

Supporting information: numerical estimation of the number of aggregates that might have sedimented during the experiment

According to the Mie theory, the number of scattering entities is proportional to the ratio of their scattering intensity to the product of their scattering efficiency (q_{scat}) and radius of the particles to the sixth power. In all calculations, 19 nm was used as the diameter of the primary particles and 630 nm was used as the average diameter of the aggregate population exceeding 400 nm in diameter.

The scattering intensity of each particle population was calculated by integrating the signal of the primary particles and of the > 400 nm aggregates. The ratio of the intensities of aggregates versus primary particles was 18.5/1.

The scattering efficiency of the particles is dependent on the particle radius and the detector angle. These values of q_{scat} were calculated using the interactive Mie Scattering Calculator developed by Scott Prahl at http://omlc.ogi.edu/calc/mie_calc.html, taking the particle diameter, resp. 19 and 630 nm and the particle concentration of 0.02310 particles/ μm^3 into account. Other important experimental parameters were the wavelength of the DLS laser beam, being 659 nm and the refractive indices of the medium and the particles, 1.33 (water) and 1.54 (silica) respectively. It was assumed that the particles did not absorb laser light. For the small particles, the angular dependence of the scattering efficiency was low, varying from 0.98 for 180° to 1 for 0° scattering angle. The scattering efficiency of the primary particles at 90° scattering angle was 0.994. For the aggregates with an average diameter of 630 nm, the angular dependence of the scattering efficiency was much more pronounced, varying from 0.0005 for 122° scattering angle to 1 for 0° scattering angle. The scattering efficiency of the aggregates in the experimental setup with 90° scattering angle was 0.010, being almost 100 times less than the scattering efficiency of the primary particles.

The ratio of the aggregates to primary particles can now be estimated using the following formula:

$$\frac{N_{\text{agg}}}{N_{\text{p}}} = \frac{I_{\text{agg}} * R_{\text{p}}^6 * q_{\text{scat_p}}}{I_{\text{p}} * R_{\text{agg}}^6 * q_{\text{scat_agg}}} = \frac{1}{7.18 \cdot 10^5}$$

With N_{agg} : the number of aggregates

N_{p} : the number of primary particles

I_{agg} : the intensity generated by the aggregates

I_{p} : the intensity generated by the primary particles

R_{agg} : the radius of the aggregates

R_{p} : the radius of the primary particles

$q_{\text{scat_agg}}$: the scattering efficiency of the aggregates at 90° scattering angle

$q_{\text{scat_p}}$: the scattering efficiency of the primary particles at 90° scattering angle

Taking the particle concentration of 70 $\mu\text{g/ml}$ into account, we calculated the number of primary particles to be $2.11 \cdot 10^{13}$, based on the TEM diameter and the density of the particles being 2.0 g/cm^3 . The number of aggregates would then be $2.94 \cdot 10^7$. The projected surface of the aggregates $N_{\text{agg}} * \pi * r^2$ equals $8.31 \cdot 10^{-6} \text{ m}^2$. Supposing that all aggregates would sediment within the 24h of the experiment, the aggregates would cover 0.0831 cm^2 or approximately 8% of the 1 cm^2 measurement tube.