NTNU | Department of Chemical Engineering

TKP4580 - Specialization Project

Droplet Distribution and Overflow Estimation for a Hydrocyclone Lab

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Abstract

Hydrocyclone separation is a commonly used technology for the treatment of produced water, which is a biproduct often occurring in the oil- and gas industry. The 'Compact Separation Laboratory' located at Norsk Hydroteknisk Laboratorium allows for studying the performance of hydrocyclone separation.

The main aim of this project is to experimentally determine some key properties of the lab setup. One of the aspects focused on is the droplet distribution of the inlet flow to the hydrocyclone. This has been investigated by the use of an offline sensor. A current issue with the lab is that the hydrocyclone overflow is not being measured. Therefore, three potential methods to estimate the unknown overflow have been tested. The methods include using the total mass balance of the hydrocyclone, the valve equation and experimentally measuring the flow. Additionally, the pressure-flow relationship for the overflow and underflow of the hydrocyclone has been researched and compared with simulated results from a previous first principles model.

Measuring the inlet droplet distribution led to the conclusion that it is log-normally distributed, and shifting when varying the inlet flow valve opening. The mass balance proved to be a reasonably accurate method of overflow estimation, as it provided results similar to the experimentally measured results. The pressure-flow relationship for the hydrocyclone followed a similar trajectory to previously simulated results.

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1 Introduction

Produced water is a common biproduct in the oil- and gas industry. The majority of offshore produced water treatment is done by use of deoiling hydrocyclones, which were introduced as an efficient and compact alternative to gravity separators in the early 1980's. In addition to their compactness, hydrocyclones require little maintenance, are easy to operate and have no moving parts. These qualities make them suitable for offshore and subsea applications [1].

A hydrocyclone separates produced water into an oil-rich overflow stream, and a treated water underflow stream. The treated water may then be discharged directly to the sea, as long as it fulfills the Norwegian requirement of a maximum of 30 mg of oil per liter of water [2].

The 'Compact Separation Laboratory' at Norsk Hydroteknisk Laboratorium allows for researching the performance of de-oiling hydrocyclones. The main aims of this project is to perform experiments to investigate certain aspects of the experimental process.

One of the main elements that will be considered in this project is the droplet distribution of the inlet flow to the hydrocyclone. This is one of the key inputs to the hydrocyclone, which is considered valuable to investigate. Also, a potential improvement that could be made for the laboratory is estimating the hydrocyclone's overflow, which is currently not being measured. An additional aim is to experimentally determine the pressure-flow relationship for the underflow and overflow, and compare the results with simulated results [3].

The report is structured in the following manner: In section 2 the necessary theoretical background for understanding the results and methods is described. The experimental methods that have been used are given in section 3, while the corresponding results are presented and analyzed in section 4. Additional analysis of the results, as well as a discussion regarding potential experimental errors and future work is presented in section 5. Finally, the main conclusions of the project are given in section 6.

2 Background

2.1 Hydrocyclones

A hydrocyclone consists of a tangential inlet chamber where the produced water enters, Q_{in}, an underflow where the treated water exits, Q_U, and an overflow where the lighter oil-rich stream comes out, Q_O. Figure 1 shows a simplified sketch of a hydrocyclone with the three mentioned streams marked.

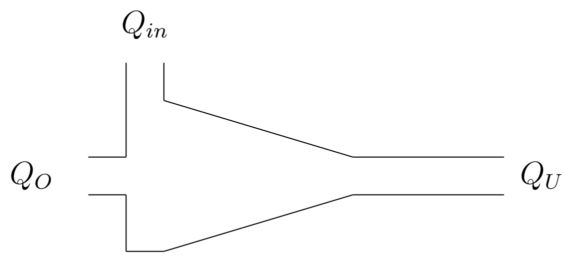

Figure 1: Hydrocyclone

The hydrocyclone works by utilising pressure energy to separate phases which have different densities. The oil-water mixture enters the inlet tangentially, which results in a vortex system. This creates a high acceleration field, which force the oil to the hydrocyclone's center, and the heavier phase through the underflow [1].

One of the key criteria for determining the hydrocyclone's performance is the separation efficiency, η, which is given as: [4]

$$\eta = 1 - \frac{c_{U,o}}{c_{in,o}} \tag{1}$$

In Equation (1), $c_{U,o}$ and $c_{in,o}$ are the concentrations of oil in the underflow stream and inlet stream respectively. The goal is to achieve the highest possible separation, meaning a value of η close to 1.

In addition to obtaining a high separation efficiency, a well-performing hydrocyclone should have low flow split, which is given by Equation (2): [3]

$$F_s = \frac{Q_O}{Q_{in}} \tag{2}$$

2.2 Droplet Distribution

The oil droplets were originally assumed to be normally distributed. However, some initial experimental analysis showed that they rather might be log-normally distributed.

The log-normal distribution applies if the natural logarithm of a random, continuous variable is normally distributed with a mean of μ and a standard deviation of σ [5]. Log-normal distributions appear when there are multiplicative processes, as opposed additive processes, which result in normal distributions [6].

An example of log-normal distributions with $\mu = 1$ and different standard deviations σ is given in Figure 2:

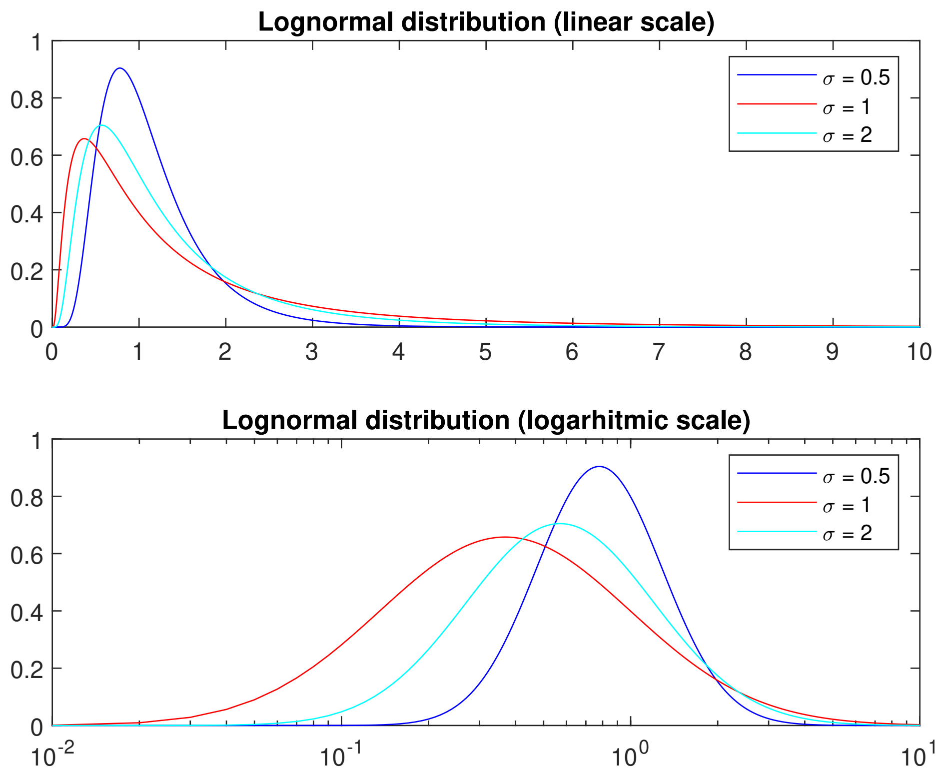

Figure 2: Log-normal distributions with varying values of σ. Generated in Matlab D.4

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As the figure illustrates, log-normal distributions have the appearance of normal distributions when plotted on a logarithmic scale, following from the property that the logarithm of a log-normally distributed variable is normally distributed.

For a normal distribution, the mode, median and mean are identical. However, for the log-normal distributions, the mode, median and mean may differ greatly. The difference is especially noticeable for large values of σ.

The probability density function for the log-normal distribution is: [7]

$$\frac{1}{\sqrt{2\pi}\sigma x}\exp\left(-\frac{(\log(x)-\mu)^2}{2\sigma^2}\right), \text{for x} > 0 \tag{3}$$

Using the calculated mean and variance from an analysed sample, it is possible to obtain an estimate for future droplet distributions at the given inlet valve opening, Z_{in}. The log-normal distribution parameters μ and σ may be calculated by using Equation (4) and (5),

$$\mu = \log(\frac{m^2}{\sqrt{v+m^2}}) \tag{4}$$

$$\sigma = \sqrt{\log(\frac{v}{m^2}+1)} \tag{5}$$

where m and v denote the calculated mean and variance respectively [8].

Using the calculated values σ and μ, it is possible to generate a log-normal distribution by applying the Matlab-function 'makedist' [9].

2.3 Overflow Estimation

While there are flowmeters installed to measure the inlet flow and underflow, the overflow of the hydrocyclone is currently not being measured. Therefore it is desirable to somehow estimate the overflow. The theoretical background for three different methods of overflow estimation is given in this section.

2.3.1 Mass Balance

The simplest way to estimate the overflow, Q_O, is to use the mass balance of the hydrocyclone:

$$Q_{in} = Q_U + Q_O \tag{6}$$

Since Q_{in} and Q_U are measured by flowmeters, the overflow can be estimated by Equation (7):

$$Q_O = Q_{in} - Q_U \tag{7}$$

One important assumption made when using the mass balance to estimate the overflow, is that the flowmeter measuring the underflow has a consistent error. This error was noticed when keeping the overflow valve opening completely closed. Both flowmeters should then give the same value, but this was not the case. Instead the flowmeter measuring the underflow showed a value approximately 0.1 m^3/h higher than the inflow measurement. This has been taken into account when using the mass balance method for overflow estimation.

2.3.2 Valve Equation

A second alternative for overflow estimation is to base it on the valve equation: [3]

$$Q_O = C_{vO} Z_O \sqrt{\frac{2\Delta P}{\rho_O}} \tag{8}$$

In Equation (8), C_{vO} represents the constant for the overflow valve, Z_O the valve opening, ρ_O the oil density, while ΔP is the pressure loss for the overflow. Z_O will be varied throughout the experiments and the corresponding pressure drop will be measured. One assumption made is that the overflow consists entirely of oil. The oil being used, 'EXXSOL D60', has a density of 793 kg/m^3 [10]. For the valve constant C_{vO}, different values have been tried to find a value that best corresponds with measured overflow values.

2.3.3 Measured Overflow

It is possible to measure the overflow by measuring the liquid level of the oil reject tank after having the overflow stream enter it for a certain amount of time. The cross-sectional area of the tank is given as:

$$A = \pi r^2 \tag{9}$$

where r is the inner radius of the tank.

The overflow can then be measured by dividing the added volume by the elapsed time:

$$Q_O = \frac{\Delta h A}{t} \tag{10}$$

where Δh is the measured difference in liquid level. The resulting average of Q_O may serve as a baseline for evaluating the results when using the mass balance or valve equation.

3 Experimental

As part of this project, numerous experiments were performed, following a few different procedures. The procedures to perform the experiments are described in this section, as well as a basic overview of the most relevant parts of the 'Compact Separation Laboratory'.

3.1 Compact Separation Laboratory

The most relevant parts of the laboratory include a hydrocyclone as previously described in subsection 2.1. Additionally, each of the hydrocyclone's streams has a corresponding valve. The valve openings are denoted by Z_{in}, Z_U and and Z_O respectively. Z_U has been kept constant at 0.6 for the experiments, while Z_{in} was varied while investigating the inlet droplet distribution. For the overflow estimation, different values of Z_O have been used.

A LabVIEW program is used for the operation of the laboratory. Key inputs such as the valve openings, and the oil-concentration and flow rate of the produced water inflow, are achieved according to set-points given in the program. The program registers the data that is necessary for the overflow estimation, that is the measured flow rates, as well as the pressure drops of the hydrocyclone.

The concentration and distribution of Q_{in} and Q_U are supposed to be measured by two online oil-in-water sensors. However, the values which the sensors produced were questionable, leading to the use of an offline sensor instead.

3.2 Droplet Distribution Measurement

An external Mastersizer 3000 (MS3000) sensor has been used to investigate the droplet size-distribution of the inlet flow. It consists of an optical unit, a dispersion unit and a measurement cell. The optical unit works by transmitting red and blue laser light through the sample. Detectors then use the light scattering pattern to provide information about the sample [11]. The sensor also has the capability of measuring the concentration, however this aspect has been focused on to a smaller extent.

The first step of the procedure is to initialize the instrument with pure water in order to measure the background and assure that the system is clean.

The next step is to extract a sample from the sample bomb, which is located after the inlet flow valve. A previously developed procedure for sample extraction has been followed [12], which is detailed in Appendix C.

The extracted sample is then added to a beaker and if necessary, diluted until an appropriate laser obscuration is achieved. The laser obscuration value should be between 10-20%, for wet samples [13].

After the measurement of the sample the system is cleaned by running the instrument with pure water, in addition to 2-propanol if needed. Additionally, the measurement cell may be ejected and cleaned using a fibreless cloth, which was deemed necessary every 3-4 samples.

A more detailed procedure, developed by Marcin Dudek and Anders Andersen, is described in Appendix B [13].

3.3 Overflow Estimation

The data used for the overflow estimation was gathered by running the system at a constant volume flow $Q_{in} \approx 2.3$ m^3/h, while varying the overflow valve opening, Z_O. The data gathered includes the inflow and underflow of the hydrocyclone, both measured by flowmeters. Equation (7) was then used for the mass balance estimation. The inlet pressure, as well as the pressure drops in both the overflow and underflow are registered as well. These values were needed to apply Equation (8).

The overflow was also measured experimentally by investigating the liquid level of the tank where the overflow exits. The level change of the tank, Δh was measured by use of a measurement tape, after keeping Z_O and Q_{in} constant for approximately 10 minutes. Equation (10) was then used to calculate the average flow rate.

3.4 Pressure-Flow Relationship

The pressure drop over the underflow and overflow, as a function of the flow rate into the hydrocyclone, was determined experimentally. This was achieved by varying the inlet flow rate, Q_O, from low to high values. Subsequently the corresponding pressure-flow relationship was observed.

4 Results

4.1 Droplet Distribution

The droplet distribution for the inlet flow to the hydrocyclone has been investigated for a variety of different inlet valve openings, Z_{in}. Different aspects of the distribution results are presented in the following sections.

4.1.1 Lognormal Distribution

Figure 3 shows the resulting droplet distribution for 50% inlet flow valve opening, plotted on a normal scale. The x-axis represents the different droplet size (diameter) categories, while the y-axis denotes the percentage of the total volume which belongs to each size.

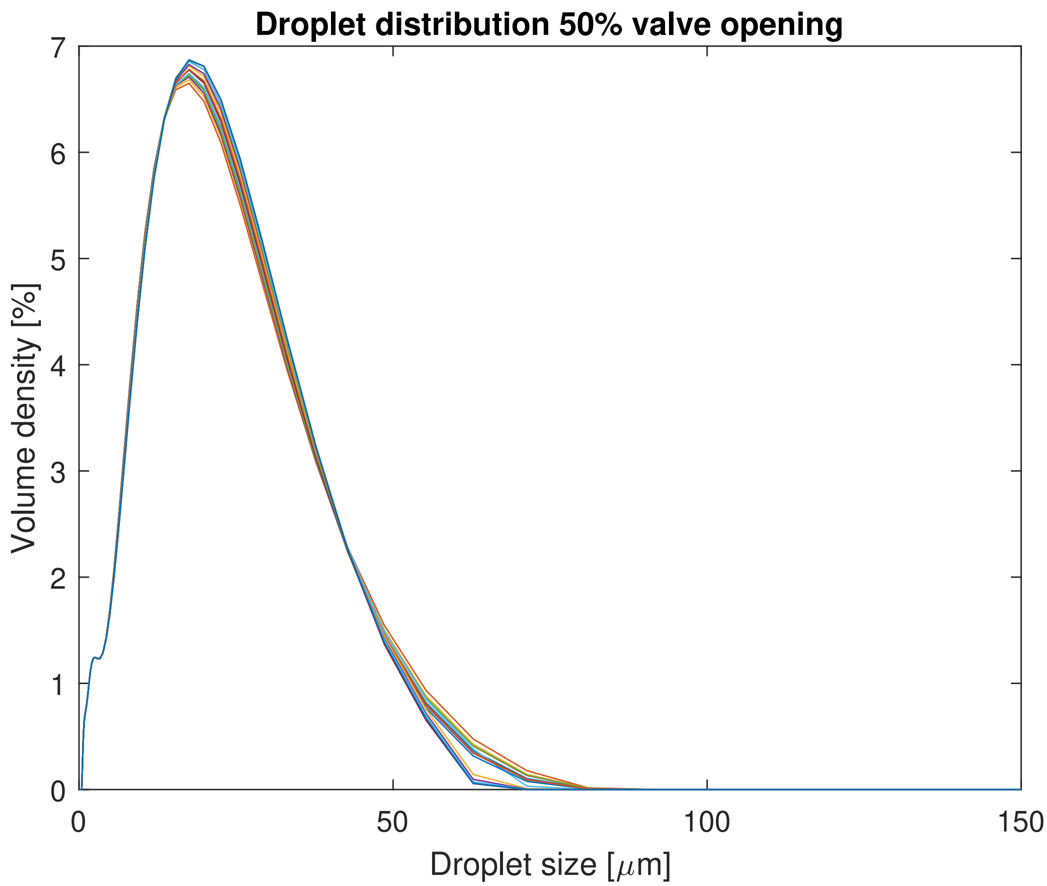

Figure 3: Droplet distribution - 50% valve opening

Figure 4 shows the exact same distribution as in Figure 3, but plotted on semi-log scale.

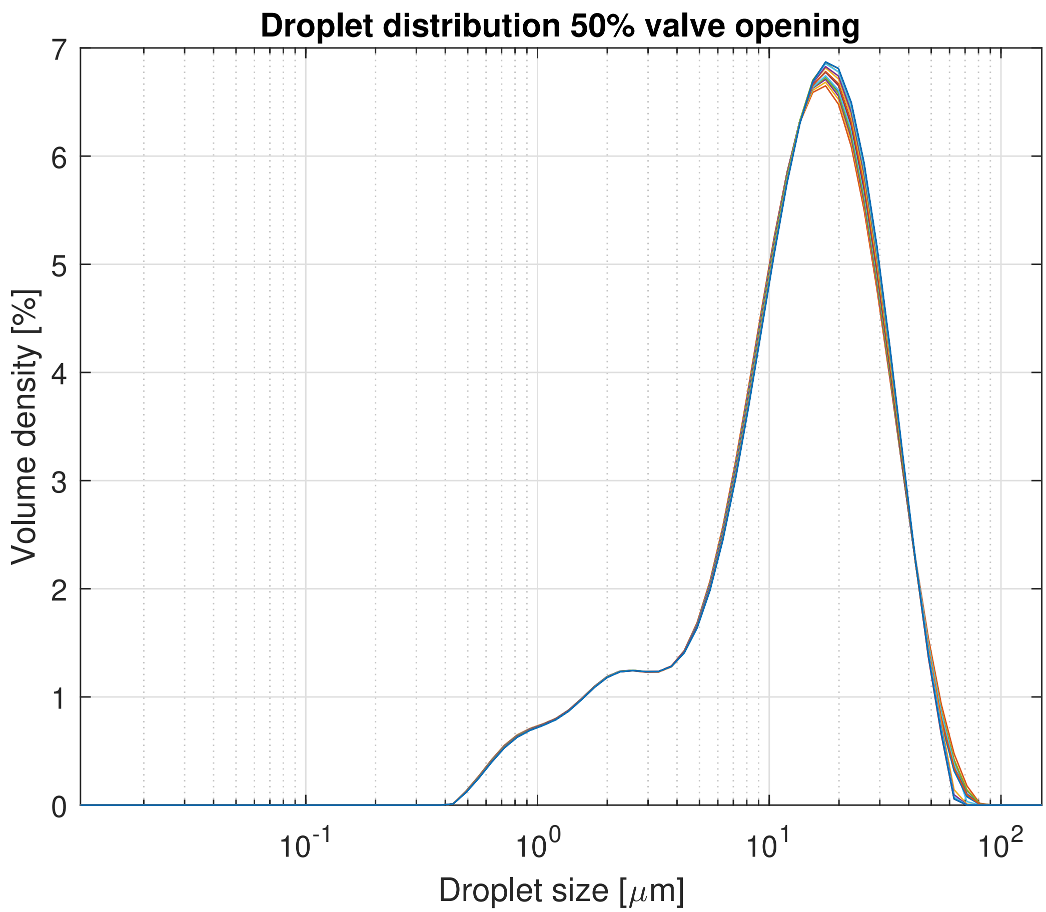

Figure 4: Droplet distribution - 50% valve opening - semi-log scale

Figure 3 and 4 importantly demonstrate that the droplet distributions are log-normally distributed. This is evident because the distribution has the appearance of a normal distribution when plotted on a semilog scale, as was demonstrated in principle in Figure 2. However, it is certainly not perfectly log-normally distributed, as it is not entirely symmetrical. Rather, the distribution of this sample has tail on left side of the x-axis, which was the case for many of the investigated samples.

It is also worth noting that both figures contain 15 different graphs, as the Mastersizer was set to perform 15 measurements for a given sample. It is observed that the curves mostly follow the same trajectory, which substantiates the validity of the given sample.

4.1.2 Comparison of Distributions

Figure 5 shows three distributions at different values of Z_{in}. In this case the 15 measurements for each sample have been combined to create an average curve for each of the three samples.

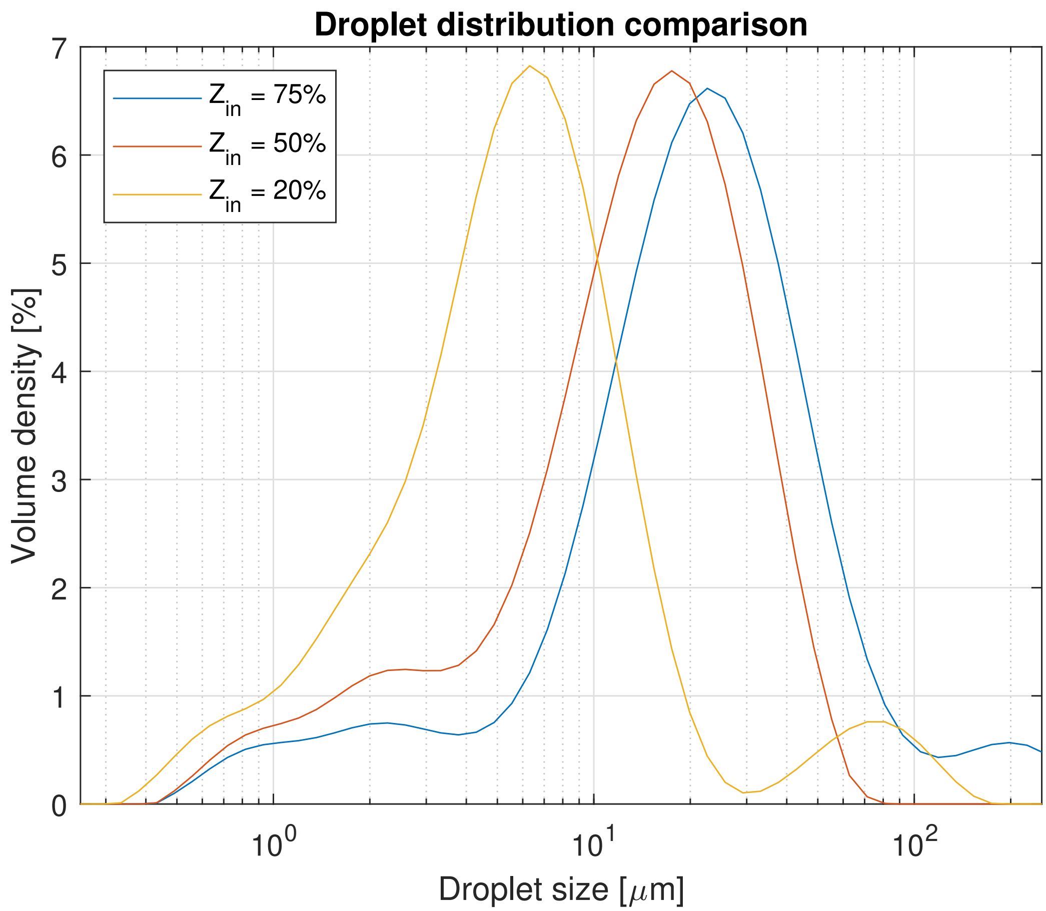

Figure 5: Comparison of droplet distributions

The main takeaway from this result is that the distribution is shifting when varying the inlet valve opening. The probable reason for this behaviour is that a portion of the droplets are breaking at smaller valve openings, due to the resulting increased pressure.

4.1.3 Distribution Trends

Figure 6 illustrates the 10th, 50th and 90th percentile, denoted by Dx10, Dx50 and Dx90 respectively, and mean droplet size for several samples. The purpose is to further demonstrate the variation as a function of the inlet flow valve opening.

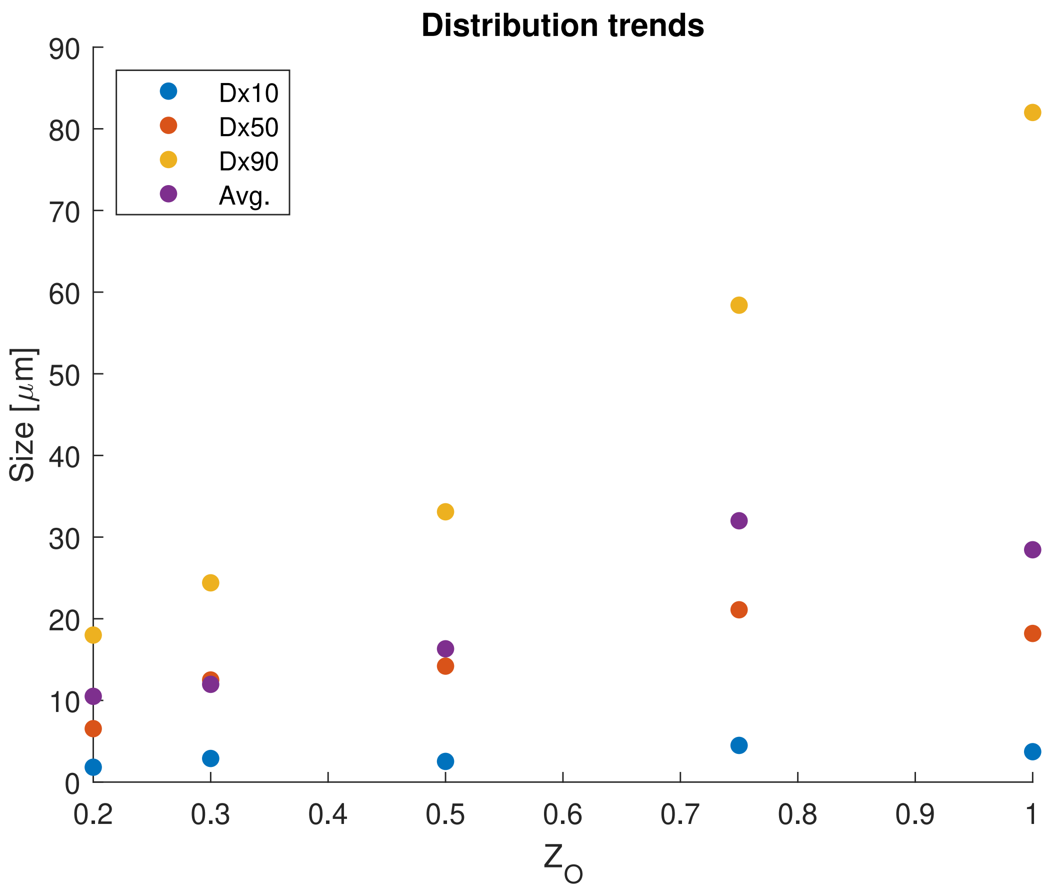

Figure 6: Dx10, Dx50, Dx90 and mean as a function of valve opening

The results are also represented in Table 1, which additionally contains the observed modes, as well as the calculated means (m) and standard deviations (s).

Table 1: Droplet distribution trends

Z_{in}	Dx10	Dx50	Dx90	Mode	m	s
20%	1.82	6.54	18.0	6.31	10.5	18.3
30%	2.89	12.5	24.4	11.9	12.0	7.11
50%	2.53	14.2	33.1	17.5	16.3	12.0
75%	4.49	21.1	58.4	22.6	32.0	43.2
100%	3.72	18.2	82.0	15.4	28.5	29.3

It is further demonstrated that the droplet distribution increases when the inlet valve opening increases. In particular, the presence of larger droplet increases, as can be observed by the rapidly increasing Dx90 values. It should also be noted that the average value for $Z_O = 100\%$ is smaller than for 75%. The probable reason for this is that the sample was of bad quality, leading to measurement errors.

4.1.4 Separable Volume

One important aspect to consider is that the hydrocyclone is unable to separate droplets under the size of 10 μm. Figure 7 shows the distribution of a sample where $Z_{in} = 30\%$. Additionally, the areas of separable and inseparable droplet sizes are indicated in the figure.

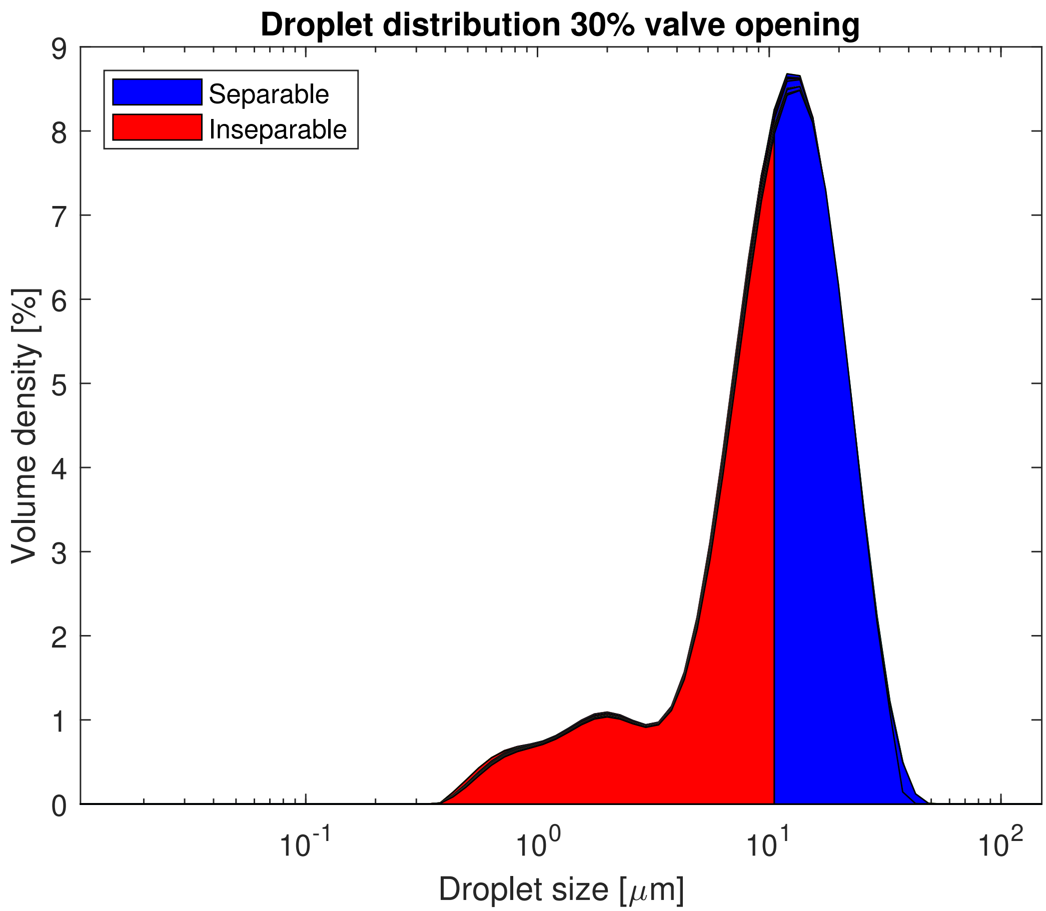

Figure 7: Separable volume - 30% valve opening

The separable volume percentage may be calculated by integrating over the relevant area of the droplet distributions. The relevant area is defined as the size range of droplets that are separable, indicated by blue in Figure 7. The volume percentage is then found by numerically integrating over the relevant area of the curve using the Matlab function 'trapz'. 'trapz' computes an approximation of the integral using the trapezoidal method [14].

Figure 8 shows the percentage of separable volume for several samples as a function of inlet flow valve opening, Z_{in}.

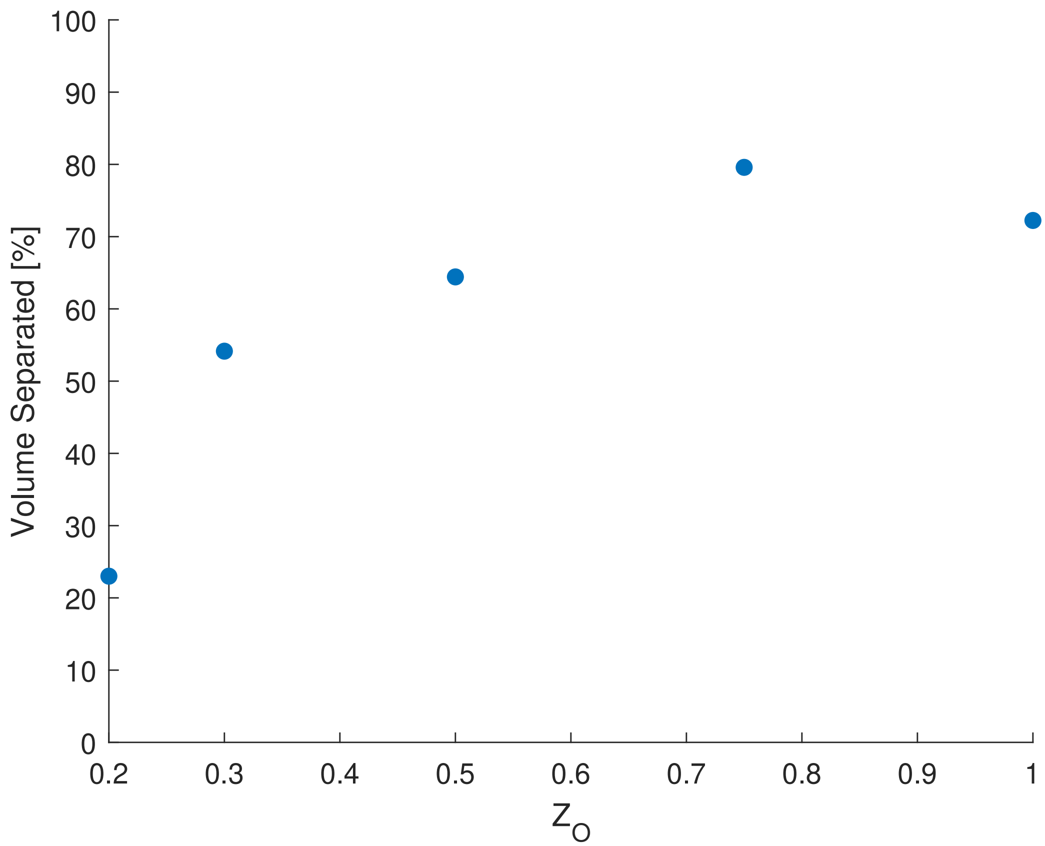

Figure 8: Percentage of separable volume

A general trend of a higher percentage of separable volume when increasing the droplet size can be observed. This is due to the samples containing a larger degree of inseparable droplets when Z_O is low. Since the goal of the hydrocyclone is to separate as much of the oil as possible, it is evident that operating at a high value of Z_O is beneficial.

4.1.5 Number Distribution

Figure 9 shows an example of the number distribution of a sample. Instead of the total percentage of volume on the y-axis, the number of droplets is plotted.

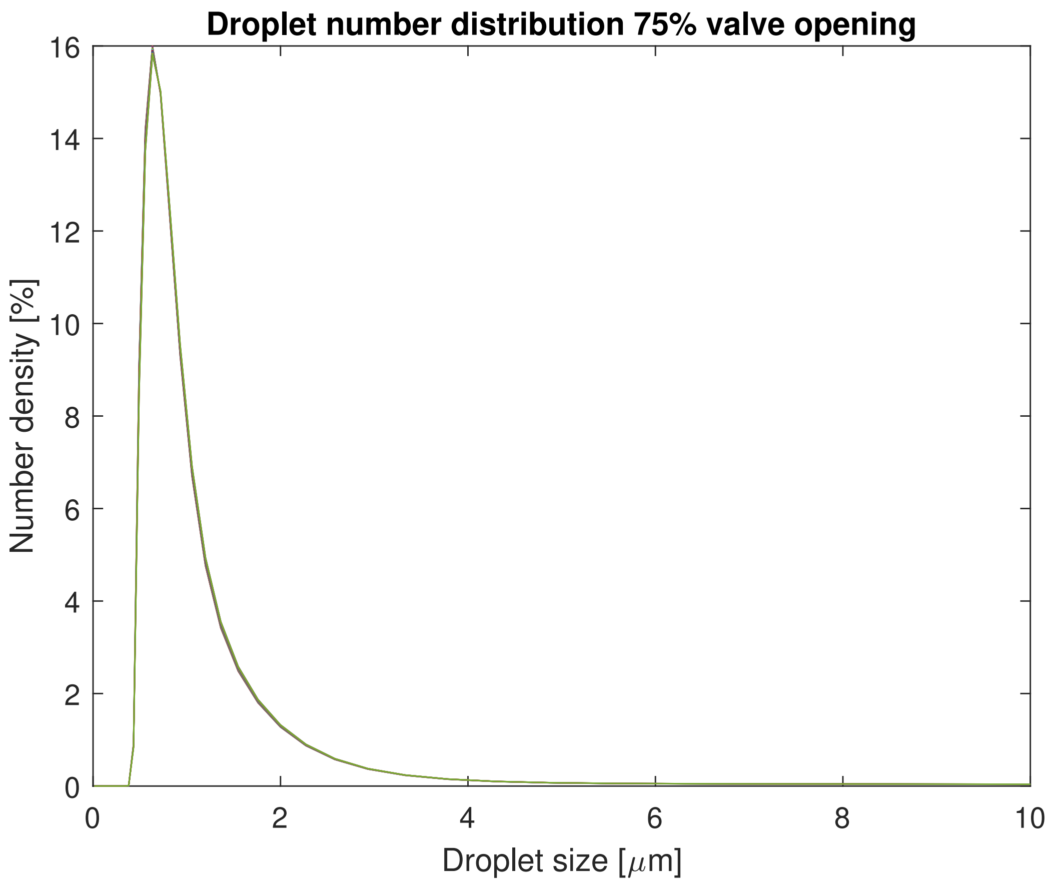

Figure 9: Number distribution - 75% valve opening

It is clear that the vast majority of the droplets belong to the smaller size categories. Most of the droplets are 4 μm or smaller, however since they are insignificantly small they make up a very small portion of the total volume.

4.1.6 Effect of Sample Concentration

The dilution process leads to somewhat differing values for the concentration of each sample. Therefore, it is desirable to know how the concentration effects the resulting distribution. Figure 10 shows the droplet distribution for the exact same conditions, except different volume concentrations, c_v. The same sample has been investigated, however at different levels of dilution. Both curves are average values of 15 measurements of the sample.

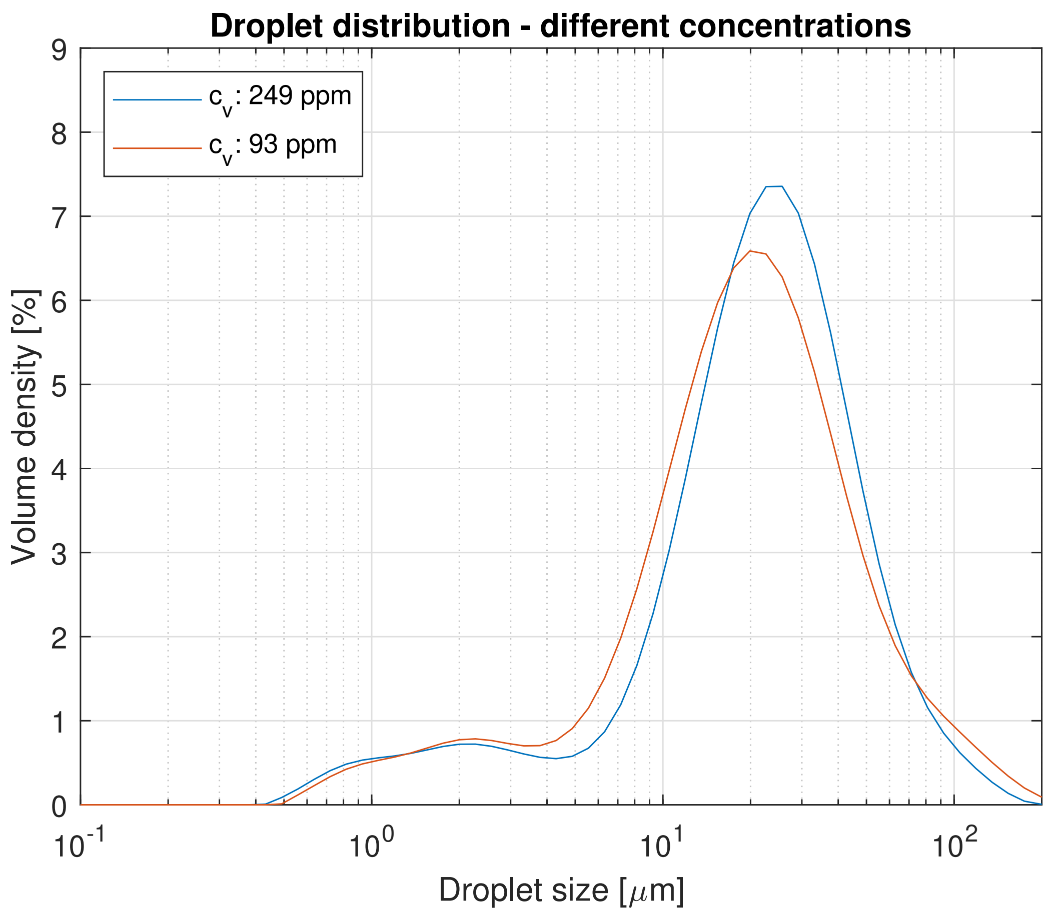

Figure 10: Average droplet distribution for 75% valve opening - differing concentrations

The sample with the higher concentration has an average distribution shifted somewhat to the right. Other than that they share a relatively similar distribution. It is therefore assumed that the concentration's effect on the sample is negligible. However, it is as previously mentioned important that the concentration is within the range that gives a suitable laser obscuration when measuring with the Mastersizer. It may be worth further investigating the effect of the sample concentration, as only one measurement may be to little data to make a definite conclusion. Nevertheless, the effect of the sample concentration is assumed to be insignificant.

4.1.7 Estimated Distribution

Figure 11 compares the actual droplet distribution for a sample where $Z_O = 50\%$ and an estimated distribution.

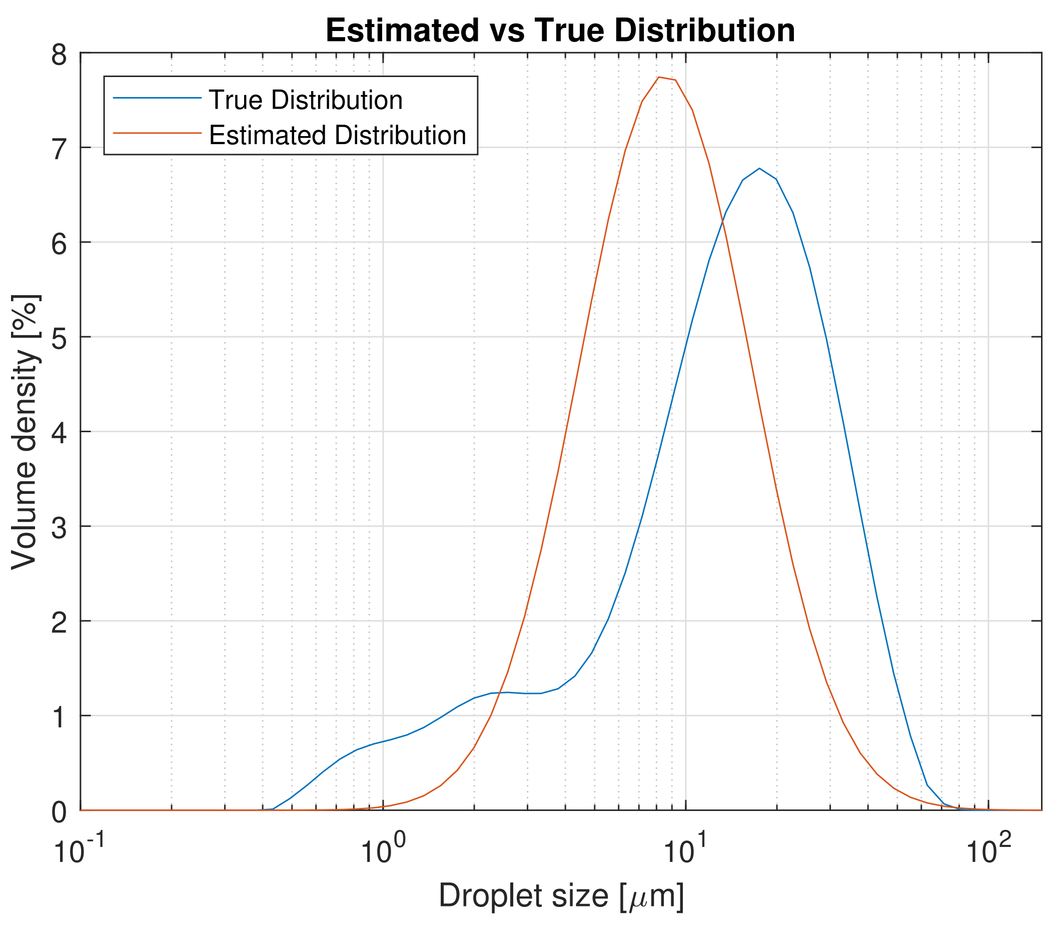

Figure 11: Estimated vs true distribution 50% valve opening

The estimation is a log-normal distribution generated in Matlab, using the method as described in subsection 2.2. More specifically, the log-normal distribution parameters are calculated and used as inputs in the 'makedist' function.

It is easily observable from the figure that the estimation is not particularly accurate, as there is quite a shift between the estimated and actual distributions. If the measured distribution was completely symmetrical, the real and estimated distributions would be identical. However, this is as previously mentioned not the case, which leads to the deviation.

4.2 Overflow Estimation

Since the overflow is currently not being measured, a few different methods to estimate it have been investigated.

4.2.1 Mass Balance

Figure 12 gives an overview of the estimated Q_O, while varying the overflow valve opening, Z_O, from 10% to 80%. The estimated results are given for 1000 samples for each Z_O-value.

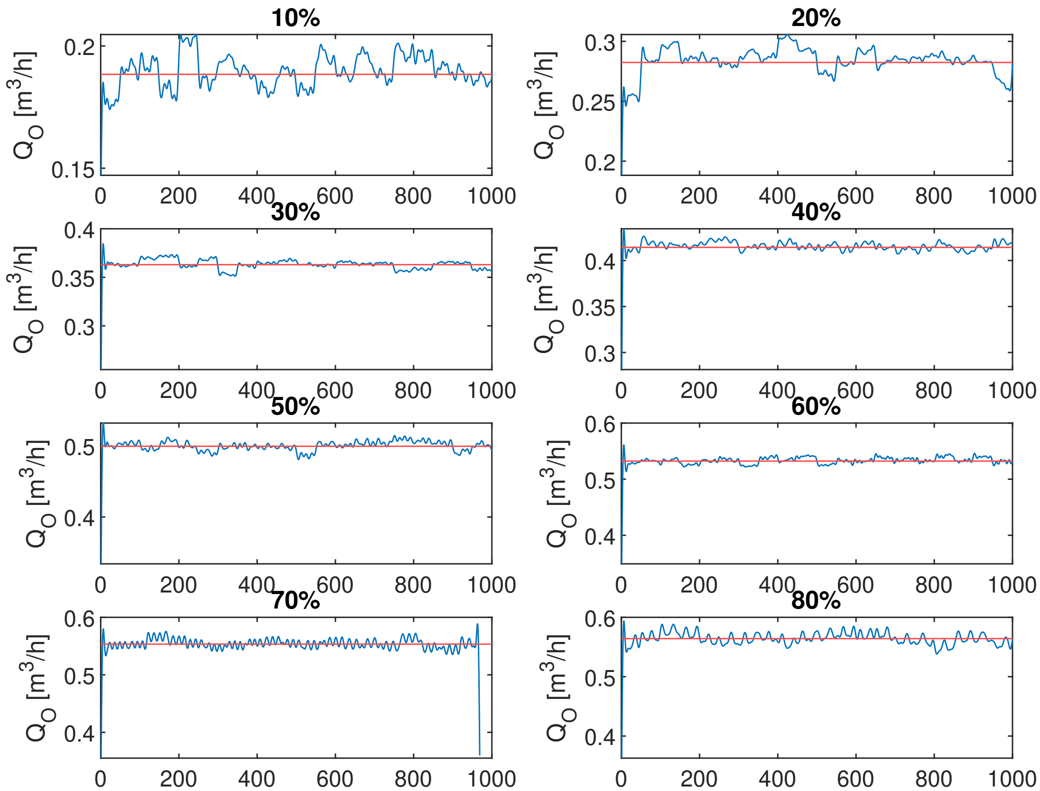

Figure 12: Mass balance estimated overflow

The horizontal red lines represent the average values, which help demonstrate the variation of the live values. The values are somewhat oscillatory, especially for lower values of Z_O. This is problematic since it is the live values that are of interest for the operation of the lab, not the averages. The estimated overflow value should ideally be as stable as possible.

4.2.2 Valve Equation

Figure 13 shows the results of applying the valve equation to estimate the overflow.

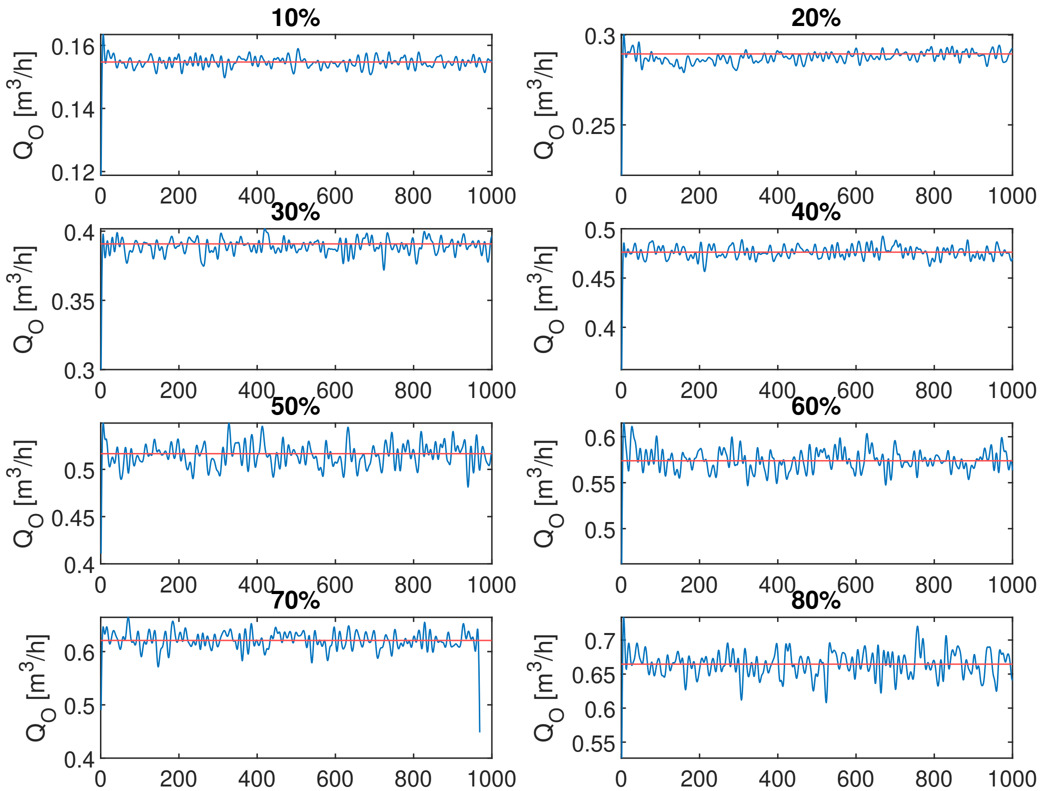

Figure 13: Valve equation estimated overflow

As with the mass balance estimation, the valve equation does not produce stable live values.

The reason why neither method provides stable results, may be due to measurement noise. The flowmeters and pressure transmitters which are providing the values, do not necessarily give entirely stable and accurate values. It is worth mentioning that these values have been filtered in Matlab using a low-pass filter. However, there is still a quite significant degree of variation.

4.2.3 Comparison with Experimental Values

Figure 14 compares the average values of the mass balance and valve equation methods and the experimentally measured overflow.

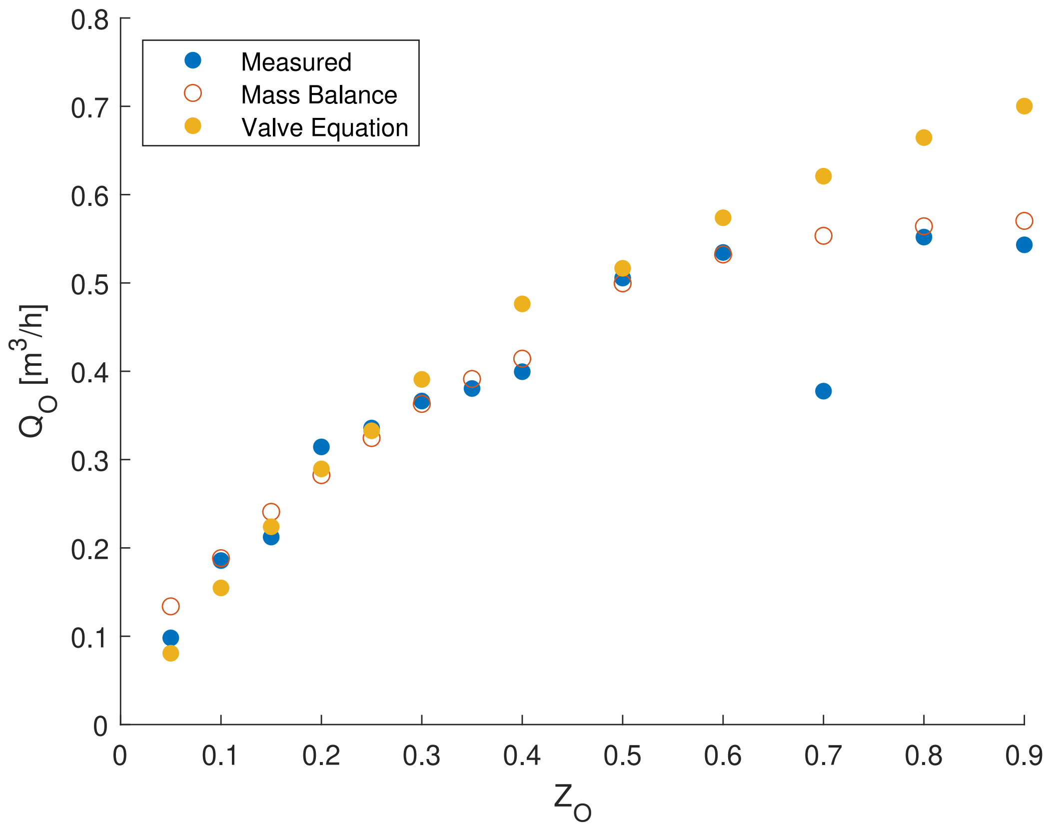

Figure 14: Measured and estimated overflow as a function of overflow valve opening.

The results are also given in Table 2. It is important to note that these values are averages, gathered over a time period of approximately 10 minutes.

Table 2: Measured vs estimated overflow

Z_O [%]	$Q_{O,meas}$ $[m^3/h]$	$Q_{O,mass}$ $[m^3/h]$	$Q_{O,valve}$ $[m^3/h]$
5	0.0982	0.134	0.0807
10	0.186	0.188	0.155
15	0.212	0.241	0.224
20	0.314	0.282	0.290
25	0.336	0.324	0.333
30	0.366	0.363	0.391
35	0.381	0.391	0.437
40	0.400	0.414	0.476
50	0.506	0.500	0.517
60	0.535	0.532	0.574
70	0.378	0.553	0.621
80	0.552	0.564	0.665
90	0.543	0.570	0.700

It is observed that the mass balance estimated and measured flow are quite closely correlated. The measured overflow for $Z_O = 0.7$ is a clear outlier, which probably is due to an erroneous measurement of the change in tank level, Δh.

The valve equation produces values that deviate to a larger degree from the measurements, especially for higher values of Z_O. The challenge of using the valve equation is obtaining the correct value for the valve constant Cv_O.

Figure 14 shows the results when Cv_O is assumed to be 0.05. This value was chosen because it produced results similar to the measured results, at least for the lower valve openings. However, it is observable that valve equation results has a higher slope than the measured and mass balance results. Due to this, whatever value is chosen will lead to deviation for a certain range of the results. There may be some non-linearity with regards to the valve equation, which should be taken into account. A potential solution may be to use different values for Cv_O, depending on the value of Z_O.

4.3 Pressure-Flow Relationship

An additional aim was to validate the pressure-flow relationship of the model. This was investigated by obtaining experimental results and comparing them with the results from the simulation. The simulated results have previously been compared to external experimental results from the 1980's and were proven to be similar [3]. However, it was deemed useful to compare them to newer results obtained from the compact separator lab. Figure 15 shows the experimentally obtained pressure drop as a function of different volume flows, Q_{in}.

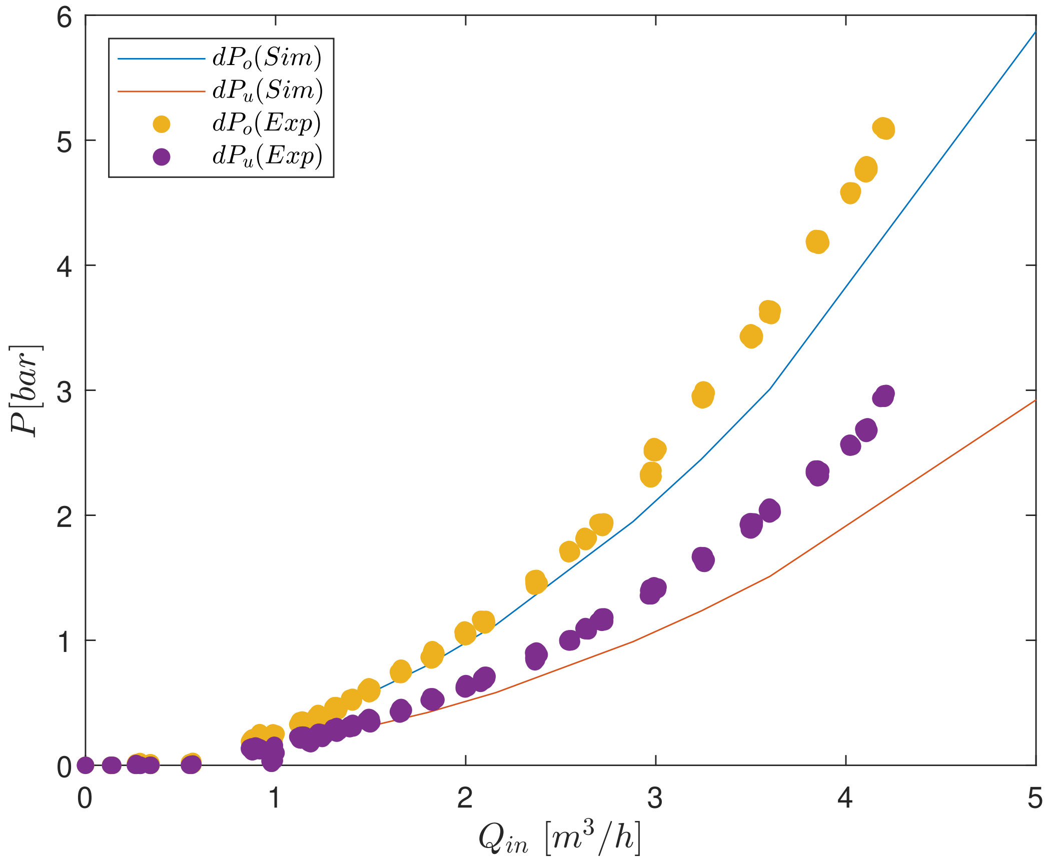

Figure 15: Pressure-flow relationship

It is evident that the pressure drop in the overflow is higher than in the underflow. The results follow the same trajectory as the simulated results, however there is some deviation. The deviation is probably due to the fact that the simulated values are missing a friction factor, which is yet to be included in the model.

5 Discussion

5.1 Droplet Distribution

There are a number of factors that should be kept in mind when considering the validity of the droplet distribution results. For one it is crucial that the equipment used to obtain the sample is clean, in order to achieve an uncontaminated sample. Ideally the sample should contain only one peak, however, many of the samples analyzed contained two or three peaks, meaning they had to be discarded.

Another thing to keep in mind is how the 15 measurements of the sample compare to each other. Ideally they should follow approximately the same trajectory, but on some occasions there were significant disparities between the measurements.

The validity of the obtained droplet distributions could be further improved by replicating the experiments. If more experiments under the same conditions would give similar results, then one could be more confident in the results. This was done to a certain degree, however even more measurements could be beneficial.

It should be noted that a portion of the larger droplets are potentially broken during the sampling process. Since the samples are extracted through a narrow passage, a certain degree of droplet breakage may have occurred. This could mean that the actual distribution is larger than what has been measured by the Mastersizer.

The distribution estimation, as seen in Figure 11, was deemed to be ineffective. This is due to the distribution being quite skewed, meaning a generated log-normal distribution will not accurately predict the actual distribution. However, the online sensors will have the capability of measuring the distribution as well, when they are fully operational. Therefore, the inaccuracy of the distribution estimation is not considered to be of great importance.

5.2 Overflow Estimation

There are a few possible errors that may have occurred during the measurement of the overflow, the must crucial being reading the wrong liquid level of the tank. This is the probable explanation for the outlier at $Z_O = 0.7$ in Figure 14. Another possible error could be inaccurate time measurement. However, since the measurements are averaged over a time of approximately 10 minutes for each measurement, an error of a few seconds would probably not have a significant impact on the results.

When estimating the overflow, an assumption was made that the flowmeter measuring the underflow is off by 0.1 m^3/h, which was taken into account when using the mass balance. This assumption was based on observing that this flowmeter showed 0.1 m^3/h higher than the inlet flowmeter when the overflow was completely closed. However, this assumption may be inaccurate. It is possible that the error is not absolute, but rather varying percentage-wise. To further elaborate, it may be the case that the measurement error changes with varying flow, instead of being consistently off by 0.1 m^3/h.

When using the valve equation, it was assumed that Q_O consists solely of oil. In reality, there is most certainly a quite large portion of water in the overflow stream, meaning this assumption probably led to some inaccuracies in the results. However, the difference in the densities of the oil and water is percentage-wise small, meaning the size of the inaccuracies are unlikely to be very significant.

5.3 Further Work

For the droplet distribution part, it would be beneficial to compare the results of the offline sensor, with the online sensors' results, when they are fully operational. This may provide insight on whether or not the droplet distribution is impacted by the sampling process, which is suspected to be the case. Different inlet flow-rates Q_{in}, could also be investigated, to observe the impact on the resulting distributions. Additionally, more work could be focused on determining the hydrocyclone's performance, with respect to the key criteria given in Equation (1) and (2).

The overflow estimation should ideally be improved to provide more stable results. If the valve equation is to be used as part of the estimation, more research should be focused on the valve constant. The overflow estimation should then be implemented in the LabVIEW program, to provide live estimated overflow values.

Additional work that may be done in the future could be centered around attempting to implement control structures, to improve the hydrocyclone's performance.

6 Conclusion

Investigating the droplet distribution at the inlet flow led to the conclusion that the droplets are close to log-normally distributed. Additionally, the distributions shifted when varying the opening of the inlet flow valve, Z_O. At smaller valve openings the distributions were smaller, as demonstrated in Figure 5. This is probably due to increased pressure, causing the larger droplets to break. Using the calculated variance and mean of the droplet distributions, an estimate of future distributions was generated, however this proved to be quite inaccurate.

Estimation of the overflow proved most successful when using the overall mass balance of the hydrocyclone. These estimated values proved to be relatively similar to the experimentally measured values, which Figure 14 illustrates.

The pressure drop over the underflow and overflow of the hydrocyclone was experimentally researched. The results, shown in Figure 15, proved to be similar to the simulated results. More specifically, the pressure drops of both the overflow and underflow increased as a function of the flowrate, with the increase over the overflow being larger.

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A List of Symbols

Symbol	**Units**	**Description**
A	m^2	Area
Cv_O	-	Valve constant overflow
c_v	ppm	Volume concentration
$c_{U,o}$	ppm	Concentration of oil in underflow
$c_{U,o}$	ppm	Concentration of oil in inlet flow
F_s	-	Flow split
Δh	m	Measured height difference
m	-	Calculated mean
P	bar	Pressure
Q_{in}	$\mathrm{m^3/h}$	Inflow-rate
Q_O	$\mathrm{m^3/h}$	Overflow-rate
Q_U	$\mathrm{m^3/h}$	Underflow-rate
r	m	Inner radius
t	s	Measurement time
v	-	Calculated variance
Z_{in}	-	Inlet flow valve opening
Z_O	-	Overflow valve opening
Z_U	-	Underflow valve opening
ρ	$\mathrm{kg/m^3}$	Density
η	-	Separation efficiency
μ	-	Mean
σ	-	Standard deviation

B Mastersizer Procedure

The following pages contain a more detailed procedure for using the Mastersizer [13].

NTNU Faculty of Natural Sciences and Technology	**PROCEDURE**	**Last revision :** **12.09.2018**
	Title: Mastersizer 3000	**Page 1 of 6**
Department: Chemical Engineering	**Procedure number: 2-0669-01-02**	
Revision number: 2	**Developed by:** **Anders Andersen, Marcin Dudek**	
	Approved by:	
	Maintained by:	

REVISION STATUS :

DOCUMENT STATUS : CONTROLLED

REV No. – DATE	REVISION made by	CHECKER	APPROVER
0 – 20.07.2016	Anders Andersen, Marcin Dudek		
1 – 25.11.2016	Marcin Dudek		
2 – 12.09.2018	Marcin Dudek		

This document is subjected to AUDIT.

ORIGINATOR: Anders Andersen, Marcin Dudek

REVISON MADE BY: Marcin Dudek

APPROVER: Camilla I. Dagsgård – Laboratory manager at UL

REVISION HISTORY	
DATE	**AMENDMENT DESCRIPTION**
25.11.2016	**Added instructions for Hydro SV accessory**
12.09.2018	**Added notes on obscuration**

1. PURPOSE
Mastersizer 3000 (Malvern Instruments) is used for quick and accurate particle size distribution analysis. It can work with both emulsions and dispersions.

2. SCOPE
Mastersizer 3000 uses laser diffraction for measuring the size of particles or droplets in a dispersion (from 0,01 to 3500 µm), by measuring the intensity of scattered light from those particles in a continuous phase. Data is then processed and presented as a size distribution.

3. RESPONSIBILITIES
The person responsible for the instrument is also responsible for updating this procedure.

4. DEFINITIONS AND ABBREVIATIONS
SOP – Standard operating procedure

5. EQUIPMENT
Mastersizer 3000
Hydro EV accessory
Beakers (400 – 1000 ml)
Hydro SV accessory
Washing station for Hydro SV

CHEMICALS
Water
Crude oil
Organic solvents (Toluene, Isopropanol)
Solid particles

6. METHOD

This procedure describes performing a manual measurement. It is possible to create an SOP (Standard operating procedure) in an analogous way, to quickly repeat similar measurements. The person responsible for the instrument can also create SOP on request, during training.

The instrument is equipped with two measurement accessories: Hydro EV (400-600 ml) and Hydro SV (6-7 ml). Depending on the amount of available sample, the user can choose whichever accessory fits best for his purposes.

Before measurements make sure that the sample and solvents used for cleaning after, are compatible with the tubing (can be changed, but normally is PVC) and sealing material (Viton®). Incompatible solvent may damage the instrument.

It is necessary to know the refractive index and the density of the dispersed material, as well as the refractive index of the dispersant before the measurement starts.

The instrument needs to be turned on 30 min before measurements to ensure thermal stability in the cell.

1. Turn on the computer and then the software on the desktop (Mastersizer 3000).
2. In the bottom-right corner check, if the instrument and accessory is connected properly (Mastersizer 3000 and Hydro EV)
3. Click **New** – **Measurement File**
4. Click **Manual Measurement** from the **Measurement** ribbon at the top
   a. Name the sample
   b. Select particle type:
      i. Emulsions: spherical
      ii. Dispersions: non-spherical (recommended, however not necessary)
   c. Select material by:
      i. Using the existing database (you can edit the database by adding known materials or chemicals)
      ii. Manual input of the refractive index and the density
   d. Select dispersant by:
      i. Database (same as in c.)
      ii. Manual input of the refractive index
   e. Measurement duration

   Not that important, anywhere between 10-20 s (usually 10s) depending on how much time you have

   f. Blue laser light measurement

   Blue laser is used for very fine particles. If you expect that your sample contains particles smaller than 150 nm, then check that option. If not, leave it unchecked, as it will prolong your measurement.

g. Sequence

Depends on how much time you want to spend on each sample. 3 runs per measurement is sufficient, more than 5 is unnecessary.

h. Obscuration

Heavily dependent on your sample and its concentration, may require some initial screening. See Additional Notes in the end of the procedure for more information. Standard values are:

- i. 10-20% for wet sample (emulsions, dispersions)
- ii. 1-10% for dry sample
- iii. 5-12% for crude oil emulsion (50-100 ppm concentration)

i. Accessories – stirrer
- i. Anywhere between 2000-3000 rpm
- ii. Note: if you have a full beaker and stir too fast, spillage may occur

j. Cleaning
- i. Normal – 3 sequences (for solid particles should be enough)
- ii. For crude oil emulsions (custom procedure)

k. Analysis Model
- i. General Purpose – most likely use this
- ii. Narrow – if you expect only a single peak (very monodisperse system)

l. Result type

It is recommended to use the volume distribution, however the choice is up to the user.

m. User sizes

It is recommended to use the default sizes.

Instructions for Hydro EV (points 5 to 10)
Instructions for Hydro SV (points 11 to 15)

5. Add beaker with dispersant (pure continuous phase) and lower the head. Make sure that the beaker is not full – 60-80% volume is usually enough for the measurement and to prevent spillage during mixing.
6. Click **Initialize the instrument** to initialize and **Start** again to measure the background. The background is of good quality, when the indicator of energy on the 1st and 20th detector is less than 100 and 20, respectively.
7. Add the sample into the beaker until you have reached sufficient obscuration (indicator on the left side of the screen), wait 30-50 seconds and then start measurements. After the measurement is done, you may preform additional measurements or skip to cleaning.
8. Start cleaning by clicking the **Clean** button (even though it may seem greyed out)
9. Follow the instructions on the screen. If you rinse the apparatus with organic solvents, make sure you use the portable fume hood and half-mask with appropriate filters. When the cleaning sequence is complete, stop the stirrer and exit the measurement window.

10. Remember to save your measurement file in your folder.

Cleaning:

- Dispersion with solid particles – rinsing the system several times with water is sufficient
- Crude oil emulsions – rinsing twice with isopropanol/toluene (50/50), then isopropanol once and then 4-5 times with tap water.

Make sure to clean any spillage to prevent damaging the equipment.
After measurements are complete, remember to clean the tray!

There is a possibility of connecting temperature control unit to the cell, to measure size distributions in various temperatures.

Instructions for Hydro SV

11. Carefully add the dispersant (pure continuous phase) to the cell by using a pipette or syringe. Avoid creating gas bubbles in the cell. Do not scratch the glass of the cell. Insert the accessory back into the instrument. You may add magnetic stirrer in the cell to have and control the mixing in the cell.
12. Click **Initialize the instrument** to initialize and **Start** again to measure the background. The background is of good quality, when the indicator of energy on the 1st and 20th detector is less than 100 and 20, respectively.
13. Take out the accessory from the instrument. Remove some dispersant and add your sample directly in the cell. This method may require some experience with the sample, as you need to be in a specific range of the obscuration (indicator on the left side of the screen). After adding the sample, put the accessory back into the instrument. If the appropriate level of obscuration is reached, wait 30-50 seconds and then start measurements. After the measurement is done, you may preform additional measurements or skip to cleaning.
14. Cleaning is performed with a washing station. Take out the accessory and unlock the cell. Put the cell in the right position in the washing station and flush the cell several times with an appropriate solvent:
    - Dispersion with solid particles – rinsing the system several times with water is sufficient
    - Crude oil emulsions – rinsing twice with toluene, then twice with isopropanol, then 4-5 times with tap water. Finish the cleaning with flushing the cell with isopropanol.
15. Put the cell on a fibreless cloth and let it dry.

The results can be accessed by opening the measurement file. The software produces graphs, different distributions and various parameters, however the raw data can still be exported to a text file or excel sheet.

It is possible to install the software on user's personal computer and access the files there. Ask the person responsible for the instrument for more details.

7. SAFETY EQUIPMENT

Safety goggles
Lab coat
Nitrile gloves
Half-mask
Portable fume hood

8. REFERENCES

Mastersizer 3000 manual
Malvern educational materials

ADDITIONAL NOTES

Obscuration tells you how much light is lost during your measurement. The software typically adjusts for 10-20% of obscuration, which means that the value above or below this range can yield inaccurate results. For example, if the concentration is too high, there is a risk of multiscattering (light is scattered off many particles). This will result in larger angle of scattering and a signal from very small particles (below 1 µm). Sometimes it can also be spotted by the presence of bimodal, non-continuous distribution of particles. Conversely, if the dispersion concentration is too low, the model in the software 'overadjusts' and can shift the distribution towards larger sizes of drops.

C Sampling Procedure

The following page contains the detailed procedure for sample extraction [12].

Chapter 4

Operation Procedures

4.1 Procedure to Operate Sampling Points

The following procedure must be followed when using the sampling points. Figure 4.1 shows a sampling point and the valve numbering which are used in the following procedure:

1. Check that the sampling bomb is drained and de-pressurized.
2. Open inlet valve (1) until the pressure in the sampling bomb matches the process pressure, then close inlet valve (1).
3. Open vent valve (2) until the pressure in the sampling bomb is at atmospheric level.
4. Extract sample from sample valve (3).
5. Fully drain the sampling bomb, then close sample valve (3) and vent valve [2].

D Matlab Code

D.1 Droplet Distribution

The following code has been used to plot the droplet distributions, calculate the average values, standard deviations and separable volumes, as well as the estimation of the distribution.

```
%% Plotting droplet distribution
clc
close all

%% Reading data from excel files
T = readtable('DropletDistribution1.xlsx'); % This contains 50% and 75% opening
T2 = readtable('DropletDistribution2.xlsx'); % This contains 40% opening
T3 = readtable('DropletDistribution3.xlsx'); % This contains 100% opening
T4 = readtable('30%.xlsx');
T5 = readtable('20%.xlsx');

%%
N = 30; % Total number of measurements first file
N2 = 15; % Total number of measurements second file
N3 = 15;
N4 = 15;
N5 = 15;

size = T{:,1};  % x-values are the same, gives droplet size
um10 = 55;  % Droplet size limits
```

```
um100 = 99;

relevantSize = T{um10:um100,1};  % relevant size area
    10-100
lowSize = T{1:um10,1};     % too small droplets

% Correction Factor 1.19948 to make it 100%
c = 1.19948;

%% Gathering y-values
samples = [];
for i = 0:N-1
    samplei = T{:,2+3*i}./c;     % These are the columns
        giving the volume percentage
    samples = [samples,samplei];
end

samples2 = [];
for i = 0:N2-1
    samplei2 = T2{:,2+3*i}./c;
    samples2 = [samples2,samplei2];
end

samples3 = [];
for i = 0:N3-1
    samplei3 = T3{:,2+3*i}./c;
    samples3 = [samples3,samplei3];
end

samples4 = [];
```

```
for i = 0:N4-1
    samplei4 = T4{:,2+3*i}./c;
    samples4 = [samples4,samplei4];
end

samples5 = [];
for i = 0:N5-1
    samplei5 = T5{:,2+3*i}./c;
    samples5 = [samples5,samplei5];
end

%% Plotting
%% 50%
figure
for i = 1:15
    plot(size, samples(:,i))
    hold on
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 7])
title('Droplet distribution 50% valve opening')
saveas(gcf,'DropletDistribution50','epsc')

figure
for i = 1:15
    semilogx(size, samples(:,i))
    hold on
```

```
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 7])
grid on
title('Droplet distribution 50% valve opening')
saveas(gcf,'DropletDistributionLog50','epsc')

%% 75%
figure
for i = 16:30
    plot(size, samples(:,i))
    hold on
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
title('Droplet distribution 75% valve opening')
saveas(gcf,'DropletDistribution75','epsc')

figure
for i = 16:30
    semilogx(size, samples(:,i),'HandleVisibility','off')
    hold on
```

```
    area(relevantSize, samples(um10:um100,i),'FaceColor', 'b')
    hold on
    area(lowSize, samples(1:um10,i), 'FaceColor', 'r')
    hold on
    Area75 = trapz(samples(um10:um100,i));
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
legend('Separable', 'Inseparable')
legend('Location','northwest')
title('Droplet distribution 75% valve opening')
saveas(gcf,'DropletDistributionLog75','epsc')

%% 40%
figure
for i = 1:15
    plot(size(1:99), samples2(:,i))
    hold on
    Area40 = trapz(samples2(um10:um100,i));
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
title('Droplet distribution 40% valve opening')
```

```
saveas(gcf,'DropletDistribution40','epsc')

figure
for i = 1:15
    semilogx(size(1:99), samples2(:,i),'HandleVisibility','off')
    hold on
    area(relevantSize, samples2(um10:um100,i),'FaceColor', 'b')
    hold on
    area(lowSize, samples2(1:um10,i), 'FaceColor', 'r')
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
legend('Separable', 'Inseparable')
legend('Location','northwest')
title('Droplet distribution 40% valve opening')
saveas(gcf,'DropletDistributionLog40','epsc')

%% 100%
figure
for i = 1:15
    plot(size(1:99), samples3(:,i))
    hold on
    Area100 = trapz(samples3(um10:um100,i));
end
```

```
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])

title('Droplet distribution 100% valve opening')
saveas(gcf,'DropletDistribution100','epsc')

figure
for i = 1:15
    semilogx(size(1:99), samples3(:,i),'
        HandleVisibility','off')
    hold on
    area(relevantSize, samples3(um10:um100,i),'
        FaceColor', 'b')
    hold on
    area(lowSize, samples3(1:um10,i), 'FaceColor', 'r')
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
legend('Separable', 'Inseparable')
legend('Location','northwest')
title('Droplet distribution 100% valve opening')
saveas(gcf,'DropletDistributionLog100','epsc')

%% 30%
figure
```

```
for i = 1:15
    plot(size(1:99), samples4(:,i))
    hold on
    Area30 = trapz(samples4(um10:um100,i));
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
title('Droplet distribution 30% valve opening')
saveas(gcf,'DropletDistribution30','epsc')

figure
for i = 1:15
    semilogx(size(1:99), samples4(:,i), 'HandleVisibility','off')
    hold on
    area(relevantSize, samples4(um10:um100,i),'FaceColor', 'b')
    hold on
    area(lowSize, samples4(1:um10,i), 'FaceColor', 'r')
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
legend('Separable', 'Inseparable')
legend('Location','northwest')
```

```
grid on
title('Droplet distribution 30% valve opening')
saveas(gcf,'DropletDistributionLog30','epsc')

%% 20%
figure
for i = 1:15
    plot(size(1:99), samples5(:,i))
    hold on
    Area20 = trapz(samples5(um10:um100,i));
end

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
title('Droplet distribution 20% valve opening')
saveas(gcf,'DropletDistribution20','epsc')

figure
for i = 1:15
    semilogx(size(1:99), samples5(:,i), '
       HandleVisibility','off')
    hold on
    area(relevantSize, samples5(um10:um100,i),'
       FaceColor', 'b')
    hold on
    area(lowSize, samples5(1:um10,i), 'FaceColor', 'r')
end
```

```
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0 150])
ylim([0 9])
legend('Separable', 'Inseparable')
legend('Location','northwest')
title('Droplet distribution 20% valve opening')
saveas(gcf,'DropletDistributionLog20','epsc')

%% Displaying percentage of area of curve which is in Separable
% AreaList = [Area20;Area30;Area40;Area50;Area75;Area100];
% disp(AreaList)

%% Average of the samples
meanSamples = [];
for i=1:length(samples)
    meanSamplei = mean(samples(i,1:15));
    meanSamples = [meanSamples;meanSamplei];
end

meanSamples1 = [];
for i=1:length(samples)
    meanSample1i = mean(samples(i,16:30));
    meanSamples1 = [meanSamples1;meanSample1i];
end

meanSamples2 = [];
for i=1:length(samples2)
```

```
    meanSample2i = mean(samples2(i,:));
    meanSamples2 = [meanSamples2;meanSample2i];
end

meanSamples3 = [];
for i=1:length(samples3)
    meanSample3i = mean(samples3(i,:));
    meanSamples3 = [meanSamples3;meanSample3i];
end

meanSamples4 = [];
for i=1:length(samples4)
    meanSample4i = mean(samples4(i,:));
    meanSamples4 = [meanSamples4;meanSample4i];
end

meanSamples5 = [];
for i=1:length(samples5)
    meanSample5i = mean(samples5(i,:));
    meanSamples5 = [meanSamples5;meanSample5i];
end

%% Plotting average curves
figure

% figure
semilogx(size,meanSamples1)
hold on
semilogx(size,meanSamples)
hold on
```

```
% semilogx(size(1:99),meanSamples2)
% hold on
% semilogx(size(1:99),meanSamples3)
% hold on
% semilogx(size(1:99),meanSamples4)
% hold on
semilogx(size(1:99),meanSamples5)
grid on
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
xlim([0.25 250])
ylim([0 7])
legend('Z_{in} = 75%', 'Z_{in} = 50%', 'Z_{in} = 20%')
legend('Location','northwest')
title('Droplet distribution comparison')
saveas(gcf,'DropletDistributionComparison20_50_75','
    epsc')
hold off

% figure
% plot(size(1:99),meanSamples4)
% hold on
% area(relevantSize, meanSamples4(um10:um100),'
    FaceColor', 'b')
% hold on
% area(lowSize, meanSamples4(1:um10), 'FaceColor', 'r')
% hold on
% xline(Av4,'--r');
% hold on
% xline(12.5,'--b');
```

```
%
% xlabel('Droplet size [\mum]')
% ylabel('Volume density [%]')
% xlim([0 150])
% ylim([0 9])
% title('Average Droplet Distribution 30%')
% grid on
% legend('V_{sep} = 54.15%', 'Separable', 'Inseparable
    ', 'Average', 'Median')
% legend('Location','northeast')
%
% figure
% semilogx(size(1:99),meanSamples4)
% hold on
% area(relevantSize, meanSamples4(um10:um100),'
    FaceColor', 'b')
% hold on
% area(lowSize, meanSamples4(1:um10), 'FaceColor', 'r')
% hold on
% xline(Av4,'--r');
% hold on
% xline(12.5,'--b');
% Qa = trapz(meanSamples4);
%
% xlabel('Droplet size [\mum]')
% ylabel('Volume density [%]')
% xlim([0 150])
% ylim([0 9])
% title('Average Droplet Distribution 30%')
% grid on
```

```
% legend('V_{sep} = 54.15%', 'Separable', 'Inseparable
    ', 'Average', 'Median')
% legend('Location','northwest')
%

%% Average value
A = size.*meanSamples;
Av = sum(A)/length(A);

A1 = size.*meanSamples1;
Av1 = sum(A1)/length(A1);

A2 = size(1:99).*meanSamples2;
Av2 = sum(A2)/length(A2);

A3 = size(1:99).*meanSamples3;
Av3 = sum(A3)/length(A3);

A4 = size(1:99).*meanSamples4;
Av4 = sum(A4)/length(A4);

A5 = size(1:99).*meanSamples5;
Av5 = sum(A5)/length(A5);

AvgList = [Av, Av1, Av2, Av3, Av4, Av5];

%% Standard deviation
var1 = [];
for i=1:length(meanSamples)
    v = (((size(i)-AvgList(1))^2)*meanSamples(i)/100);
```

```
    var1 = [var1;v];
end

var2 = [];
for i=1:length(meanSamples1)
    v = (((size(i)-AvgList(2))^2)*meanSamples1(i)/100);
    var2 = [var2;v];
end

var3 = [];
for i=1:length(meanSamples2)
    v = (((size(i)-AvgList(3))^2)*meanSamples2(i)/99);
    var3 = [var3;v];
end

var4 = [];
for i=1:length(meanSamples3)
    v = (((size(i)-AvgList(4))^2)*meanSamples3(i)/99);
    var4 = [var4;v];
end

var5 = [];
for i=1:length(meanSamples4)
    v = (((size(i)-AvgList(5))^2)*meanSamples4(i)/99);
    var5 = [var5;v];
end

var6 = [];
for i=1:length(meanSamples5)
    v = (((size(i)-AvgList(6))^2)*meanSamples5(i)/99);
```

```
    var6 = [var6;v];
end


varianceList = [sum(var1),sum(var2),sum(var3),sum(var4)
    ,sum(var5),sum(var6)];
stdList = [sqrt(varianceList(1)),sqrt(varianceList(2)),
    sqrt(varianceList(3)),sqrt(varianceList(4)),sqrt(
    varianceList(5)),sqrt(varianceList(6))];
%
% for i=1:length(meanSamples2)
%     if meanSamples2(i) > 0
%         meanSamplesFit = [meanSamplesFit;meanSamples2
    (i)];
%     end
% end

%% LogNormal Distribution Estimation
muList = [];
sigmaList = [];
for i=1:length(AvgList)
    mu = log(AvgList(i)^2/(sqrt(AvgList(i)^2+
        varianceList(i))));
    muList = [muList;mu];
    sigma = sqrt(log(varianceList(i)/AvgList(i)^2+1));
    sigmaList = [sigmaList;sigma];
end
```

```
%% 50%
pd1 = makedist('Lognormal','mu',muList(1),'sigma', sigmaList(1));
y1 = pdf(pd1,size);
A1 = (trapz(y1));

figure
semilogx(size, meanSamples)
hold on
semilogx(size, y1*(1/A1)*100) %% scaling: y1*(1/A)*100
xlim([0.1 150])
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
legend('True Distribution', 'Estimated Distribution')
legend('Location', 'Northwest')
title('Estimated vs True Distribution')
grid on
saveas(gcf,'EstimatedDistribution50','epsc')

%% 75%
pd2 = makedist('Lognormal','mu',muList(2),'sigma', sigmaList(2));
y2 = pdf(pd1,size);
A2 = (trapz(y2));

figure
semilogx(size, meanSamples1)
hold on
semilogx(size, y2*(1/A2)*100) %% scaling: y1*(1/A)*100
xlim([0.1 150])
```

```
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
legend('True Distribution', 'Estimated Distribution')
legend('Location', 'Northwest')
title('Estimated vs True Distribution')
grid on
saveas(gcf,'EstimatedDistribution75','epsc')

%% 30%
pd5 = makedist('Lognormal','mu',muList(5),'sigma',
    sigmaList(5));
y5 = pdf(pd5,size);
A5 = (trapz(y5));

figure
semilogx(size(1:99), meanSamples4)
hold on
semilogx(size, y5*(1/A5)*100) %% scaling: y1*(1/A)*100
xlim([0.1 150])
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
legend('True Distribution', 'Estimated Distribution')
legend('Location', 'Northwest')
title('Estimated vs True Distribution')
grid on
saveas(gcf,'EstimatedDistribution30','epsc')

%% 20%
pd6 = makedist('Lognormal','mu',muList(6),'sigma',
    sigmaList(6));
```

```
y6 = pdf(pd6, size);
A6 = (trapz(y6));

figure
semilogx(size(1:99), meanSamples5)
hold on
semilogx(size, y6*(1/A6)*100) %% scaling: y1*(1/A)*100
xlim([0.1 150])
xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
legend('True Distribution', 'Estimated Distribution')
legend('Location', 'Northwest')
title('Estimated vs True Distribution')
grid on
saveas(gcf,'EstimatedDistribution30','epsc')

%% Concentration
T = readtable('75%vs75%.xlsx');

N = 30; % Total number of measurements first file
c = 1.19948;

figure
semilogx(T{:,1}, T{:,2}./c)
hold on
semilogx(T{:,1}, T{:,5}./c)
hold off

xlabel('Droplet size [\mum]')
ylabel('Volume density [%]')
```

```
legend('c_v: 249 ppm','c_v: 93 ppm')
legend('Location','northwest')
xlim([0.1 200])
ylim([0 9])
grid on
title('Droplet distribution - different concentrations'
    )
saveas(gcf,'DropletDistribution75Concentration','epsc')

%% Number distribution
T = readtable('NumberDistribution.xlsx');
size = T{:,1};
N = 12;

%% Correction Factor 1.19948 to make it 100%
c = 1.19948;

%% Gathering y-values
samples = [];
for i = 0:N-1
    samplei = T{:,2+3*i}./c;     % These are the columns
        giving the volume percentage
    samples = [samples,samplei];
end

%% Plotting
figure
for i = 1:N
     plot(size(1:99), samples(:,i))
     hold on
```

```
end

xlabel('Droplet size [\mum]')
ylabel('Number density [%]')
xlim([0 10])
% ylim([0 9])
title('Droplet number distribution 75% valve opening')
saveas(gcf,'NumberDistribution75','epsc')
```

D.2 Distribution Trends

The following code has been used to create the plots given in Figure 6 and 8.

```
%% Distribution trends
clc
close all

Dx10 = [1.82,2.89,2.53,4.49,3.72];
Dx50 = [6.54,12.5,14.2,21.1,18.2];
Dx90 = [18.0,24.4,33.1,58.4,82.0];
Av = [10.5;11.97;16.32;32.00;28.45];
z = [0.20,0.30,0.50,0.75,1.00];

figure
scatter(z,Dx10,'filled');
hold on
scatter(z,Dx50,'filled');
hold on
scatter(z,Dx90,'filled');
hold on
```

```
scatter(z,Av,'filled');
hold off

% set(gca,'yscale','log')
xlabel('Z_O','Interpreter','tex')
ylabel('Size [\mum]','Interpreter','tex')
title('Distribution trends','Interpreter','tex')
legend('Dx10','Dx50','Dx90', 'Avg.','Interpreter','tex'
    )
legend('location','northwest')
saveas(gcf,'DistributionTrends','epsc')

figure
volSep = [23.0020,54.1463,64.4268,79.5904,72.2397];
scatter(z, volSep,'filled')
xlabel('Z_O','Interpreter','tex')
ylabel('Volume Separated [%]','Interpreter','tex')
ylim([0 100])
saveas(gcf,'VolumeSeparated','epsc')
```

D.3 Overflow Estimation

This code has been used for the results relating to the overflow estimation.

```
%% Overflow estimation
clc
close all

%% Parameters
r = 0.25;    % Inner radius
```

```
c = 0.1;      % Correction factor
t = 605;
A = pi*r^2;
Cvo = 0.05;      % Valve constant
rhoOil = 793;  %kg/m^3

%% Measurements
h0 = 0.159; h1 = 0.243; h2 = 0.418; h3 = 0.575; h4 =
    0.802; h5 = 1.103; h6 = 1.426; h7 = 1.741;
h8 = 2.096; h9 = 2.3190; h10 = 2.542; h11 = 2.702; h12
    = 2.951; h13 = 3.172;
h = [h0,h1,h2,h3,h4,h5,h6,h7,h8,h9,h10,h11,h12,h13];
dhList = [];

for i=1:length(h)-1
    dh = h(i+1) - h(i);
    dhList = [dhList;dh];
end

%% Overflow valve openings
z0 = 0; z1 = 0.05; z2 = 0.10; z3 = 0.15; z4 = 0.20; z5
    = 0.25; z6 = 0.30; z7 = 0.35;
z8 = 0.40; z9 = 0.50; z10 = 0.60; z11 = 0.70; z12 =
    0.80; z13 = 0.90;
zlist = [z1,z2,z3,z4,z5,z6,z7,z8,z9,z10,z11,z12,z13];

%% Measured times
t0 = 0; t1 = 604.91; t2 = 1271.1; t3 = 1793.8; t4 =
    2304.2; t5 = 2937.9;     t6 = 3561.2; t7 = 4146.4;
```

```
t8 = 4774.5; t9 = 5086.3; t10 = 5381.2; t11 = 5680.8;
    t12 = 5999.7; t13 = 6287.3;
t = [t0,t1,t2,t3,t4,t5,t6,t7,t8,t9,t10,t11,t12,t13];

dtList = [];

for i=1:length(t)-1
     dt = t(i+1) - t(i);
     dtList = [dtList;dt];
end

Qlist = [];

%% Experimentally measured overflow
for i=1:length(h)-1
     Q = A*(dhList(i))/dtList(i)*3600;
     Qlist = [Qlist;Q];
end

%% Reading data from excel file
T1 = readtable('TankLevel1.xlsx');

n1 = 4402;  n2 = 8501; n3 = 9952;   n4 = 13001; n5 =
    14402; n6 = 15950;
n7 = 4052; n8 = 8351; n9 = 9802; n10 = 12751;
n11 = 3052; n12 = 7452; n13 = 8852; n14 = 11001; n15 =
    12652; n16 = 14850;
n17 = 2702; n18 = 4901; n19 = 5852; n20 = 7400; n21 =
    8283; n22 = 9251; n23 = 10552;
n24 = 11751; n25 = 12502; n26 = 13451;
```

```
%% Mass balance (filtering the raw values in Matlab)
%% 5
f = 0.1;
Qin1 = lowpass(T1{n1:n2,2},f);
Qu1 = lowpass(T1{n1:n2,4},f);
Qo1 = Qin1-Qu1;

%% 10
Qin2 = lowpass(T1{n3:n4,2},f);
Qu2 = lowpass(T1{n3:n4,4},f);
Qo2 = Qin2-Qu2;

%% 15
Qin3 = lowpass(T1{n5:n6,2},f);
Qu3 = lowpass(T1{n5:n6,4},f);
Qo3 = Qin3-Qu3;

T2 = readtable('TankLevel2.xlsx');

%% 20
Qin4 = lowpass(T2{n7:n8,2},f);
Qu4 = lowpass(T2{n7:n8,4},f);
Qo4 = Qin4-Qu4;

%% 25
Qin5 = lowpass(T2{n9:n10,2},f);
Qu5 = lowpass(T2{n9:n10,4},f);
Qo5 = Qin5-Qu5;

```

```
T3 = readtable('TankLevel3.xlsx');

%% 30
Qin6 = lowpass(T3{n11:n12,4},f);
Qu6 = lowpass(T3{n11:n12,5},f);
Qo6 = Qin6-Qu6;

%% 35
Qin7 = lowpass(T3{n13:n14,4},f);
Qu7 = lowpass(T3{n13:n14,5},f);
Qo7 = Qin7-Qu7;

%% 40
Qin8 = lowpass(T3{n15:n16,4},f);
Qu8 = lowpass(T3{n15:n16,5},f);
Qo8 = Qin8-Qu8;

T4 = readtable('TankLevel4.xlsx');

%% 50
Qin9 = lowpass(T4{n17:n18,4},f);
Qu9 = lowpass(T4{n17:n18,5},f);
Qo9 = Qin9-Qu9;

%% 60
Qin10 = lowpass(T4{n19:n20,4},f);
Qu10 = lowpass(T4{n19:n20,5},f);
Qo10 = Qin10-Qu10;

%% 70
```

```
Qin11 = lowpass(T4{n21:n22,4},f);
Qu11 = lowpass(T4{n21:n22,5},f);
Qo11 = Qin11-Qu11;

%% 80
Qin12 = lowpass(T4{n23:n24,4},f);
Qu12 = lowpass(T4{n23:n24,5},f);
Qo12 = Qin12-Qu12;

%% 90
Qin13 = lowpass(T4{n25:n26,4},f);
Qu13 = lowpass(T4{n25:n26,5},f);
Qo13 = Qin13-Qu13;

%% Delta P for valve equation
dPo1 = lowpass(T1{n1:n2,3},f) - lowpass(T1{n1:n2,8},f);      % z = 0.05
dPo2 = lowpass(T1{n3:n4,3},f) - lowpass(T1{n3:n4,8},f);      % z = 0.10
dPo3 = lowpass(T1{n5:n6,3},f) - lowpass(T1{n5:n6,8},f);      % z = 0.15
dPo4 = lowpass(T2{n7:n8,3},f) - lowpass(T2{n7:n8,8},f);      % z = 0.20
dPo5 = lowpass(T2{n9:n10,3},f) - lowpass(T2{n9:n10,8},f);    % z = 0.25
dPo6 = lowpass(T3{n11:n12,6},f) - lowpass(T3{n11:n12,7},f);     % z = 0.30
dPo7 = lowpass(T3{n13:n14,6},f) - lowpass(T3{n13:n14,7},f);     % z = 0.35
```

```
dPo8 = lowpass(T3{n15:n16,6},f) - lowpass(T3{n15:n16,7},f);    % z = 0.40
dPo9 = lowpass(T4{n17:n18,6},f) - lowpass(T4{n17:n18,7},f);    % z = 0.50
dPo10 = lowpass(T4{n19:n20,6},f) - lowpass(T4{n19:n20,7},f);    % z = 0.60
dPo11 = lowpass(T4{n21:n22,6},f) - lowpass(T4{n21:n22,7},f);    % z = 0.70
dPo12 = lowpass(T4{n23:n24,6},f) - lowpass(T4{n23:n24,7},f);    % z = 0.80
dPo13 = lowpass(T4{n25:n26,6},f) - lowpass(T4{n25:n26,7},f);    % z = 0.90


%% Valve equation

Qov1 = [];
for i = 1:length(dPo1)
    Q = zlist(1)*Cvo*sqrt(2*dPo1(i)*10^5/rhoOil);
    Qov1 = [Qov1; Q];
end

Qov2 = [];
for i = 1:length(dPo2)
    Q = zlist(2)*Cvo*sqrt(2*dPo2(i)*10^5/rhoOil);
    Qov2 = [Qov2; Q];
end

Qov3 = [];
for i = 1:length(dPo3)
```

```
    Q = zlist(3)*Cvo*sqrt(2*dPo3(i)*10^5/rhoOil);
    Qov3 = [Qov3; Q];
end

Qov4 = [];
for i = 1:length(dPo4)
    Q = zlist(4)*Cvo*sqrt(2*dPo4(i)*10^5/rhoOil);
    Qov4 = [Qov4; Q];
end

Qov5 = [];
for i = 1:length(dPo5)
    Q = zlist(5)*Cvo*sqrt(2*dPo5(i)*10^5/rhoOil);
    Qov5 = [Qov5; Q];
end

Qov6 = [];
for i = 1:length(dPo6)
    Q = zlist(6)*Cvo*sqrt(2*dPo6(i)*10^5/rhoOil);
    Qov6 = [Qov6; Q];
end

Qov7 = [];
for i = 1:length(dPo7)
    Q = zlist(7)*Cvo*sqrt(2*dPo7(i)*10^5/rhoOil);
    Qov7 = [Qov7; Q];
end

Qov8 = [];
for i = 1:length(dPo8)
```

```
    Q = zlist(8)*Cvo*sqrt(2*dPo8(i)*10^5/rhoOil);
    Qov8 = [Qov8; Q];
end

Qov9 = [];
for i = 1:length(dPo9)
    Q = zlist(9)*Cvo*sqrt(2*dPo9(i)*10^5/rhoOil);
    Qov9 = [Qov9; Q];
end

Qov10 = [];
for i = 1:length(dPo10)
    Q = zlist(10)*Cvo*sqrt(2*dPo10(i)*10^5/rhoOil);
    Qov10 = [Qov10; Q];
end

Qov11 = [];
for i = 1:length(dPo11)
    Q = zlist(11)*Cvo*sqrt(2*dPo11(i)*10^5/rhoOil);
    Qov11 = [Qov11; Q];
end

Qov12 = [];
for i = 1:length(dPo12)
    Q = zlist(12)*Cvo*sqrt(2*dPo12(i)*10^5/rhoOil);
    Qov12 = [Qov12; Q];
end

Qov13 = [];
for i = 1:length(dPo13)
```

```
    Q = zlist(13)*Cvo*sqrt(2*dPo13(i)*10^5/rhoOil);
    Qov13 = [Qov13; Q];
end

%% Calculating average of estimated overflow and correcting for flowmeter error
QmeanList = [mean(Qo1)+c,mean(Qo2)+c,mean(Qo3)+c,mean(Qo4)+c,mean(Qo5)+c,mean(Qo6)+c,mean(Qo7)+c,mean(Qo8)+c,mean(Qo9)+c,mean(Qo10)+c,mean(Qo11)+c,mean(Qo12)+c,mean(Qo13)+c];

%% Calculating average of valve equation estimations
QovAvgList = [sum(Qov1)/length(Qov1);sum(Qov2)/length(Qov2);sum(Qov3)/length(Qov3);sum(Qov4)/length(Qov4);sum(Qov5)/length(Qov5);sum(Qov6)/length(Qov6);sum(Qov7)/length(Qov7);sum(Qov8)/length(Qov8);sum(Qov9)/length(Qov9);sum(Qov10)/length(Qov10);sum(Qov11)/length(Qov11);sum(Qov12)/length(Qov12);sum(Qov13)/length(Qov13)];

%% Plotting Mass balance estimated results (including correction)
figure
title('Mass balance estimation')
subplot(4,2,1)  %10
plot(Qo2+c)
hold on
yline(QmeanList(2), 'r')
hold off
xlim([0 1000])
```

```
title('10%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,2)  %20
plot(Qo4+c)
hold on
yline(QmeanList(4), 'r')
hold off
xlim([0 1000])
title('20%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,3)  %30
plot(Qo6+c)
hold on
yline(QmeanList(6), 'r')
hold off
xlim([0 1000])
title('30%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,4)  %40
plot(Qo8+c)
hold on
yline(QmeanList(8), 'r', 'Interpreter','tex')
hold off
xlim([0 1000])
title('40%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

```

```
subplot(4,2,5)   %50
plot(Qo9+c)
hold on
yline(QmeanList(9), 'r')
hold off
xlim([0 1000])
title('50%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,6)   %60
plot(Qo10+c)
hold on
yline(QmeanList(10), 'r')
hold off
xlim([0 1000])
title('60%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,7)   %70
plot(Qo11+c)
hold on
yline(QmeanList(11), 'r')
hold off
xlim([0 1000])
title('70%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,8)   %80
plot(Qo12+c)
hold on
```

```
yline(QmeanList(12), 'r')
hold off
xlim([0 1000])
title('80%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

saveas(gcf,'EstimatedOverflowMassbalance','epsc')

%% Plotting Valve equation estimated results

figure
subplot(4,2,1)  %10
plot(Qov2)
hold on
yline(QovAvgList(2), 'r')
hold off
xlim([0 1000])
title('10%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,2)
plot(Qov4)
hold on
yline(QovAvgList(4), 'r')
hold off
xlim([0 1000])
title('20%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,3)  %30
```

```
plot(Qov6)
hold on
yline(QovAvgList(6), 'r')
hold off
xlim([0 1000])
title('30%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,4)  %40
plot(Qov8)
hold on
yline(QovAvgList(8), 'r')
hold off
xlim([0 1000])
title('40%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,5)  %50
plot(Qov9)
hold on
yline(QovAvgList(9), 'r')
hold off
xlim([0 1000])
title('50%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,6)  %60
plot(Qov10)
hold on
yline(QovAvgList(10), 'r')
```

```
hold off
xlim([0 1000])
title('60%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,7) %70
plot(Qov11)
hold on
yline(QovAvgList(11), 'r')
hold off
xlim([0 1000])
title('70%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

subplot(4,2,8) %80
plot(Qov12)
hold on
yline(QovAvgList(12), 'r')
hold off
xlim([0 1000])
title('80%')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')

saveas(gcf,'EstimatedOverflowValveEquation','epsc')


%% Plotting Comparison of the methods' average values
figure
scatter(zlist,Qlist,'filled')
hold on
```

```
scatter(zlist,QmeanList)
hold on
scatter(zlist,QovAvgList, 'filled')
hold off

legend('Measured', 'Mass Balance', 'Valve Equation', 'Interpreter','tex')
legend('Location','northwest')
xlabel('Z_O', 'Interpreter','tex')
ylabel('Q_O [m^3/h]', 'Interpreter','tex')
saveas(gcf,'OverflowEstimationVsTankLevel','epsc')
```

D.4 Lognormal Distribution

This code has been used to create Figure 2.

```
%% LogNormal distribution figures
clc
close all

pd1 = makedist('Lognormal','mu',log(1),'sigma',0.5);
pd2 = makedist('Lognormal','mu',log(1),'sigma',1);
pd3 = makedist('Lognormal','mu',log(1),'sigma',0.75);

x = (0:0.01:10);
y1 = pdf(pd1,x);
y2 = pdf(pd2,x);
y3 = pdf(pd3,x);

figure

```

```
subplot(2,1,1)

plot(x,y1,'b')
hold on
plot(x,y2,'r')
hold on
plot(x,y3,'c')

title('Lognormal distribution (linear scale)')
legend('\sigma = 0.5', '\sigma = 1', '\sigma = 2')

subplot(2,1,2)

semilogx(x,y1,'b')
hold on
semilogx(x,y2,'r')
hold on
semilogx(x,y3,'c')

title('Lognormal distribution (logarhitmic scale)')
legend('\sigma = 0.5', '\sigma = 1', '\sigma = 2')

saveas(gcf,'LogNormalExample','epsc')
```

D.5 Pressure Flow Relationship

The following code has been used to plot simulated and experimental pressure-flow relationship. It is mostly created by Mishiga Vallabhan, but modified to include the experimental values.

```
clear all
par = initHC_Lab();
%xu=[0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8];
xu=0.6; %underflow valve opening
xo=[0.1];%0.4 %overflow valve opening
P2=300000; %initla guess of pressure
P3=400000; %initla guess of pressure
Ain=pi*par.Rin^2; %Inlet area
Au=pi*(0.005)^2;  %Underflow area
Ao=pi*(0.001)^2; %Oveflow area

%% Experimental values
T = readtable('PressureDrop.xlsx');
flow = T{:,2};
dPo = T{:,8};
dPu = T{:,9};

%% Her boudary condtion is inflow rate
Qin1=[0.0004 0.0005 0.0006 0.0008 0.0009 0.001 0.0015
    0.0016]; %Inflow rate
PQ=[]; % This matrix gives you Qo,Qu,P2, P3
Pin=[]
for i=1:length(Qin1)
     PQ=[PQ;Press_Flow_Relation(xu,xo,P2,P3,Qin1(i),par)
         ]
     Pin= [Pin;PQ(i,3)+(par.Rho_o/2)*(PQ(i,2)/Ao).^2+(
         par.Rho_o/4)*((0.175*Qin1(i)*par.R1).^2*(0.002)
         ^2)/(pi^2*par.Rin^4*(0.3718*0.005)^4)-(par.
         Rho_in/2)*(Qin1(i)/Ain).^2];
end
```

```
DPo=Pin-PQ(:,3);
DPu=Pin-PQ(:,4);

figure
plot(Qin1*3600,DPo*10^-5);
hold on;
plot(Qin1*3600,DPu*10^-5);
hold on
scatter(flow, dPo, 'filled')
hold on
scatter(flow, dPu, 'filled')
xlabel('$Q_{in}\ [m^3/h]$','FontSize', 12 );
ylabel('$P [bar]$','FontSize', 12 );
xlim([0 5]);
legend('$dP_o(Sim)$','$dP_u(Sim)$','$dP_o(Exp)$','$dP_u
    (Exp)$');
legend('Location', 'northwest');
saveas(gcf,'PressureFlowRelationship','epsc')
```