Flocculation of natural organic matter in Swedish lakes

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Flocculation is an important part of the carbon cycle. It is therefore crucial to understand how flocculation is regulated and how different environmental factors impact. A dilemma is that it has been found difficult to measure flocculation experimentally.

In this thesis, flocculation of dissolved organic carbon in a Swedish lake was measured in a series of laboratory experiments. The method used was Dynamic Light Scattering (DLS). DLS is used to determine the size distribution profile of, for instance, small particles in suspension. DLS measures Brownian motion and relates it to the particle size by measuring the fluctuation in scattering intensity. It is not very effective to measure the frequency spectrum contained in the intensity fluctuations directly, so instead, a digital auto correlator is used.

Since factors such as pH, salinity and calcium chloride content varies in lakes and is thought to have an impact on flocculation, this was investigated as well. As pH was changed in a range of 3 to 9, small changes in size distribution could be detected. Salinity and calcium chloride content have quite an impact on flocculation. Time also has a great impact, samples that were set to rest for a week showed a significant increase in particle size.

For DLS to work, the samples need to be filtered of centrifuged to get rid of large particles. Different types of filters were tested to see which filter material was the best to use. When filtering the water we only want to filter out the large particles. Natural organic matter has a hydrophobic component which adsorbs to some filter types but not to others. It is crucial to know which filters this hydrophobic component adsorbs to, so that the loss of dissolved organic carbon during filtration can be minimalized.
Summary

Flocculation is an important part of the carbon cycle. It is therefore crucial to understand how flocculation is regulated and how different environmental factors impact. A dilemma is that it has been found difficult to measure flocculation experimentally.

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Introduction

Flocculation

IUPAC defines flocculation as “a process of contact and adhesion whereby the particles of a dispersion form larger-size clusters.”[1]

A lot of organic carbon comes into Swedish lakes. The three essential things that happen to that organic carbon are mineralization, sedimentation and export via rivers. There are many Swedish lakes and there is a significant amount of carbon in the water as well as in the sediment in these lakes. The carbon that is brought into a lake by incoming water represents a size continuum of molecules, colloids and particles and is divided into dissolved organic carbon (DOC) and particulate organic carbon (POC). These two fractions can be separated by filtration.[2] DOC is defined as the organic carbon that is small enough to pass through a filter, while POC is defined as the particles of organic carbon that are too large to pass through a filter and are filtered out.[3]

Little is known about flocculation of DOC in Swedish lakes. Flocculation is not even included as a flux of carbon in most of the conceptual models of the carbon cycle. It is not so well known to what extent sediment processes contribute to the mineralization or the preservation of organic carbon entering the lakes. Where the mineralization occurs, in the water or in the sediments, is influenced by the rate and extent of the flocculation. Hence, flocculation and subsequent sedimentation together with mineralization of the flocculated material might be very important in the metabolism of carbon in lakes.[2]

Given the potential importance of flocculation (as highlighted in the paragraph above), it is crucial to understand how flocculation is regulated and how different environmental factors impact. It is important to have as much knowledge as possible of flocculation processes to be able to understand the carbon cycle in Swedish lakes. A dilemma has been that it has been difficult to measure the flocculation rates experimentally.

pH in Swedish lakes is highly variable. So is salt, calcium and magnesium content. This thesis aims to find out how these factors affect the flocculation of natural organic matter. It also aims to test the Dynamic Light Scattering (DLS) method in order to be able to study flocculation in shorter time and with greater accuracy.

Dynamic Light Scattering (DLS)

Dynamic Light Scattering is also known as Photon Correlation Spectroscopy. It is used to determine the size distribution profile of small particles in suspension or polymers in a solution.

DLS measures and relates Brownian motion to the size of particles by measuring the fluctuation in the scattering intensity. The rate of the intensity fluctuations depends on the particle size. Brownian motion is when a particle moves randomly when struck by the solvent molecules surrounding it. The Brownian motion will be slower when the particles are larger and faster when the particles are smaller. That is because smaller particles move further and faster when hit by solvent molecules.

The frequency spectrum contained in the intensity fluctuations can be measured directly, but it is not very effective to do so. It is better to use a digital auto correlator. A correlator is
in fact a signal comparator. It measures the similarity between two signals or compares one
signal with itself at different points in time. If the intensity of a randomly fluctuating signal is
compared with itself at two points far apart in time, the intensities are not related in any way,
there will be no correlation. But if the intensity of the signal is compared with itself at two
points close to each other in time, they will be strongly correlated. The correlation is
decreasing with time, so, the more time there is between the points in time at which the signal
is compared to itself, the weaker the correlation. The correlation will decrease slower if the
particles are large because the signal will then be changing slower. If the particles are small
and hence moving faster, the correlation will decrease faster. Perfect correlation is indicated
as 1,00 and no correlation at all is indicated as 0,00.

A correlogram provides a lot of information about the measured sample. The point in time
at which the correlation starts to decrease considerably indicates the mean size of the sample.
If the sample is very monodisperse, the line will be very steep. If the sample is very
polydisperse, the line will be very flat. Dispersity can be defined as the heterogeneity of
sizes of molecules or particles in a mixture. If the molecules or particles in a sample have the
same size, shape or mass, the sample is monodisperse. If the particles or molecules in a
sample have different size, shape or mass, the sample is polydisperse.
Aims of the Thesis

My hypothesis is that dissolved organic carbon in Swedish lakes flocculates and that factors such as pH, salinity and calcium and magnesium content affects the flocculation. I investigated which of these factors affect the flocculation and how they affect it. I also investigated and tested different kinds of filters because for the DLS to work it was necessary to filter the water. My hypothesis is that the hydrophobic component of natural organic matter adsorbs to some filter materials but not to others.
Methods

Filters and filter holders

In order for the DLS method to work, large particles needed to be filtered out. Otherwise they would disturb the light scattering. Therefore, different types of filters needed to be tested to see which type was most suitable to use, this because natural organic matter has a hydrophobic component which adsorbs to some types of filter materials and we do not want the organic carbon that is not particulate to stick to the filter. Samples of the lake water were filtered through different types of filters. The filters that were used and their specifications are presented in Table 1. Both vacuum and pressure filter holders were used. Both types of filter holders are shown in Figure 1. The vacuum filter holder was plastic and for 45mm filters. The pressure filter holder was also plastic but for 25mm filters and the water was pushed through by a syringe. First, 500 ml lake water was filtered through a 0,7μm×45mm glass fiber filter to get rid of flocs and particles. Then, samples of 100 ml of the filtered water were filtered through five different types of filters.

<table>
<thead>
<tr>
<th>Filter type</th>
<th>Diameter (mm)</th>
<th>Pore size (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass fiber</td>
<td>45</td>
<td>0,7</td>
</tr>
<tr>
<td>Cellulose acetate</td>
<td>45</td>
<td>0,45</td>
</tr>
<tr>
<td>HV</td>
<td>45</td>
<td>0,45</td>
</tr>
<tr>
<td>HA</td>
<td>25</td>
<td>0,45</td>
</tr>
<tr>
<td>GTTP</td>
<td>25</td>
<td>0,2</td>
</tr>
</tbody>
</table>

Table 1. The filters used in the analysis.

Figure 1. (a) and (b) Vacuum filter holders. (c) and (d) Pressure filter holders.
In two additional experiments, 500 ml lake water, filtered through a glass fiber filter, was acidified by adding HCl and 500 ml likewise filtered lake water was alkalized by adding NaOH. The water was not changed to any exact measured pH because it was not necessary to know the pH in this experiment. Three different samples of 100 ml filtered acidified lake water and three different samples of filtered alkalized lake water were filtered through glass fiber-, cellulose acetate- and HV filters respectively. This was done to see if pH had an impact on the adsorption of the hydrophobic component of natural organic matter to the different filter materials.

Reference samples
When using an ALV Dynamic Light Scattering system, reference samples are not necessary. That is because they will simply not yield any correlation or particle distribution. Still, measurements were done on a number of reference samples that contained Milli-Q water. First, raw Milli-Q water was measured and it was soon revealed that the Milli-Q water provided was not clean. Good correlation was seen and the particle distribution showed large particles. In an attempt to purify the Milli-Q water, it was filtrated through a glass fiber filter and then measured in the DLS system. There was still good correlation and the size distribution curve showed even more and larger particles. Instead, LC-MS water was used. This water was also measured in the DLS system to make sure it was pure, which, turned out, it was. The LC-MS water was then filtered through a glass fiber filter and the filtered water was then measured, only to reveal that there were now particles in the water. The conclusion was drawn that the particles had arisen from the filters, since the LC-MS water was clean before it was filtrated and was full of particles after it was filtrated. This lead to the conclusion that the lake water could not be purified by filtration. Another means of purifying was needed. So, instead of filtration, centrifugation was used.

ALV Dynamic Light Scattering system and measurements
To measure whether flocs were formed in the prepared samples, an ALV Dynamic Light Scattering system was used. A Dynamic Light Scattering system is shown in Figure 2. All samples had the volume of 1 ml. Measurements were made on three samples of acidic lake water, each to which hydrochloric acid was added between runs so that measurements were made at pH 3, 4, 5 and 6, and three samples of alkaline lake water to which sodium hydroxide was added between runs so that measurements were made at pH 6, 7, 8 and 9. The pH of the raw lake water was measured to pH 6. Measurements were also made on three samples of lake water with 5, 15 and 35 g/l NaCl and one sample with 100 µl CaCl₂ in 1 ml water. All the samples were left for a week and were then measured again. All the acidic samples had pH 3 and all the alkaline samples had pH 9. The combinations of measurements are presented in Tables 2-4 below.
Table 2. The pH’s the acidic and alkaline samples were measured at.

<table>
<thead>
<tr>
<th>Sample</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td>1a</td>
<td>3 4 5 6</td>
</tr>
<tr>
<td>2a</td>
<td>3 4 5 6</td>
</tr>
<tr>
<td>3a</td>
<td>3 4 5 6</td>
</tr>
<tr>
<td>1b</td>
<td>6 7 8 9</td>
</tr>
<tr>
<td>2b</td>
<td>6 7 8 9</td>
</tr>
<tr>
<td>3b</td>
<td>6 7 8 9</td>
</tr>
</tbody>
</table>

Table 3. The concentrations/amounts of NaCl/CaCl₂ in measured samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Conc. NaCl/CaCl₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>1c</td>
<td>5 g/l NaCl</td>
</tr>
<tr>
<td>2c</td>
<td>15 g/l NaCl</td>
</tr>
<tr>
<td>3c</td>
<td>35 g/l NaCl</td>
</tr>
<tr>
<td>4c</td>
<td>100 µl CaCl₂</td>
</tr>
</tbody>
</table>

Table 4. The samples measured after a week.

<table>
<thead>
<tr>
<th>Sample</th>
<th>pH/NaCl/CaCl₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>1a</td>
<td>3</td>
</tr>
<tr>
<td>2a</td>
<td>3</td>
</tr>
<tr>
<td>3a</td>
<td>3</td>
</tr>
<tr>
<td>1b</td>
<td>9</td>
</tr>
<tr>
<td>2b</td>
<td>9</td>
</tr>
<tr>
<td>3b</td>
<td>9</td>
</tr>
<tr>
<td>1c</td>
<td>5 g/l NaCl</td>
</tr>
<tr>
<td>2c</td>
<td>15 g/l NaCl</td>
</tr>
<tr>
<td>3c</td>
<td>35 g/l NaCl</td>
</tr>
<tr>
<td>4c</td>
<td>100 µl CaCl₂</td>
</tr>
</tbody>
</table>

Figure 2. Dynamic Light Scattering system. The cap in the middle is taken off and the sample, in a cylindrical cuvette, is inserted. The cuvette is surrounded by toluene to reduce or eliminate scattering on the surface of the cuvette. The laser comes through the tube on the right side. The gray tubing is for water to hold the temperature of the system.
Results and Discussion

Filtering
After filtering the lake water through the different types of filters it was clear that some filters had been colored by the water and others had not been. It was also evident that the acidification and alkalization had an impact on the results. The results are presented in Table 5 and Table 6.

Table 5. Results from filtration of neutral lake water.

<table>
<thead>
<tr>
<th>Filter type</th>
<th>Color</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass fiber</td>
<td>No</td>
</tr>
<tr>
<td>Cellulose acetate</td>
<td>No</td>
</tr>
<tr>
<td>HV</td>
<td>Yes</td>
</tr>
<tr>
<td>HA</td>
<td>No</td>
</tr>
<tr>
<td>GTTP</td>
<td>Yes</td>
</tr>
</tbody>
</table>

Table 6. Results from filtering the acidic and alkaline lake water through three different types of filters. (*The first filtration of neutral water took very long because there was no good flow of the water through the filter. The most reasonable explanation is that something went wrong when the vacuum filter holder was put together for this filtration.). The discoloration is presented in a scale from 0 to 6 where 0=No discoloration, 1=Almost no discoloration, 2=Very, very slight discoloration, 3= Very slight discoloration, 4=Slight discoloration, 5=Less discoloration and 6=Discoloration. When this experiment was done, these three were the only filter types available.

<table>
<thead>
<tr>
<th>pH</th>
<th>Filter nr.</th>
<th>Glass fiber</th>
<th>HV</th>
<th>Cellulose acetate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutral</td>
<td>1</td>
<td>4</td>
<td>4</td>
<td>6*</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>0</td>
<td>3</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>-</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td>Acidic</td>
<td>1</td>
<td>2</td>
<td>6</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>0</td>
<td>5</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>-</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Alkaline</td>
<td>1</td>
<td>1, strange stains</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>0</td>
<td>1</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>-</td>
<td>1</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 5 indicates that the hydrophobic component of natural organic matter adsorbs to the HV- and GTTP filters but not to the glass fiber-, cellulose acetate-, and HA filters. It also indicates that the filters are more colored by the water at the first filtration than at the second and third. This seems to apply to all samples and all filters. The reason that this happens is most likely that most of the hydrophobic components adsorb to the first filter and there is little left to adsorb to the second and third filter. Unfortunately, further specifications for the filters were very hard to find, making it hard to draw any conclusions as to why the hydrophobic component of natural organic matter adsorbs to some filter types but not to others.
From Table 6 it is clear that pH has an impact on both flocculation and adsorption of the hydrophobic component of natural organic matter to the filters. However, it is hard to give separate pictures of the impact on the respective processes. The results from the glass fiber filters show no big difference between natural, acidic and alkaline water. This is most likely because the hydrophobic component of natural organic matter does not adsorb to the glass fiber filters. The results from the HV filters, on the other hand, show a clear difference between natural, acidic and alkaline water. When the acidic water was filtrated there was more discoloration on the HV filter than when natural water was filtrated. When alkaline water was filtrated there was less discoloration on the HV filter than when natural water was filtrated. From this, the conclusion can be drawn that it is evident that pH has an impact on the adsorption. The hydrophobic component of natural organic matter adsorbs better to the HV filter when the water is acidic and is less prone to adsorb when the water is alkaline. The results from the cellulose acetate filters are almost the same as for the glass fiber filters, setting aside the error in the first filtration of natural water through the cellulose acetate filters. But for cellulose acetate there is a small difference between neutral, acidic and alkaline water. It can be seen that there is a little more discoloration when the acidic water is filtrated and a little less discoloration when the alkaline water is filtrated.

**Dynamic Light Scattering**

After running all the samples through the ALV Dynamic Light Scattering system it was evident that flocs were indeed formed in the water. All the measurements gave good correlation and the size distribution curves showed that there were indeed particles formed in the water. It was also clear that pH, salinity and CaCl$_2$ concentration had impact on the flocculation. The results from the measurements are presented in Figures 3 to 22. The raw data can be found in Figures A1 to A38 in the Appendix.

![Figure 3](image.png)

*Figure 3.* Curves for Vial 1 acidic samples pH 3-6. Green = pH 6, Red = pH 5, Blue = pH 4 and Yellow = pH 3.
Figure 4. Curves for Vial 2 acidic samples pH 3-6. Green = pH 6, Red = pH 5, Blue = pH 4 and Yellow = pH 3.

Figure 5. Curves for Vial 3 acidic samples pH 3-6. Green = pH 6, Red = pH 5, Blue = pH 4 and Yellow = pH 3.

Figure 6. Curves for Vial 1 alkaline samples pH 6-9. Green = pH 6, Red = pH 7, Blue = pH 8 and Yellow = pH 9.
Figure 7. Curves for Vial 2 alkaline samples pH 6-9. Green = pH 6, Red = pH 7, Blue = pH 8 and Yellow = pH 9.

Figure 8. Curves for Vial 3 alkaline samples pH 6-9. Green = pH 6, Red = pH 7, Blue = pH 8 and Yellow = pH 9.

Figure 9. Curves for pH 6 acidic samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.
Figure 10. Curves for pH 5 acidic samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.

Figure 11. Curves for pH 4 acidic samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.

Figure 12. Curves for pH 3 acidic samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.
Figure 13. Curves for pH 6 alkaline samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.

Figure 14. Curves for pH 7 alkaline samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.

Figure 15. Curves for pH 8 alkaline samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.
Figure 16. Curves for pH 9 alkaline samples Vial 1-3. Green = Vial 1, Red = Vial 2 and Blue = Vial 3.

Figure 17. Curves for pH 3 acidic samples Vial 1 Fresh and a week old. Green = Fresh and Red = 1 week.

Figure 18. Curves for pH 3 acidic samples Vial 2 Fresh and a week old. Green = Fresh and Red = 1 week.
Figure 19. Curves for pH 3 acidic samples Vial 3 Fresh and a week old. Green = Fresh and Red = 1 week.

Figure 20. Curves for pH 9 alkaline samples Vial 1 Fresh and a week old. Green = Fresh and Red = 1 week.

Figure 21. Curves for pH 9 alkaline samples Vial 2 Fresh and a week old. Green = Fresh and Red = 1 week.
When studying the correlation curves in the Appendix, it can be seen that they are generally rather steep. This means that the samples are quite monodisperse. It is also evident that the correlation is very good. One exception is Figure A38, but looking at Figure A34, which is the result from measuring the same sample as in Figure A38 but a week earlier, the conclusion can be drawn that some contamination must have happened between the measurements. Figure A38 can also be compared to Figures A36 and A37, leading to the same conclusion. Another exception is Figure A35. Comparing Figure A35 to Figure A31, which is the result from measuring the same sample as in Figure A35 but a week earlier, leads to the conclusion that some contamination must have happened here too. If the samples in Figures A35 and A38 have not been contaminated, something drastic has happened to them.

When studying the size distribution curves in Figures A1 to A12 it is clear that there is a consistency between the measurements of the samples when comparing the measurements from the same pH. When looking at the measurements within each sample as pH changes, a trend can be found in the form of a small increase in particle radius as pH decreases. This means that the more acidic the water, the faster the flocculation. However, the size distribution curve in Figure A12 indicates that the sample might have been contaminated when the acid was added before this measurement. Figures 3 to 5 show the results from Figures A1 to A12 put together in three graphs to make it easier to compare the results from the different pH measurements within each sample. Some samples are clearly contaminated but ignoring those, small changes between the pH measurements can be detected. Figures 9 to 12 show the results from figures A1 to A12 put together in four graphs according to pH to easier compare the samples to each other. There is clearly a consistency between the samples.

When studying the size distribution in Figures A13 to A24 it is even here clear that there is a consistency between samples. There is also a trend within samples, showing a small decrease in particle radius as pH increases. This means that the more alkaline the water, the slower the flocculation. The size distribution curve in Figure A23 indicates that the sample might have been contaminated before this measurement, but the fact that the size distribution curve in Figure A24 looks normal suggests that the contamination might have been on the outside of the cuvette or that it was just some particles floating by during the measurement.
Figures 6 to 8 show the results from Figures A13 to A24 put together in three graphs to make it easier to compare the results from the different pH measurements within each samples. Even here, some samples have been contaminated, but again, ignoring those, small changes between the pH measurements can be detected here as well. Figures 13 to 16 show the results from figures A13 to A24 put together in four graphs according to pH to easier compare the samples to each other. There is clearly a consistency between the samples even here.

When looking at Figures A25 to A30, which is the measurements of the acidic and alkaline samples at pH 3 and 9 after leaving them for a week, and comparing them to Figures A4, A8, A12, A16, A20 and A24, which are the curves for the samples at pH 3 and 9 from the first measurements, it is clear that time has an impact on flocculation. In both the acidic and alkaline samples, the particle radius has increased significantly. The increase in particle radius doesn’t differ very much between the acidic and alkaline samples. By the looks of the size distribution curve in Figure A26, that sample has most likely been contaminated. For this to easier be done, the results from measuring the fresh and a week old samples for each vial has been put together in six graphs that are presented in Figures 17 to 22. Looking at these graphs, it is quite clear that time has an impact on flocculation, regardless of the pH.

When looking at Figure A31 it is clear that CaCl$_2$ has quite an impact on flocculation since the particle radius is significantly larger than for all previous samples. Comparison is especially done to samples with pH 6, which is the pH of the raw lake water. Comparison to Figure A35, however, can not be done since there seem to have been contamination to the sample during the week between measurements. But looking briefly at Figure A35, the particle radius seems to be in the same range as in Figure A31.

When looking at Figures A32 to A34 it can be noted that the particle radius increases when the NaCl concentration increases. This means that salinity has an impact on flocculation. Comparing Figure A32 to Figure A36, Figure A33 to Figure A37 and Figure A34 to Figure A38 leads to the conclusion that time has an impact on flocculation even in saltier water.
Conclusions

1. The hydrophobic component in natural organic matter adsorbs to some filter materials but not to others. It is therefore important to consider which type of filter to use depending on what kind of solution to be filtrated.


3. pH has an impact on flocculation of DOC.

4. Salinity has an impact on flocculation of DOC.

5. CaCl₂ content has an impact on flocculation of DOC.

6. Time has an impact on flocculation of DOC in acidic, alkaline and salty water as well as water with a high CaCl₂ content.

7. Both the raw centrifuged water and all the samples is quite monodisperse.
References


Appendix

Figure A1. (a) Size distribution for acidic samples, vial 1 pH 6.  
(b) Correlation for acidic samples, vial 1 pH 6.

Figure A2. (a) Size distribution for acidic samples, vial 1 pH 5. 
(b) Correlation for acidic samples, vial 1 pH 5.

Figure A3. (a) Size distribution for acidic samples, vial 1 pH 4. 
(b) Correlation for acidic samples, vial 1 pH 4.

Figure A4. (a) Size distribution for acidic samples, vial 1 pH 3.  
(b) Correlation for acidic samples, vial 1 pH 3.
Figure A5. (a) Size distribution for acidic samples, vial 2 pH 6.
(b) Correlation for acidic samples, vial 2 pH 6.

Figure A6. (a) Size distribution for acidic samples, vial 2 pH 5.
(b) Correlation for acidic samples, vial 2 pH 5.

Figure A7. (a) Size distribution for acidic samples, vial 2 pH 4.
(b) Correlation for acidic samples, vial 2 pH 4.

Figure A8. (a) Size distribution for acidic samples, vial 2 pH 3.
(b) Correlation for acidic samples, vial 2 pH 3.
Figure A9. (a) Size distribution for acidic samples, vial 3 pH 6.
(b) Correlation for acidic samples, vial 3 pH 6.

Figure A10. (a) Size distribution for acidic samples, vial 3 pH 5.
(b) Correlation for acidic samples, vial 3 pH 5.

Figure A11. (a) Size distribution for acidic samples, vial 3 pH 4.
(b) Correlation for acidic samples, vial 3 pH 4.

Figure A12. (a) Size distribution for acidic samples, vial 3 pH 3.
(b) Correlation for acidic samples, vial 3 pH 3.
Figure A13. (a) Size distribution for alkaline samples, vial 1 pH 6. (b) Correlation for alkaline samples, vial 1 pH 6.

Figure A14. (a) Size distribution for alkaline samples, vial 1 pH 7. (b) Correlation for alkaline samples, vial 1 pH 7.

Figure A15. (a) Size distribution for alkaline samples, vial 1 pH 8. (b) Correlation for alkaline samples, vial 1 pH 8.

Figure A16. (a) Size distribution for alkaline samples, vial 1 pH 9. (b) Correlation for alkaline samples, vial 1 pH 9.
Figure A17. (a) Size distribution for alkaline samples, vial 2 pH 6. (b) Correlation for alkaline samples, vial 2 pH 6.

Figure A18. (a) Size distribution for alkaline samples, vial 2 pH 7. (b) Correlation for alkaline samples, vial 2 pH 7.

Figure A19. (a) Size distribution for alkaline samples, vial 2 pH 8. (b) Correlation for alkaline samples, vial 2 pH 8.

Figure A20. (a) Size distribution for alkaline samples, vial 2 pH 9. (b) Correlation for alkaline samples, vial 2 pH 9.
Figure A21. (a) Size distribution for alkaline samples, vial 3 pH 6.
(b) Correlation for alkaline samples, vial 3 pH 6.

Figure A22. (a) Size distribution for alkaline samples, vial 3 pH 7.
(b) Correlation for alkaline samples, vial 3 pH 7.

Figure A23. (a) Size distribution for alkaline samples, vial 3 pH 8.
(b) Correlation for alkaline samples, vial 3 pH 8.

Figure A24. (a) Size distribution for alkaline samples, vial 3 pH 9.
(b) Correlation for alkaline samples, vial 3 pH 9.
Figure A25. (a) Size distribution for acidic samples, vial 1 pH 3, after a week. (b) Correlation for acidic samples, vial 1 pH 3, after a week.

Figure A26. (a) Size distribution for acidic samples, vial 2 pH 3, after a week. (b) Correlation for acidic samples, vial 2 pH 3, after a week.

Figure A27. (a) Size distribution for acidic samples, vial 3 pH 3, after a week. (b) Correlation for acidic samples, vial 3 pH 3, after a week.

Figure A28. (a) Size distribution for alkaline samples, vial 1 pH 9, after a week. (b) Correlation for alkaline samples, vial 1 pH 9, after a week.
Figure A29. (a) Size distribution for alkaline samples, vial 2 pH 9, after a week. 
(b) Correlation for alkaline samples, vial 2 pH 9, after a week.

Figure A30. (a) Size distribution for alkaline samples, vial 3 pH 9, after a week. 
(b) Correlation for alkaline samples, vial 3 pH 9, after a week.

Figure A31. (a) Size distribution for sample with 100 µl CaCl₂, pH 6. 
(b) Correlation for sample with 100 µl CaCl₂, pH 6.

Figure A32. (a) Size distribution for sample with 5 g/l NaCl, pH 6. 
(b) Correlation for sample with 5 g/l NaCl, pH 6.
Figure A33. (a) Size distribution for sample with 15 g/l NaCl, pH 6.
   (b) Correlation for sample with 15 g/l NaCl, pH 6.

Figure A34. (a) Size distribution for sample with 35 g/l NaCl, pH 6.
   (b) Correlation for sample with 35 g/l NaCl, pH 6.

Figure A35. (a) Size distribution for sample with 100 µl CaCl$_2$, pH 6, after a week.
   (b) Correlation for sample with 100 µl CaCl$_2$, pH 6, after a week.

Figure A36. (a) Size distribution for sample with 5 g/l NaCl, pH 6 after a week.
   (b) Correlation for sample with 5 g/l NaCl, pH 6, after a week.
**Figure A37.** (a) Size distribution for sample with 15 g/l NaCl, pH 6 after a week.  
(b) Correlation for sample with 15 g/l NaCl, pH 6, after a week.

**Figure A38.** (a) Size distribution for sample with 35 g/l NaCl, pH 6 after a week.  
(b) Correlation for sample with 35 g/l NaCl, pH 6, after a week.