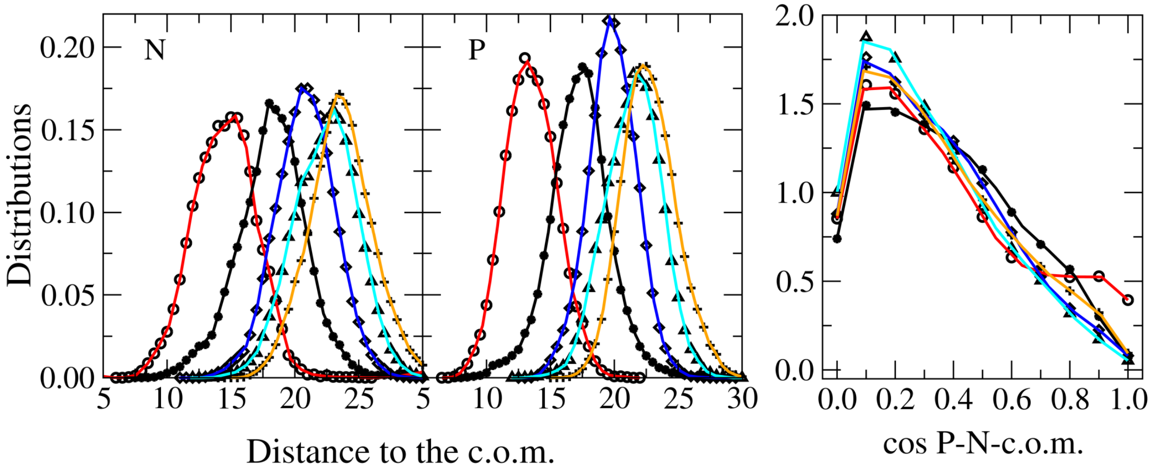
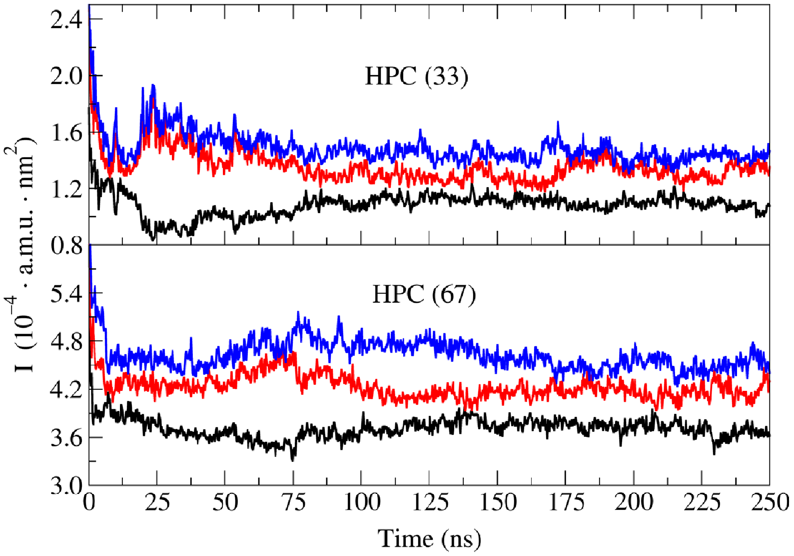
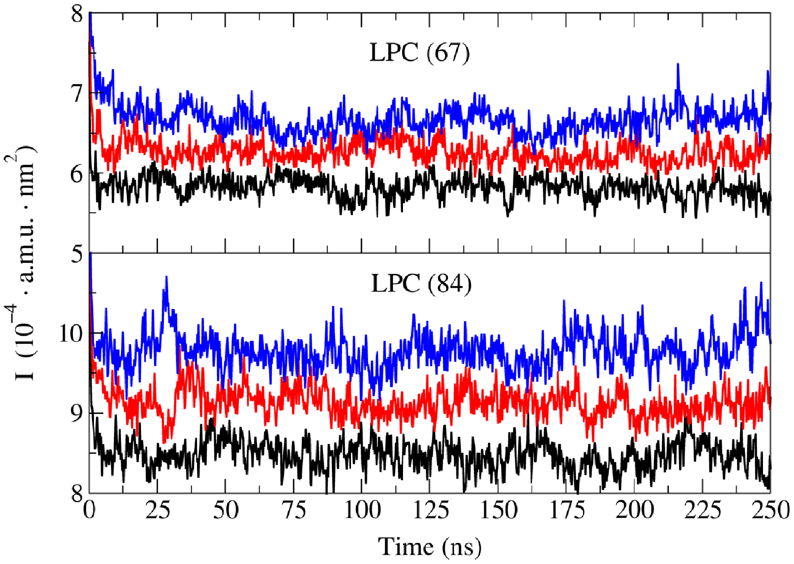


Figure S3.- Left and center: Distribution of the N-c.o.m. and P-c.o.m. distances (see Figure 6). Right: Distribution of the cosines of the P-N-c.o.m. angle (see Figure S5). Symbols for the different systems (dot type, line color): 33-HPC (○, red), 67-HPC (●, black), 67-LPC (◊, blue), 84-LPC (Δ, cyan), and 84-PPC (+, orange) micelles. Lines are intended to guide the eye.





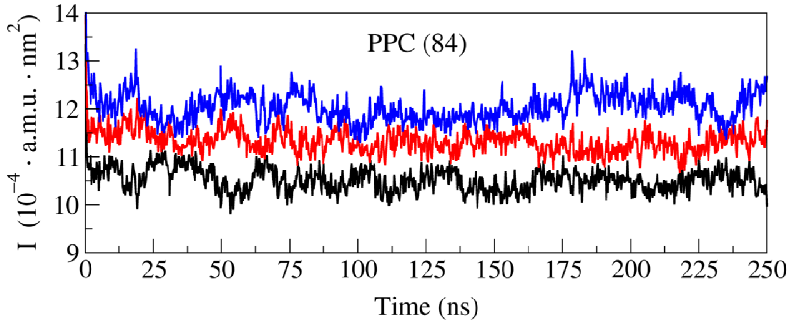


Figure S4.- Principal moments of inertia of the micellar aggregates as a function of time. The type and number of molecules constituting each micelle are indicated in each plot.

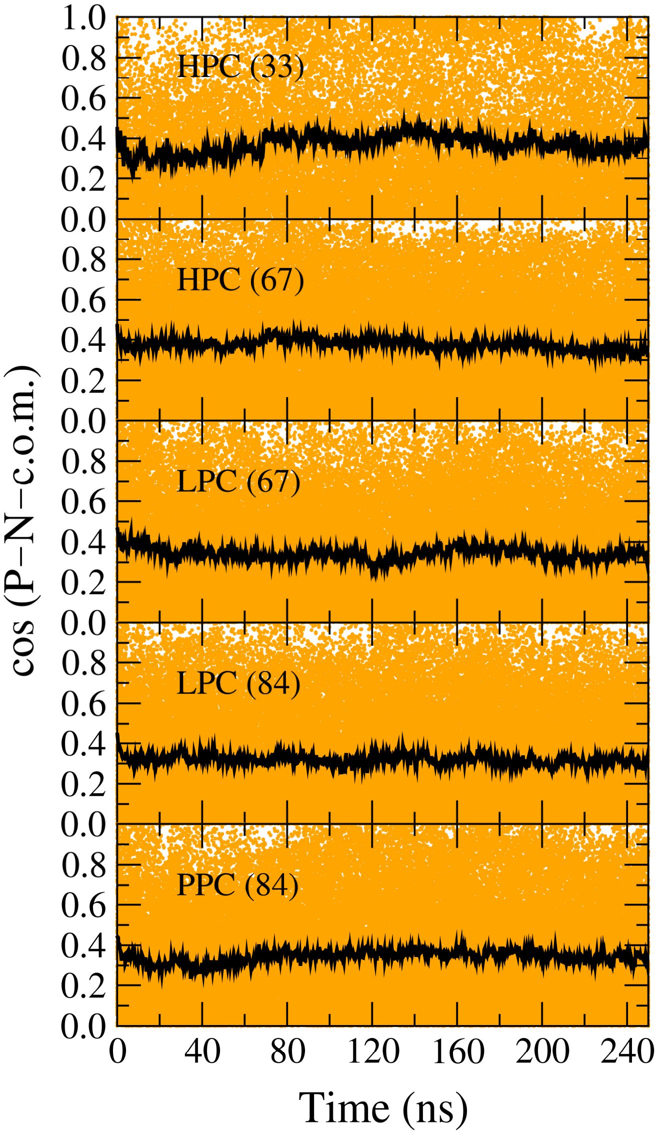


Figure S5.- Cosine of the angle formed by the atoms P and N of each surfactant molecule and the center of mass of the micelle as a function of time (orange spots). The value averaged over all the micelle molecules as a function of time is also plotted (black curve). The type and number of lyso-C$_n$PC is indicated in each panel.

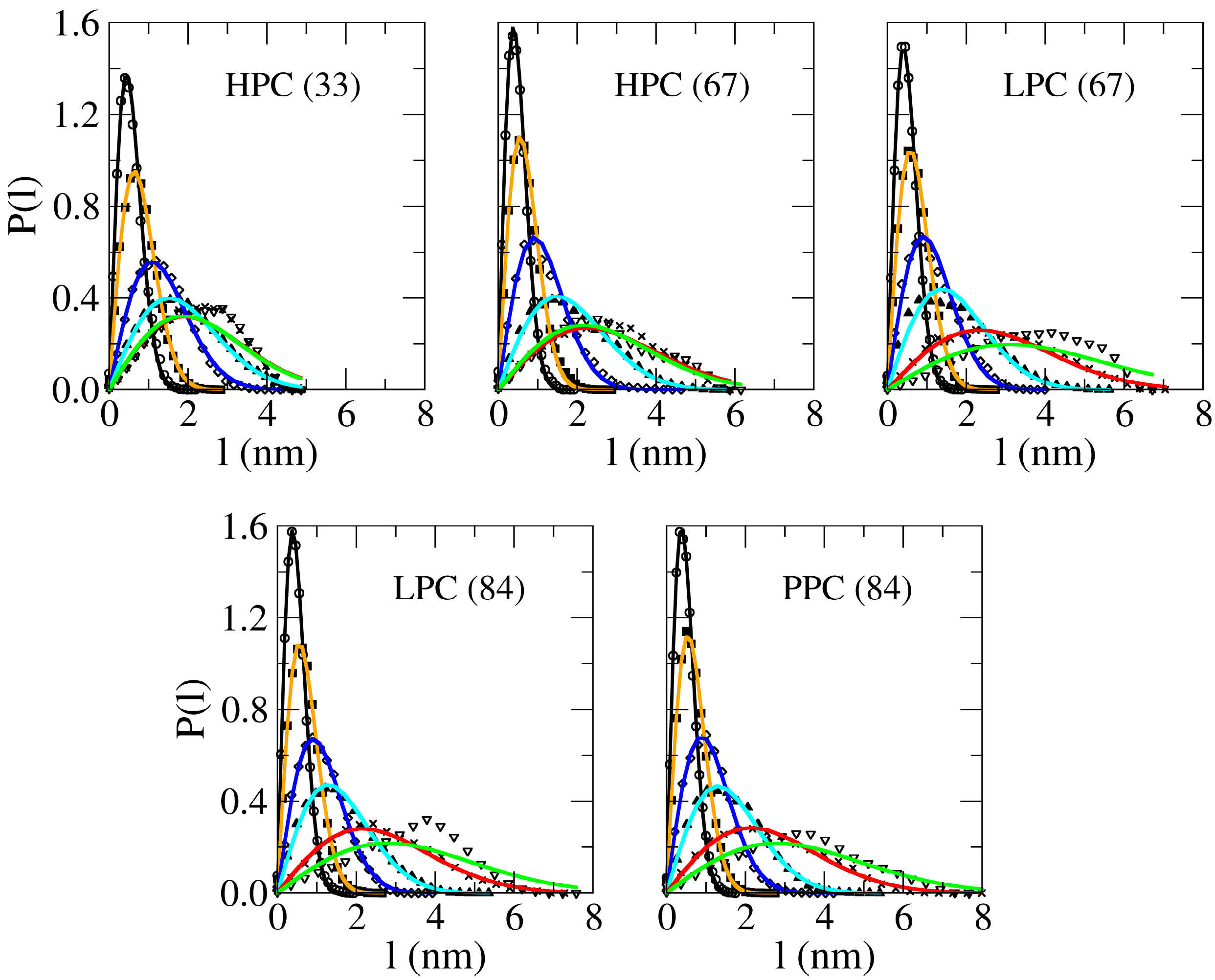


Figure S6.- Distribution of the lateral displacements (*l*) of the P atoms over the surface of the studied aggregates for 6 different Δ*t* values. Data were fitted to the expected two-dimensional random-walk distribution as detailed by Niemelä et al.[77] (see eq. 3 in the SI of the referenced work) to obtain the lateral diffusion coefficients displayed in Figure S7. Symbols (dot type, curve color): 1 ns (○, black), 2 ns (■, orange), 5 ns (◊, blue), 10 ns (▲, cyan), 25 ns (x, red), and 50 ns (∇, green).

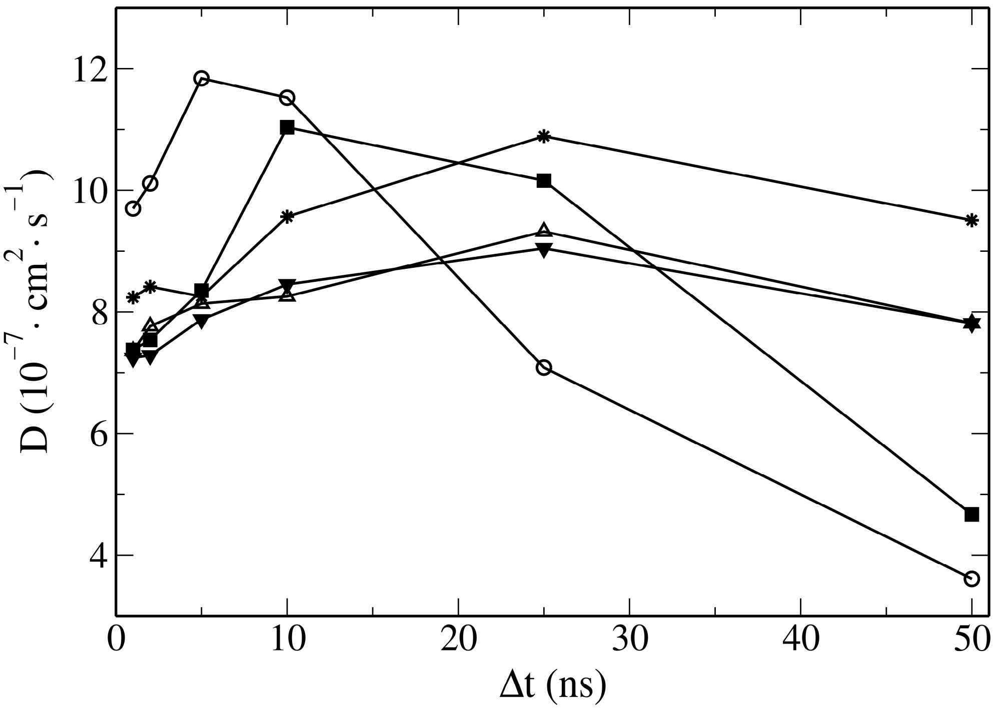


Figure S7.- Lateral diffusion coefficients obtained from the MD simulation of the systems consisting of 33 HPC (○), 67 HPC (■), 67 LPC (*), 84 LPC (Δ), and 84 PPC (▼) molecules as a function of Δ*t*. Each point is the result of fitting the corresponding distribution in Figure S6 to the expected two-dimensional random-walk distribution as detailed by Niemelä et al.[77] (see eq. 3 in the SI of the referenced work). Lines are intended to guide the eye.

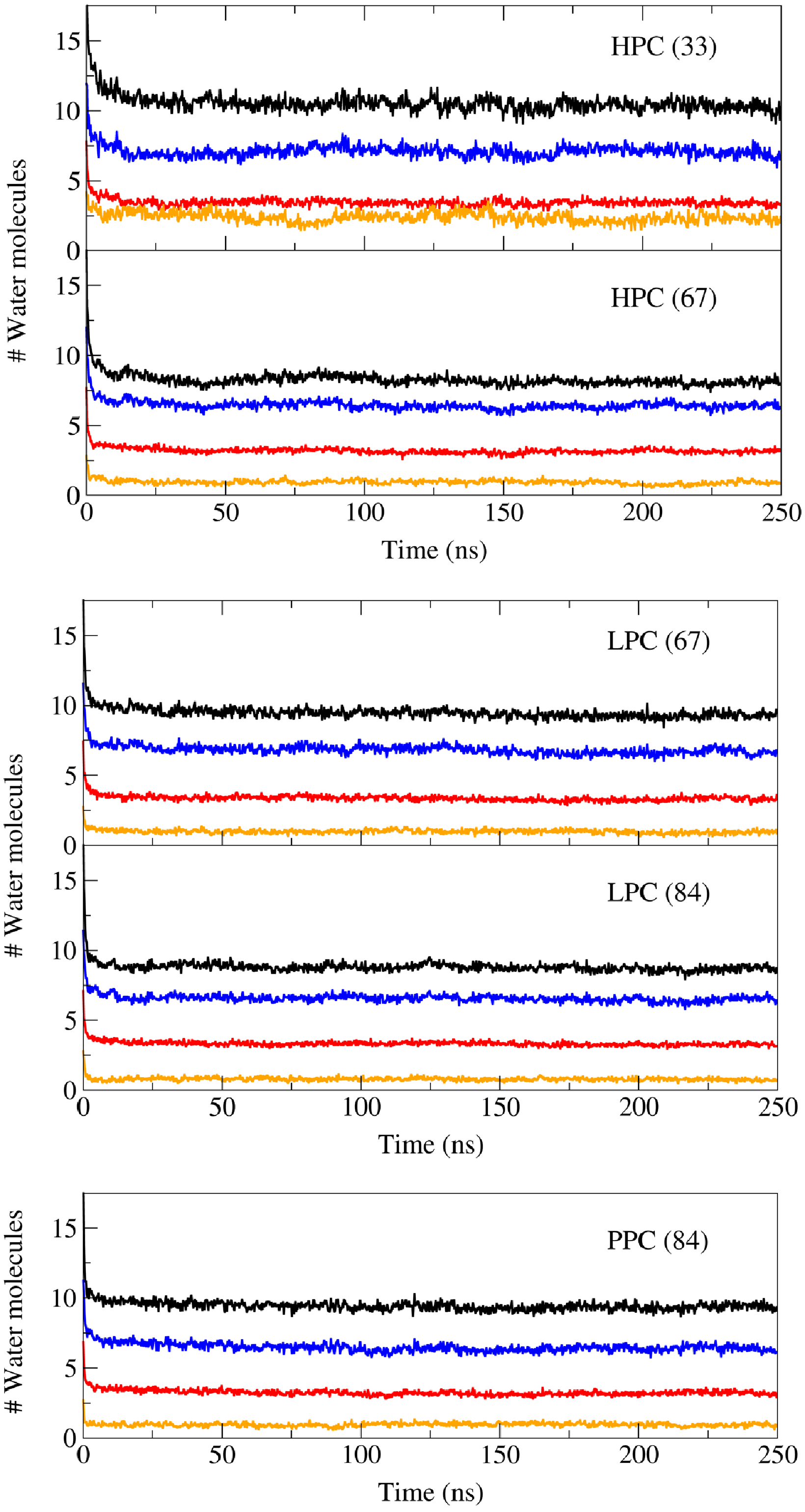


Figure S8.- Number of water molecules at less than 4 Å from any atom of groups G (black), N (blue), P (red), and T (orange), as defined in Figure 1. The type and number of molecules constituting each micelle are indicated in each plot.

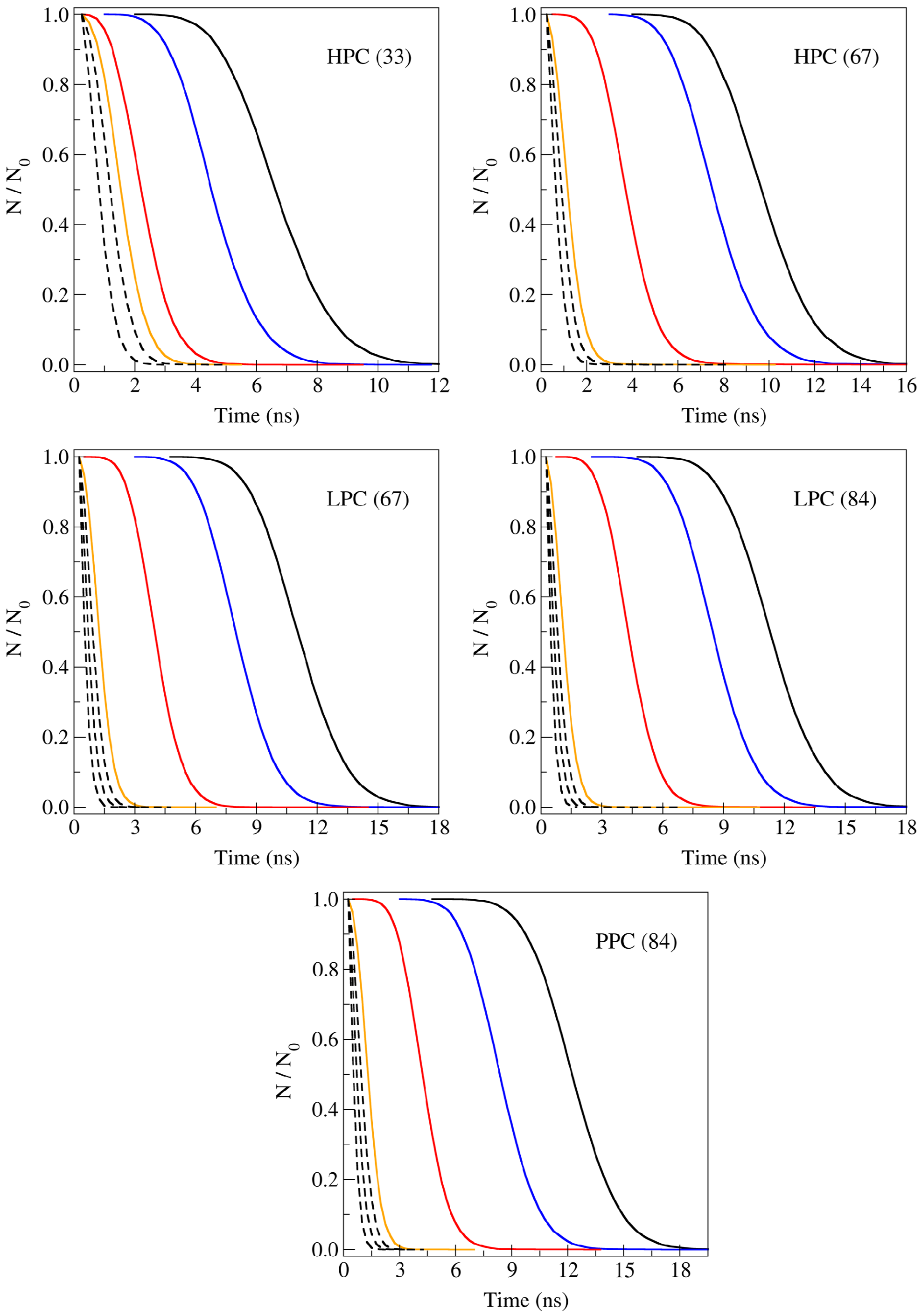


Figure S9.- Decay functions for the normalized water-surfactant contact number, N/N_0, intended to illustrate the time the interactions stand. Contacts are defined by using a cutoff of 4 Å. The number of contacts for a given group at the starting point (50 ns of MD simulation), N_0, is used as normalizing constant and taken as reference to follow their lifetime through the variable N. The calculations were performed for the last 200 ns of the atomistic trajectories and for the atomic groups G (solid black), N (blue), P (red), and T (orange), as defined in Figure 1. Dashed black curves stand for the sequential deletion in T group of the C-atom pairs that are closest to the surfactant polar head and show the corresponding decrease in the mean lifetime of the interaction.

Table S1.- Splitting of the SAS area per surfactant in its hydrophilic and hydrophobic contributions, with indication of the percentage corresponding to the latter.

	Total SAS area per surfactant (nm^2)	Hydrophilic SAS area (nm^2)	Hydrophobic SAS area (nm^2)	Hydrophobic percentage (%)
HPC (33)	1.741 ± 0.050	1.160 ± 0.041	0.581 ± 0.037	33.4
HPC (67)	1.416 ± 0.042	1.036 ± 0.028	0.381 ± 0.026	26.9
LPC (67)	1.749 ± 0.042	1.128 ± 0.030	0.621 ± 0.032	35.5
LPC (84)	1.655 ± 0.036	1.100 ± 0.028	0.555 ± 0.027	33.5
PPC (84)	1.846 ± 0.036	1.085 ± 0.025	0.761 ± 0.030	41.2

Table S2.- Comparison of the hard-sphere, geometric radius (given by the average distance from the N or P atoms to the center of mass) with the hydrodynamic radius.[a, b]

	Average distance (nm)		Hydrodynamic radius (nm)
	N-c.o.m.	P-c.o.m.	
HPC (10 ± 2)			1.4 [c]
HPC (33)	1.45 ± 0.24	1.35 ± 0.20	
HPC (67)	1.82 ± 0.27	1.73 ± 0.23	
LPC (unspecified n_s)			2.52 ± 0.12 [d]
LPC (55 ± 7)			2.6 [c]
LPC (67)	2.08 ± 0.23	1.99 ± 0.19	
LPC (84)	2.24 ± 0.25	2.15 ± 0.21	
PPC (unspecified n_s)			2.8 [e]
PPC (84)	2.36 ± 0.25	2.27 ± 0.21	

[a] For a given lyso-C_nPC, data are ordered by increasing n_s. [b] All the hydrodynamic radii were reported at 298 K. [c] By PGSTE-NMR spectroscopy in D_2O, above the cmc.[72] [d] By dynamic light scattering, at 20 mM concentration (28.6 times the cmc).[57] [e] By dynamic light scattering, at 0.2 mM concentration (28.6 times the cmc).[76]