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Rain-out investigation: initial droplet size measurement

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Abstract

Droplet size distribution inside water flashing jets and corresponding rain-out fraction were measured. Mass distribution showed that a few droplets are “large” ($> 150 \mu\text{m}$) and count for more than 85 % of the liquid mass in the jet because of their large individual mass. This could be due to incomplete thermal fragmentation. It could explain the rain-out falling near the orifice or pipe exit.

Keywords: rain-out, loss of containment, flashing liquid jets, thermal fragmentation, droplet size distribution

1. Introduction

The rain-out problem is part of the loss of containment scenario about liquefied gases reservoirs: how much of the released liquid phase will fall on the ground? This fraction will not directly participate to the toxic or flammable cloud.

Experimental data about this topic are seldom (Johnson 1999, Hocquet 2002). Models are either not sufficiently validated because of this lack of experimental data or even not in good agreement with the existing ones (Wheatley 1987, Ianello 1989, Epstein 1990, Papadourakis 1991, UDM, Johnson 1999).

A former work (Hocquet 2002) showed that rain-out forms a few meters downstream the accidental breach *under* the horizontal aerosol. However literature predicts small droplets ($d_d < 150 \mu\text{m}$) will be formed through the thermal fragmentation mechanism (Brown 1962, Bushnell 1968, Gooderum 1969, Lienhard 1970, Oza 1983-1984, Razzaghi 1989, Reitz 1990, Park 1994, Ramsdale 1998, Witlox 2000-2001). Such small droplets would normally be

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entrained by the aerosol. The aim of this work is to understand how this non homogeneous behaviour can occur.

2. Experimental

We used two different experimental set-ups in order to measure drop sizes, temperature and rain-out fraction for known initial conditions T_{res} and P_{res} . Both set-ups used the same test sections. The tested fluid was water.

2.1. Test sections: orifices and pipes

We used both stainless steel orifices and pipes (fig. 1). The 2 mm inner diameter orifice (D2) and pipe (D2L100) were mainly used. But some experiments were conducted with D5 and D8 orifices, D5L250 and D8L400 pipes.

2.2 Rain-out, mass flow rate and jet temperature measurements

The experimental set-up for these measurements is mainly composed of an upstream tank, the test section, jet temperature probes and rain-out capture basins (fig. 2. & 3.). The upstream tank consists of a jacketed stainless steel column (4.5 m high, .257 m inner diameter, 0.233 m³ volume). It allows for a few minutes steady-state experiments up to $P_{res} = 1.1$ MPa and $T_{res} = 170^{\circ}\text{C}$. A $\pm 0.1^{\circ}\text{C}$ temperature uniformity for the tested fluid inside the tank was obtained. Pressure at the test section level is maintained constant within ± 2 kPa.

Jet temperature is measured both on its axis and just above the capture basins through 20 equally spaced (1 m) Pt temperature probes ($\pm 0.75\text{K}$ measurement uncertainty). Mass flow rate through the test section is measured by comparing the change of the pressure head above the test section during an experiment to the change when a known amount of liquid is added ($\pm 1\%$ accuracy). 15 capture basins (1 m * 2 m or 1 m * 1 m) allow rain-out water capture (fig. 2. & 3.). Water captured in each capture basin is weighted. Ratio of mass in one

capture basin to the total mass released from the upstream tank is a measure of the rain-out density function (± 2 % accuracy).

2.3 Droplet velocity and size measurements

The experimental set-up for these measurements is mainly composed of another upstream tank, the same test sections and a PDA system. The test section is connected to the upstream tank through a 3 m long, 0.013 m inner diameter, heat insulated flexible hose (fig. 4.).

This upstream tank ($\sim 1 \text{ m}^3$) allows the same initial thermodynamic conditions (1.1 MPa, 170°C) to be obtained as the other one. Internal electrical heating is provided, as well as N_2 inlet. Pressure and temperature change just upstream the test section during an experiment are typically of $\pm 10 \text{ kPa}$ and $\pm 0.5 \text{ }^\circ\text{C}$.

A DANTEC Dual-PDA¹ allows local measurements (typically 1 mm^3 volume) of both two components of individual droplet velocity and diameter.

The measurement volume can be moved in the three directions (2.7 m - .6 m - 1.5 m). Thus different measurement locations can be tested during the same experiment. We generally covered the vertical plane which includes the jet axis as described in fig. 5.

2.4. Droplet size measurement accuracy

The PDA stops when 3000 measures are collected or after 5 seconds if less than 3000 measures were collected. At 25 and 50 mm downstream of the D2L100 pipe, the apparatus only detects a few droplets (~ 200) during the 5 s which are allowed. The difficulty is probably linked to the optical density of the jet in this region: laser beams generally cannot cross the jet without being refracted. At intermediate distances (generally comprising 200 mm

¹ 2 300 mW argon lasers in two orthogonal planes, 60X Fiber Flow series, 57X80 and 58N81 detectors, BSA P70 electronic module, mask C.

downstream the orifice or pipe) 3000 droplets are measured. The number of measures decreases again farther than 400 or 600 mm. The main reason is probably the decrease in the droplets density. Sometimes droplets impact the lenses of the apparatus...

We thus decided to use a distance of 200 mm downstream of the orifice or pipe on the jet axis as a reference location. Another justification for this location is that the change in the size distribution is very important in the first few centimetres and clearly slower after 200 to 400 mm.

We estimate the Sauter mean diameter (d_{32}) from the sample collected by the PDA. This sample has to be large in order that the estimated value is close to the actual one. Most of the measured droplets are “small droplets” ($d_d < 150 \mu m$).

Figure 6. shows that estimated mean value for the small droplets is correct within a few micrometers when the sample has 3000 droplets. The “larger drops” play a predominant role when estimating d_{32} for the whole population because of their large V/S ratio. But these drops are relatively few among the whole population ($\sim 40 \text{ à } 50 \%$ at $T_{\text{res}} \sim 105^\circ\text{C}$, 3 to 10% at $T_{\text{res}} \sim 165^\circ\text{C}$). When the apparatus stops because he counted 3000 droplets, the sample of larger ones is still limited (100 and 300 droplets for the two tests depicted in figure 6). It is the reason why the convergence is slower. We can estimate the d_{32} uncertainty as $\pm 100 \mu m$ for a sample of 3000 droplets.

3. Results and discussion

Tested reservoir conditions are all sub-cooled. They are summarized in figures 7. (pipes) and 8. (orifices).

3.1. Flow type

Figure 9. shows mass velocity (W/A) data for *orifices* (A is the nominal section of the orifice) versus relative reservoir pressure. Lines show Bernoulli equation flow

$(G = C_D \sqrt{2\rho_l (P_{res} - P_a)})$ for different discharge coefficient values. There is a very good consistency if discharge coefficient is fitted ($C_D = 0.67$ for D2, $C_D = 0.70$ for D5). This shows that *observed flows are purely liquid*, as is expected for ideal orifices. Using “standard” discharge coefficient ($C_D = 0.61$) leads to 10 or 15% mass flow rate uncertainty.

Mass flow rates from experiments with *pipes* at temperatures lightly higher than the boiling one (up to 120 °C) also show a complete agreement ($\pm 5\%$) with purely liquid flow assumption (constant linear friction coefficient: $\lambda_c = 0.016$). Figure 10. represents mass velocities measured at higher temperatures ($T > 120^\circ\text{C}$). The agreement with the Bernoulli-like approach (or Lackmé approach: $G = \sqrt{2\rho_l (P_{res} - P_{sat}(T_{res}))}$) is quite good. It is better than using a model for initially saturated fluid (Fauske 1985 for example) even at low sub-cooling (10 to 20 kPa here). This means that the *flow is liquid in the main part of the pipe but there is some form of vaporisation just upstream its end*.

3.2. Rain-out measurements

Figure 11. shows typical spatial distributions for the rain-out downstream both orifice and pipes experiments. They are very similar one to the other (for similar reservoir conditions) as opposed to the completely different behaviour downstream a longer pipe (D8L4000; Hocquet 2002). This suggests a large influence of pressure gradient $(P_{res} - P_{sat}(T_{res}))/L$ on two-phase flow structure inside the pipe. The 2-phase flow region is much shorter for the shorter pipe, so that almost *no internal fragmentation* can occur (when $\Delta P/L > 1$ MPa/m instead of $\Delta P/L < .1$ MPa/m). This leads to the small pipe (D2L100) behaving like an orifice. *Our pipe measurements are representative of such highly sub-cooled flows only. All measurements we made were very similar downstream orifices and pipes. We thus will subsequently present orifice data only.*

Figure 12. shows the global rain-out we measured versus reservoir temperature T_{res} . Two important features of these data are that there is *no significant influence of the reservoir pressure on rain-out fraction* and that *data after a pipe are not so different from ones obtained after an orifice*. The agreement is also good with water data from the CCPS experiments (Johnson 1999).

3.3. Droplet diameter measurements; Number and mass distribution

Figures 13. shows that all distributions have a peak in the $[0, 150 \mu\text{m}]$ range, whatever T_{res} et P_{res} . Similar distributions are again obtained downstream a pipe. The shape of this peak is similar to what was reported in the literature (Brown 1962, Bushnell 1968, Gooderum 1969, Lienhard 1970, Oza 1983-1984, Razzaghi 1989, Reitz 1990, Park 1994, Ramsdale 1998, Witlox 2000-2001), qualitatively near a log-normal or a Rosin–Rammler distribution. Our measurements show that another population seems to exist. At low temperature ($T_{\text{res}} = 80^\circ\text{C}$, 104°C and 124°C), one observes a quasi-uniform distribution (almost the same number of droplets in each size class) up to $600 \mu\text{m}$ (PLDA limit for the used configuration). At higher temperatures (167°C) a few droplets with diameter larger than $150 \mu\text{m}$ still exist, most of them between 200 et $300 \mu\text{m}$.

Droplet mass is more important than droplet number as far as rain-out phenomenon is concerned. This led us to represent size distribution using the mass probability density function. Figures 14. shows a completely different trend. *The “small droplets” ($dd < 150 \mu\text{m}$), which represent the larger number ($\tau > 90 \%$ for $T_{\text{res}} \geq 136^\circ\text{C}$ and still $\tau > 45 \%$ for $T_{\text{res}} \geq 80^\circ\text{C}$) only represent 0,5 to 17 % of the mass! And the large ones count for the complement, i.e. 83 to 99,5 % of the mass!* The mass fraction for the « large droplets » in our temperature range is always the larger one. It represents almost all the mass when $T_{\text{res}} < 125^\circ\text{C}$. It then slowly decreases to approximately 85% at $T_{\text{res}} \sim 165^\circ\text{C}$ (fig. 15.). The

sensitivity of this fraction to pressure seems to be low, probably lower than the measurement uncertainty.

It seems to us that such a large fraction of large drops was never mentioned in the literature up to now especially in case of thermal fragmentation. This could be due to experimental considerations : in order to obtain best measurements where most of the droplets are present, it is often better to limit the measurement range. With our apparatus configuration the signal was very weaker in the $[0, 600 \mu\text{m}]$ (mask C) range than in the $[0, 200 \mu\text{m}]$ (mask A) range. Another reason is that most of the literature is devoted to very small orifices, generally less than $500 \mu\text{m}$ (Diesel injection for example). However preliminary experiments using shadow visualization of droplets under the aerosol confirm that they have diameters larger than $150 \mu\text{m}$.

3.4. Change with P_{res} et T_{res}

The distribution which appeared as quasi-uniform in number at the lower temperatures ($T_{res} < 125^\circ\text{C}$) appears increasing with diameter when represented in mass because mass increases as diameter to the cube. The mass representation shows that *the same distribution still exists at the higher temperatures for the larger droplets* (we define them from the histograms as : $d_d > 325 \mu\text{m}$). Further observing the mass histograms we are tempted to distinguish a third population: a peak between $150 \mu\text{m}$ and $325 \mu\text{m}$ is almost absent at the lower temperatures ($T_{res} < 105^\circ\text{C}$), but grows when temperature increases. When temperature is higher than $T_{res} = 136^\circ\text{C}$ its mass is more than that of « small droplets ». These comments apply to both D2 orifice and D2L100 pipe.

Histogram at 80°C (fig. 13.) can only be understood as resulting from pure mechanical fragmentation. It consists of a quasi-uniform distribution between 0 and $600 \mu\text{m}$, plus a peak of very fine droplets (around $d_d = 15 \mu\text{m}$). The histograms at 104°C or 124°C are qualitatively

similar. The only difference is that the peak moved to lightly larger diameters. Fragmentation at such T_{res} probably still corresponds to essentially mechanical fragmentation.

Thermal fragmentation is thus not the only one which can form very fine droplets ($d_d < 50 \mu\text{m}$). Thus we cannot decide if the peak of “small droplets” ($d_d < 150 \mu\text{m}$, more than 90 % of the droplets at high temperature) has a thermal or mechanical origin. There is perhaps superimposition of a mechanical peak and a thermal one at lightly larger diameters. The growth of the thermal peak could eventually explain that the global peak moves towards slightly larger diameters when temperature increases.

The “medium” peak ($150 \mu\text{m} < d_d < 325 \mu\text{m}$) is almost absent at the lower temperatures (fig. 15.) ($T_{\text{res}} < 125^\circ\text{C}$). It is visible at $T_{\text{res}} \sim 165^\circ\text{C}$. This leads us to associate it with thermal fragmentation. The “larger drops” ($d_d > 325 \mu\text{m}$) would be a remaining part of the primary fragmentation which would not have undergone secondary one.

This would mean that the thermal fragmentation would only concern a fraction of the liquid, with the “larger drops” staying in a meta-stable state ! Fig 16. shows that the mass fraction for these droplets is very high, even if it decreases when temperature increases: 90 % at $T_{\text{res}} \sim 105^\circ\text{C}$, still $\sim 60 \%$ at $T_{\text{res}} \sim 165^\circ\text{C}$. Perhaps the number of nuclei is not large enough for thermal fragmentation to be complete. This type of fragmentation would be more intensive at higher temperatures because more nuclei would be activated

3.5. Axial change of mean size

Fig. 16. represents the probability density function data at different locations downstream the orifice or pipe. Fig. 17. shows the corresponding mass distributions.

The number fraction of « small droplets » ($d_d < 150 \mu\text{m}$) represents between 80 and 90 % at 30 mm. The maximum is reached between 200 and 400 mm downstream the orifice or pipe. Meanwhile a peak between $150 \mu\text{m}$ and $325 \mu\text{m}$ appears on the mass histograms.

This means that there exists at least one *external mechanism* which breaks the droplets mainly in the first few centimetres, and continues during 200 to 400 mm.

The Sauter mean diameter decreases when the number of « small droplets » increases (from pipe exit to ~ 200-400 mm downstream; fig. 18.). It can then increase when their relative weight decreases (farther than 200 to 400 mm). Their relative weight decreases slower when temperature is lower, which could explain that the trend for d_{32} to increase comes farther. Note that the observed diameter increase is generally smaller than the uncertainties but the trend is observed for all the experiments.

4. Conclusion

Two pilot scale experimental set-up allowed us to characterize flashing water jets. We measured mass flow rate, initial droplet size and velocity distributions, plus spatial distribution of rain-out. Test sections were both orifices and pipes, mainly 2 mm inner diameter.

Mass flow rate measurements and spatial distribution of rain-out showed that almost no internal fragmentation in the pipe occurs when reservoir sub-cooling is large enough ($\Delta P / L > 1 \text{ MPa} / \text{m}$). In such conditions both pipes and orifices exhibit the same jet behaviour (rain-out fraction and rain-out spatial distribution).

Reservoir pressure (P_{res}) has not a significant influence on total rain-out fraction as compared to reservoir temperature T_{res} .

All droplet size distributions exhibit a peak in the $[0 \div 150 \mu\text{m}]$ range as described in the literature. But some larger droplets are also detected. These last ones count for more than 85 % of the liquid mass even when they count for less than 10 % of the droplets number!

The "small droplets" ($d_d < 150 \mu\text{m}$) form mainly in the first few centimetres with the maximum number reached between 200 and 400 mm downstream the orifice or pipe. Mass

histograms show that another peak forms between 150 and 325 μm . These two peaks mean that an *external secondary fragmentation mechanism* exists.

The former observations lead to the following scheme:

- A part of the liquid phase undergoes very rapidly (mostly less than 10 mm) a mechanical fragmentation and probably a thermal fragmentation. This leads to a population of “small droplets” ($d_d < 150 \mu\text{m}$). This mechanism is stronger at the higher temperatures.
- When temperature increases another fraction of the droplets is concerned with thermal fragmentation. This leads to the “medium droplets” ($150 \mu\text{m} < d_d < 325 \mu\text{m}$).
- An important residual quantity of droplets do not undergo secondary fragmentation. They constitute the “large droplets” population ($325 \mu\text{m} < d_d < 600 \mu\text{m}$).

The large mass fraction of large droplets ($d_d > 150 \mu\text{m}$) could be an explanation for our former observation that rain-out is heterogeneous in nature (Hocquet 2002) : it is composed of droplets which fall *under* the almost horizontal two-phase jet. Our next aim will be to prove that such size distributions can be a basis in order to improve rain-out prediction models. Future work will also have to take into account near saturated reservoir conditions and fluids other than water.

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Fig. 1.: scheme of the test sections (orifices and pipes)

Fig 2. : scheme of the set-up for rain-out measurements

Fig 3. : photograph of the jet with the capture basins .

Fig. 4.: scheme of the set-up for droplet size measurement

Fig. 5 : typical PDA measurement locations

Fig 6.: d_{32} measurement accuracy versus droplet number (small droplets ; whole population).

$$T_{\text{res}} = 164^{\circ}\text{C}, P_{\text{res}} = 0.82 \text{ Mpa}$$

fig. 7.: Experimental reservoir conditions. Water jets from orifices.

fig. 8.: Experimental reservoir conditions. Water jets from pipes.

Figure 9. : Mass velocity versus relative reservoir pressure (orifices)

Fig 10. : mass velocity versus $(P_{\text{res}} - P_{\text{sat}}(T_{\text{res}}))$ (pipes)

Fig 11. : Typical spatial distribution of rain-out ($T_{\text{res}} = 164^{\circ}\text{C} / P_{\text{res}} = 0.82 \text{ MPa}$).

Fig. 12. : rain-out fraction versus reservoir temperature.

Fig. 13. : Number size distribution at different reservoir conditions
(200 mm down-stream from D2 orifice)

Fig. 14. : mass size distribution at different reservoir conditions

(200 mm down-stream from D2 orifice)

Fig. 15. : mass % of large droplets versus reservoir temperature

Fig. 16: number size distribution at different locations

(D2 orifice, $T_{\text{res}} = 164^{\circ}\text{C}$; $P_{\text{res}} = 0.82 \text{ MPa}$)

Fig. 17: mass size distribution at different locations

(D2 orifice, $T_{\text{res}} = 164^{\circ}\text{C}$; $P_{\text{res}} = 0.82 \text{ MPa}$)

Fig. 18. : Change of SAUTER mean diameter with distance from the orifice

D2 orifice, $T_{\text{res}} = 167^{\circ}\text{C}$, $P_{\text{res}} = 1.04 \text{ MPa}$



Fig. 1.: scheme of the test sections (orifices and pipes)

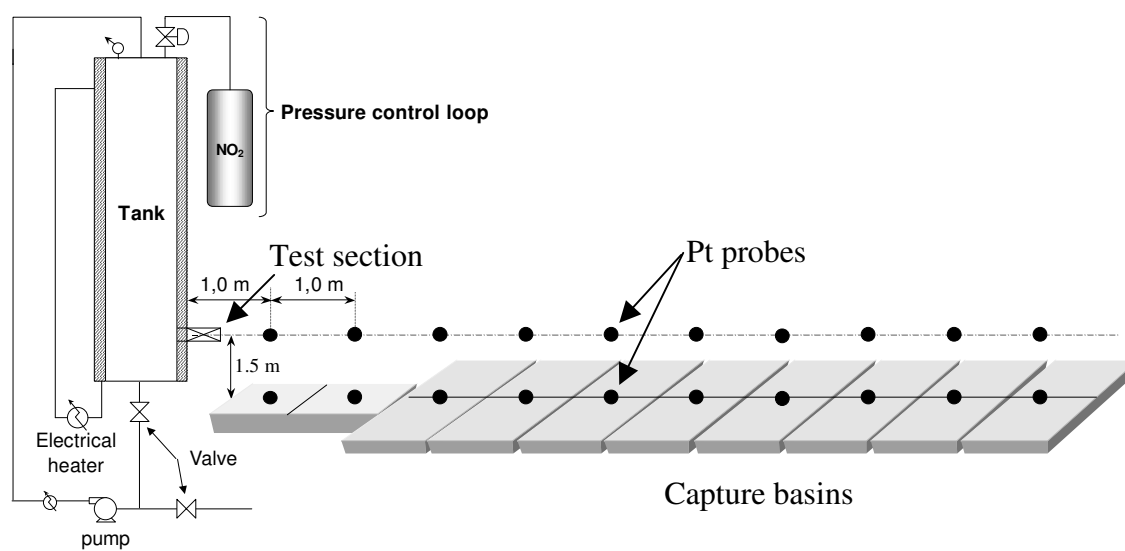


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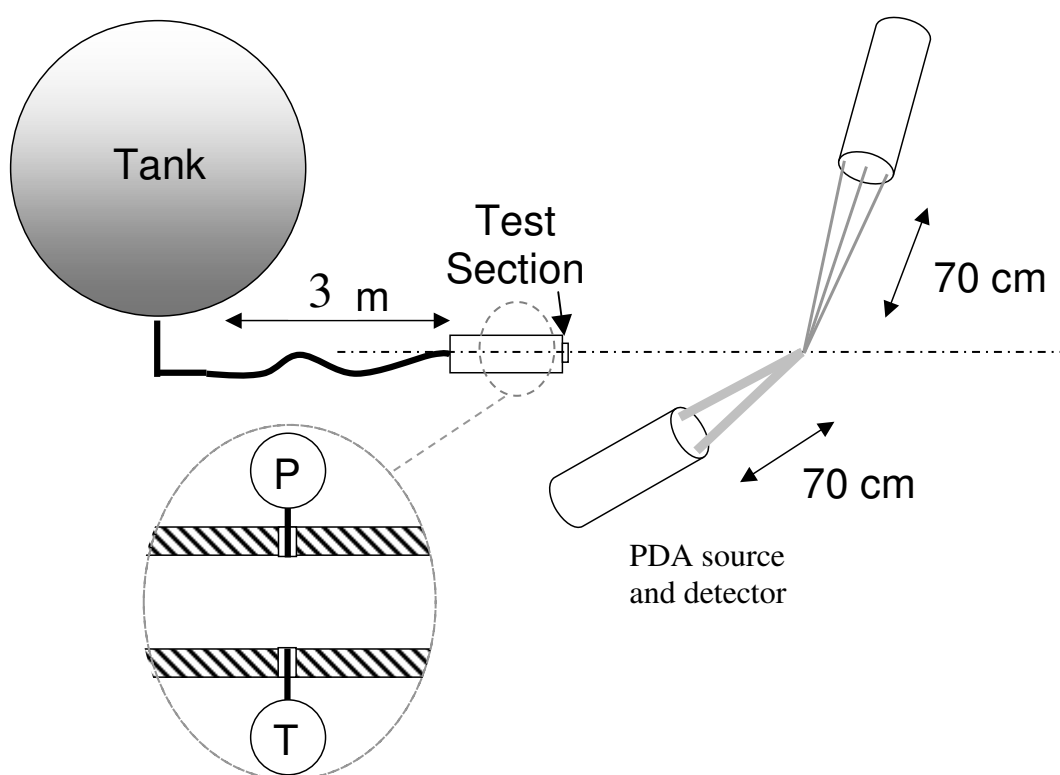


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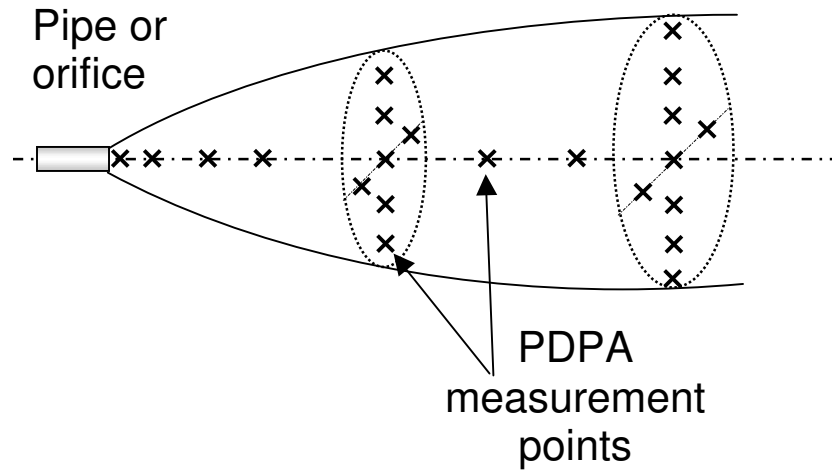


Fig. 5 : typical PDA measurement locations

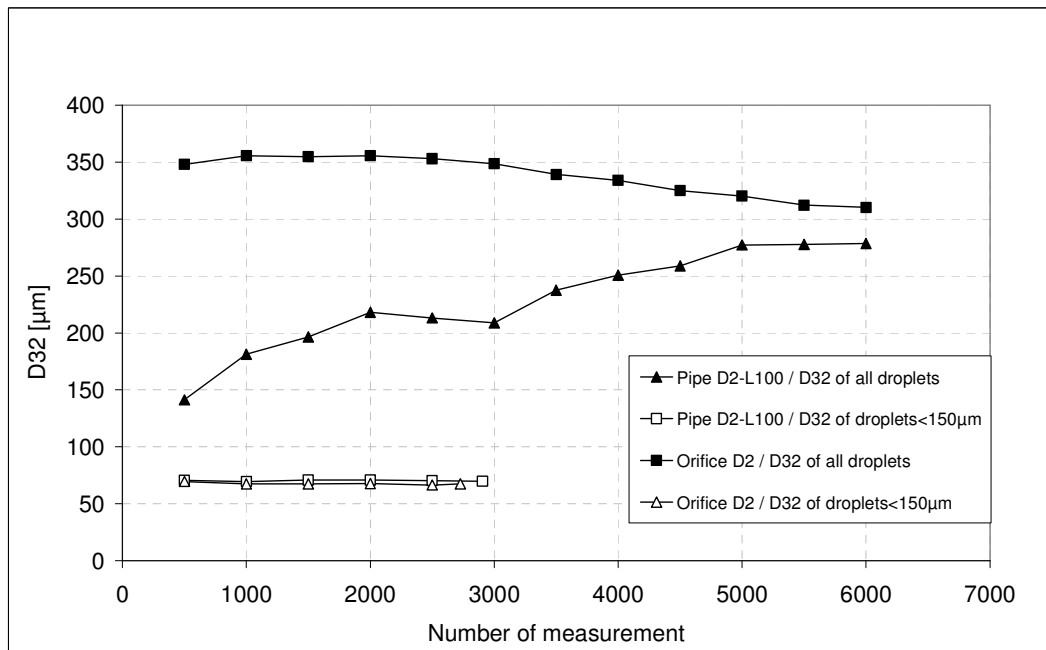


Fig 6.: d_{32} measurement accuracy versus droplet number (small droplets ; whole population).

$$T_{\text{res}} = 164^{\circ}\text{C}, P_{\text{res}} = 0.82 \text{ MPa}$$

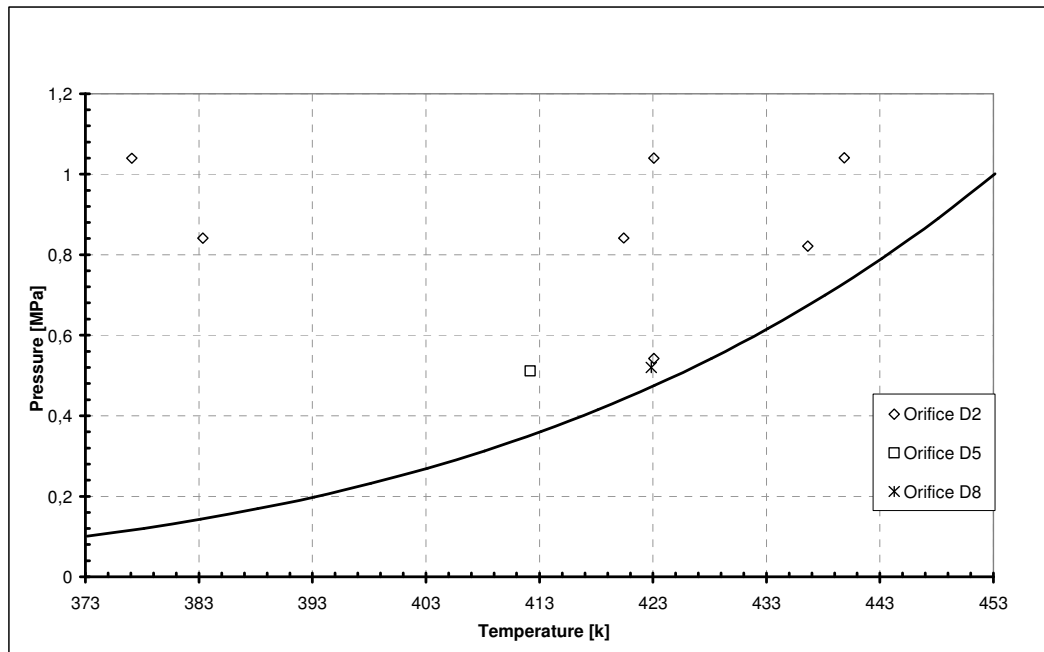


fig. 7.: Experimental reservoir conditions. Water jets from orifices.

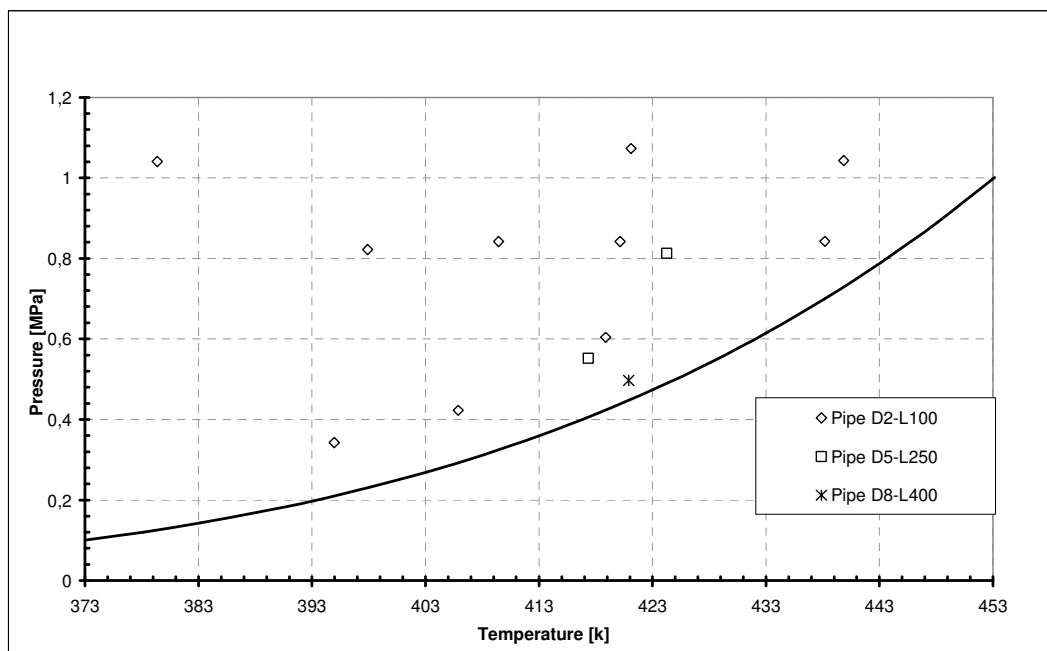


fig. 8.: Experimental reservoir conditions. Water jets from pipes.

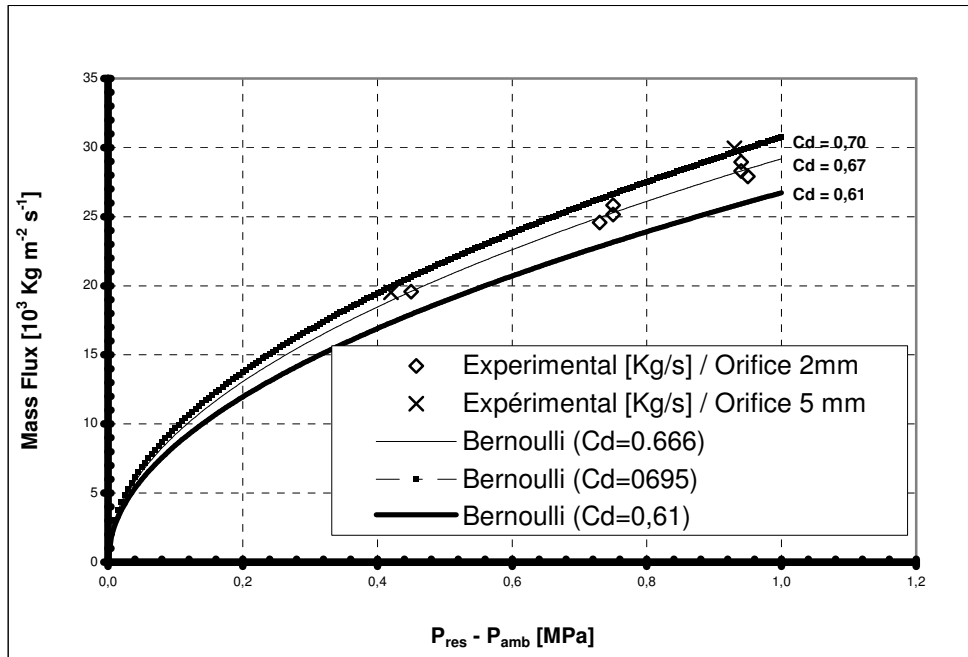


Figure 9. : Mass velocity versus relative reservoir pressure (orifices)

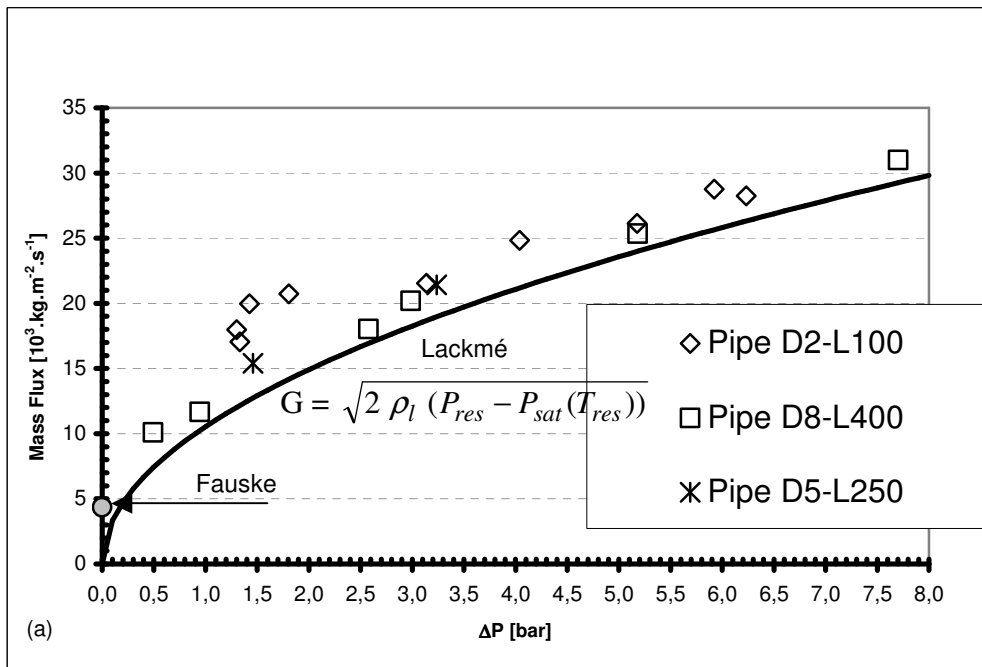


Fig 10. : mass velocity versus $(P_{\text{res}} - P_{\text{sat}}(T_{\text{res}}))$ (pipes)

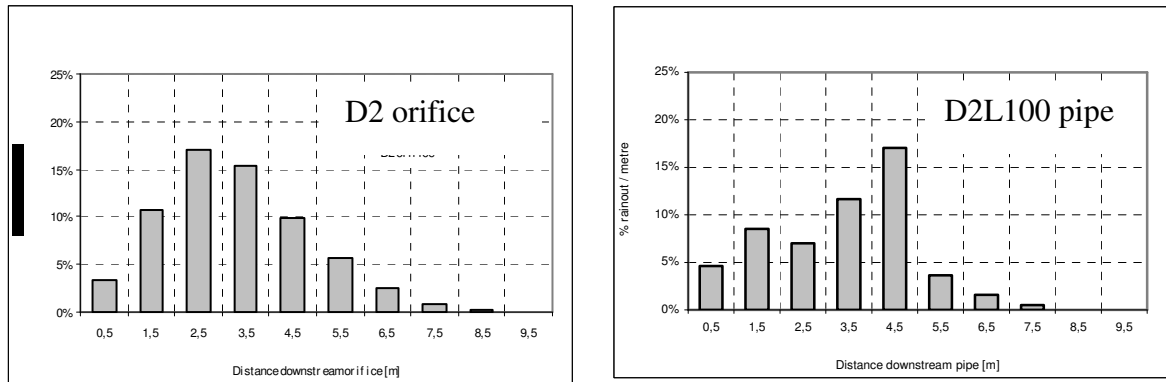


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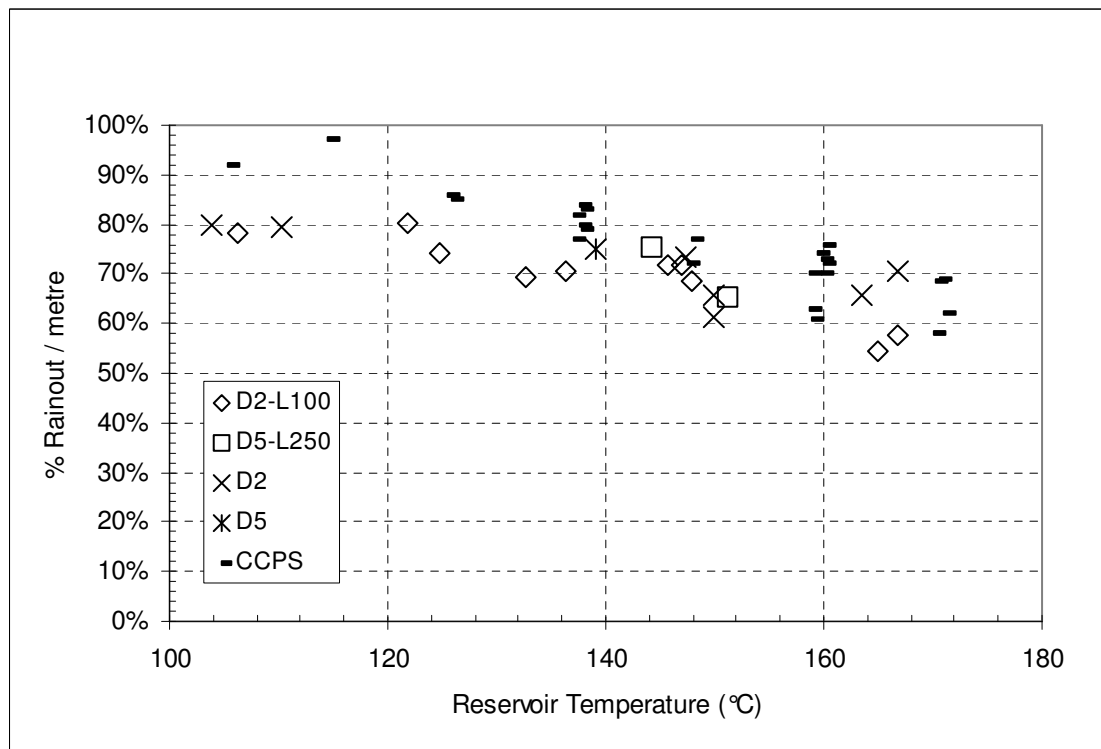
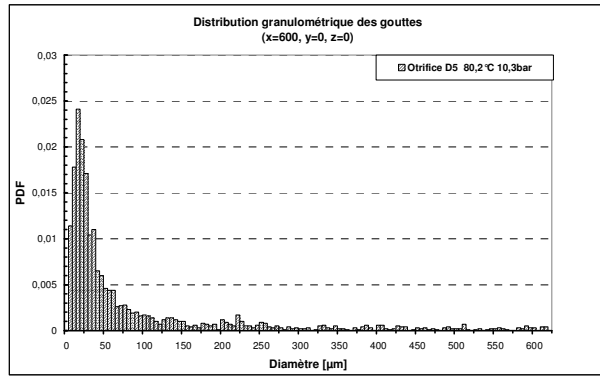
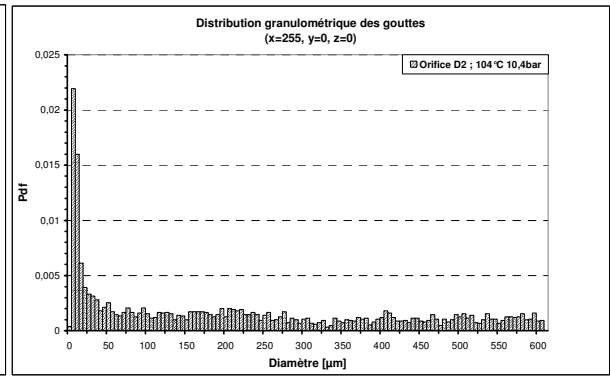


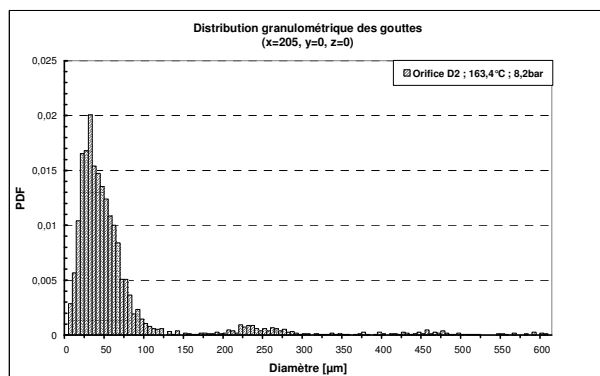
Fig. 12. : rain-out fraction versus reservoir temperature.



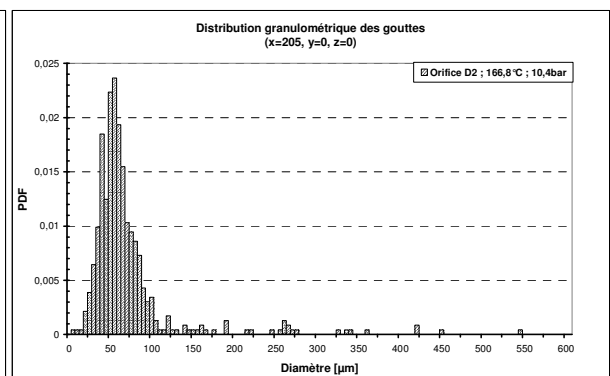
$$T_{\text{res}} = 80.2 \text{ °C} / P_{\text{res}} = 1.03 \text{ MPa}$$



$$T_{\text{res}} = 104 \text{ °C} / P_{\text{res}} = 1.04 \text{ MPa}$$



$$T_{\text{res}} = 163.4 \text{ °C} / P_{\text{res}} = 0.82 \text{ MPa}$$



$$T_{\text{res}} = 166.8 \text{ °C} / P_{\text{res}} = 1.04 \text{ MPa}$$

Fig. 13. : Number size distribution at different reservoir conditions
(200 mm down-stream from D2 orifice)

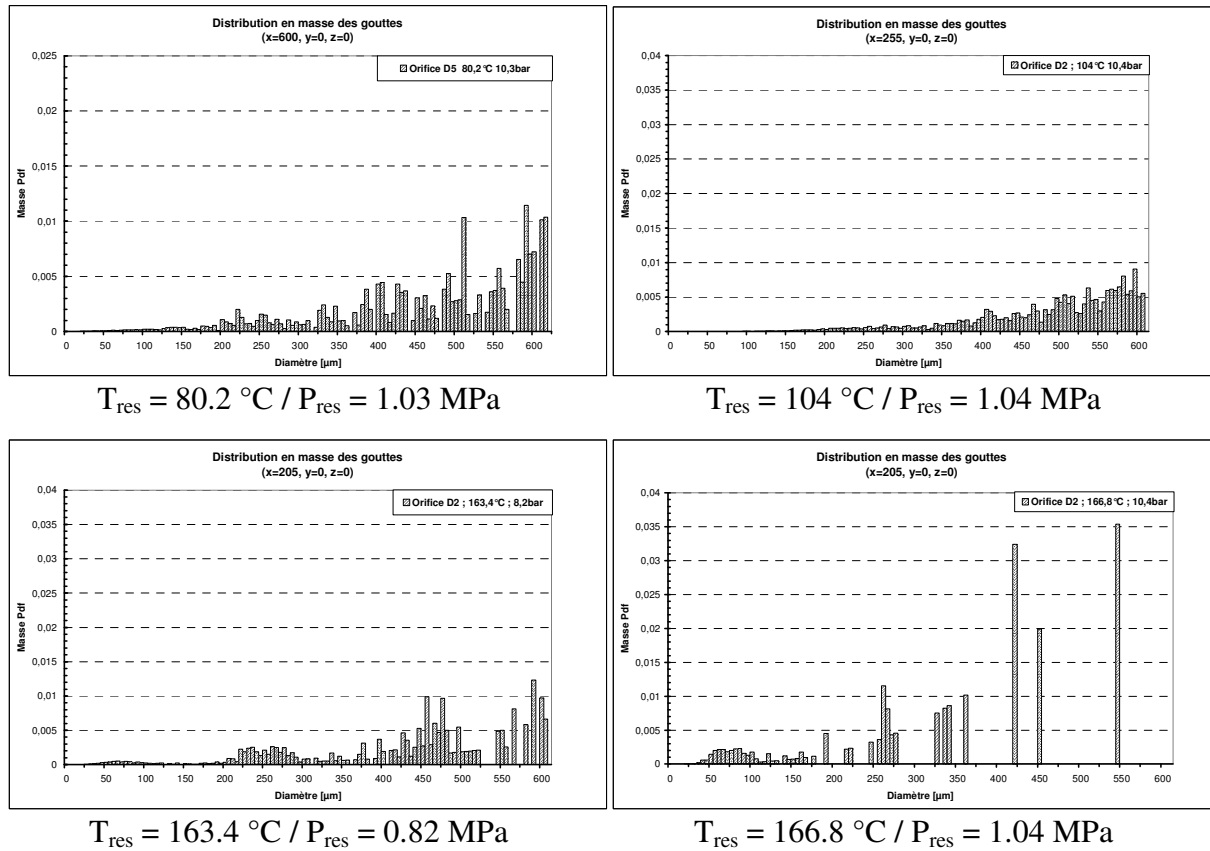


Fig. 14. : mass size distribution at different reservoir conditions
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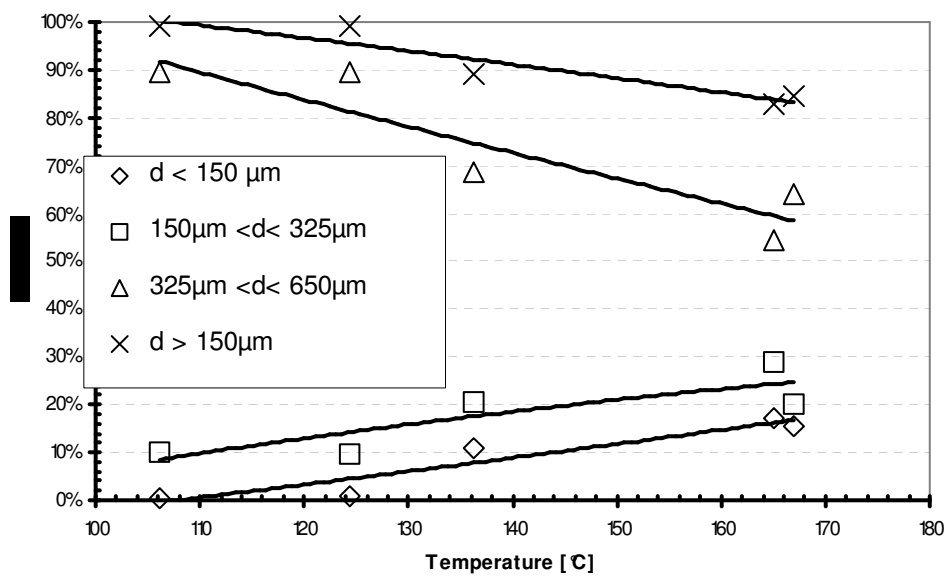


Fig. 15. : mass % of large droplets versus reservoir temperature

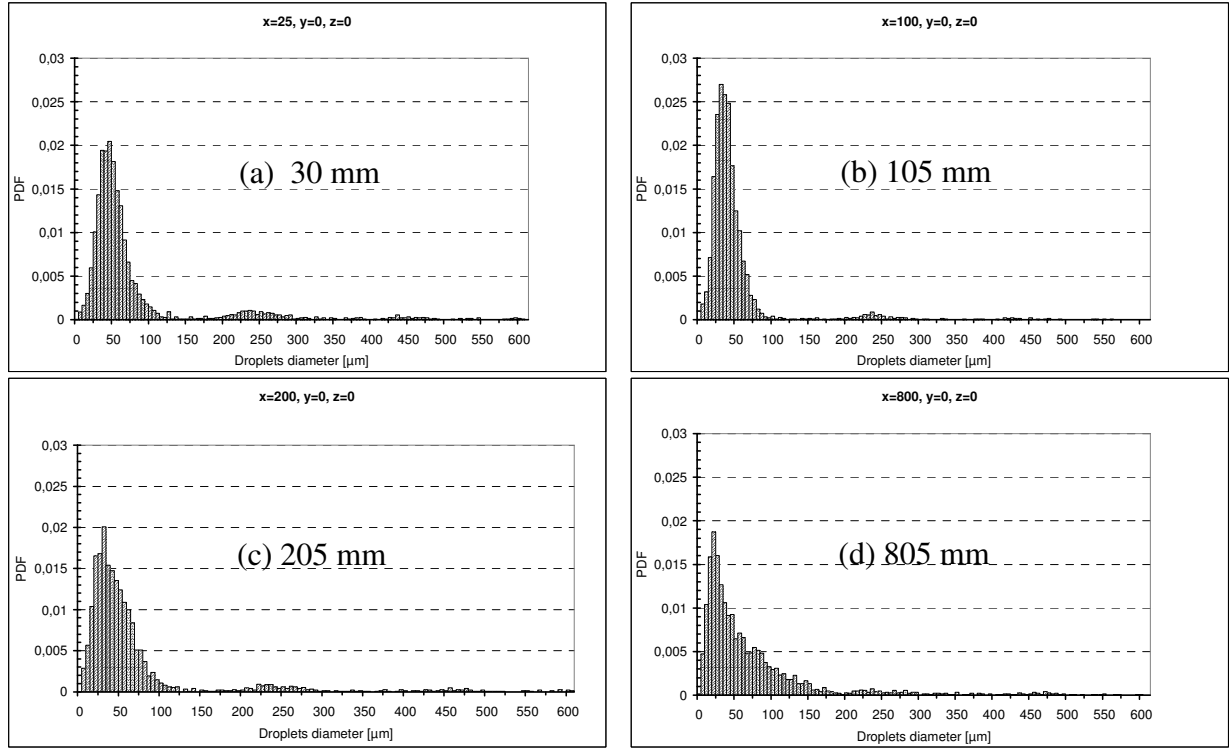


Fig. 16: number size distribution at different locations

(D2 orifice, $T_{\text{res}} = 164^{\circ}\text{C}$; $P_{\text{res}} = 0.82 \text{ MPa}$)

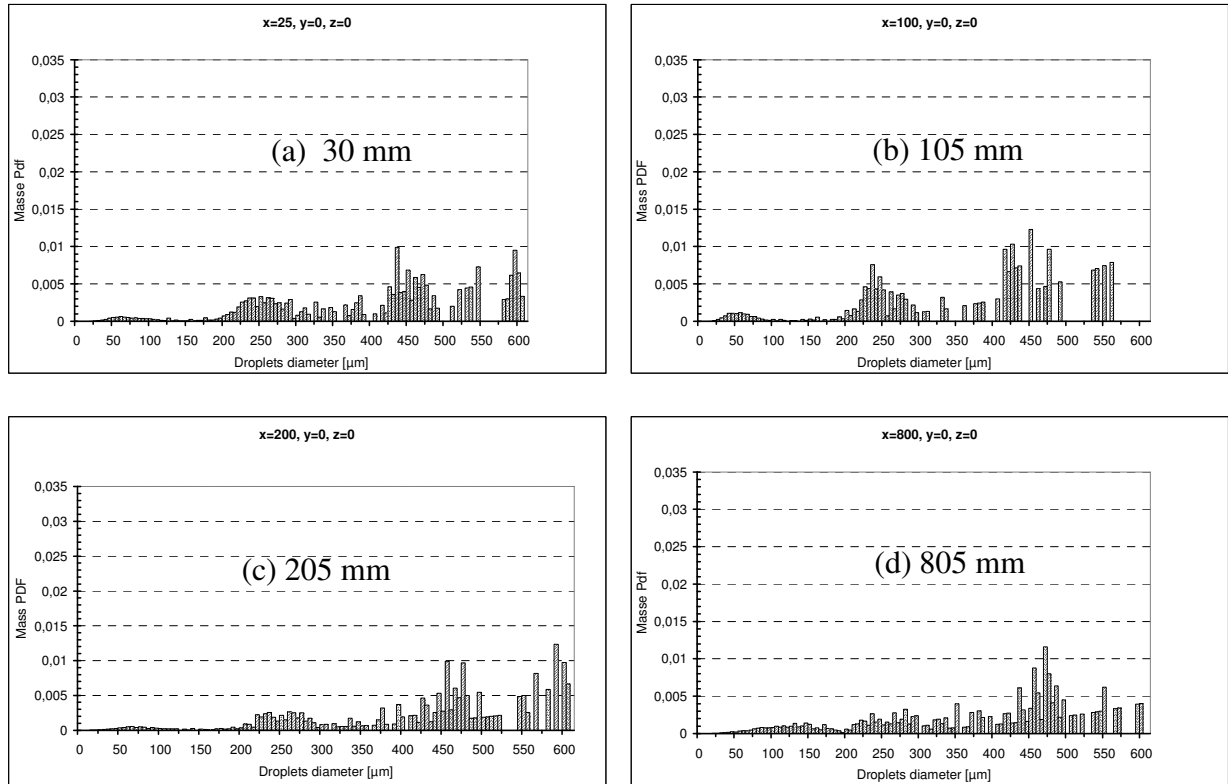


Fig. 17: mass size distribution at different locations

(D2 orifice, $T_{\text{res}} = 164 \text{ }^{\circ}\text{C}$; $P_{\text{res}} = 0.82 \text{ MPa}$)

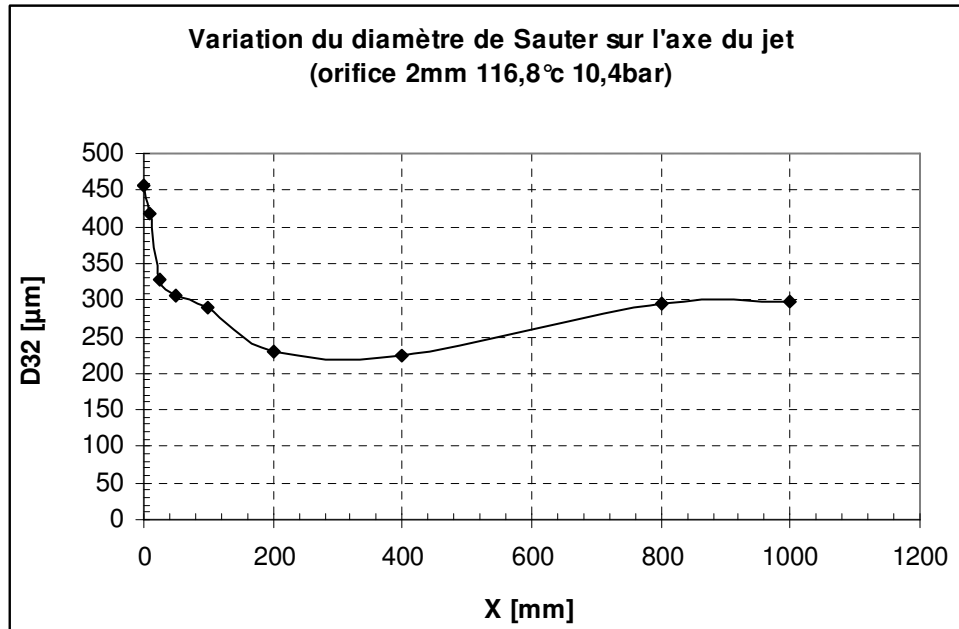


Fig. 18. : Change of SAUTER mean diameter with distance from the orifice

D2 orifice, $T_{\text{res}} = 167^{\circ}\text{C}$, $P_{\text{res}} = 1.04 \text{ MPa}$