

Application of fuel tracers with different volatilities for planar LIF/Mie drop sizing in evaporating systems

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Dropsizing imaging on the base of the ratio of laser induced fluorescence (LIF) and Mie scattering is frequently used for planar non-intrusive measurement of the Sauter mean diameter (SMD) in sprays. In evaporating sprays, the variation of tracer concentration in the liquid phase influences the measurement. We investigate the systematic effects that occur applying this method in evaporating sprays with tracers of different volatilities. The investigations were carried out in a ethanol-fueled spray flame in a coflow of air. For different fuel tracers the resulting two-dimensional distributions of apparent SMD are presented and compared to a simple evaporation model.

1. Introduction

The atomization of liquid fuel plays an important role in many technical processes, especially in combustion processes. It can be found in Diesel, gasoline direct injection and aircraft engines as well as in domestic heating systems. To improve the energy efficiency and pollutant formation knowledge of the atomization, evaporation and mixture generation is vital. For the measurement of the droplet sizes of a spray various techniques exist. Most provide information only at single points. For instantaneous imaging of droplet Sauter mean diameter (SMD) distributions, a non-intrusive, laser-based technique was suggested by Yeh et al. [1] and since then applied to various spray configurations [2]. The SMD is defined by the average droplet volume divided by the average droplet surface [3] and given by the following expression:

$$SMD = \frac{1}{N} \sum_{i=1}^N D_i^3 \bigg/ \frac{1}{N} \sum_{i=1}^N D_i^2 \quad (1)$$

N represents the number of droplets and D_i the diameter of the respective droplets. The average droplet surface is determined by the detection of Mie scattering which, at spherical droplets with a diameter significantly larger than the wavelength of the scattered light, is $S_{Mie} = C_{Mie} D^2$. While strong signal variations depending on minor diameter variations and detection angles occur, it is generally accepted that the equation holds for an ensemble of droplets with a spectrum of droplet sizes [2,4]. C_{Mie} is the fitting constant. The averaged droplet volume is measured by laser-induced fluorescence (LIF) emitted from a tracer added to the (otherwise non-fluorescing) fuel. If the absorption of the incident laser beam and the self-absorption of the LIF signal in the droplet are negligible and self-quenching and lasing does not occur, the LIF signal I_{LIF} is connected via C_{LIF} with the droplet volume: $S_{LIF} = C_{LIF} D^3$. Hence, the ratio of average LIF and Mie signals is proportional to the local SMD [1].

$$\frac{S_{LIF}}{S_{Mie}} = \frac{C_{LIF}}{C_{Mie}} \sum_{i=1}^N D_i^3 \bigg/ \sum_{i=1}^N D_i^2 \propto SMD \quad (2)$$

When the probe volume is illuminated and the signals are detected with CCD cameras this method yields a 2d spatially resolved information. Care must be taken that the number of droplets N is identical for both measurements, which is typically ensured by measuring both quantities simultaneously. Calibration is possible by comparing the signal ratio with one measured in a defined monodisperse spray.

While this technique shows good results in non-evaporating sprays [2], care must be taken in evaporating systems due to local and temporal variation of tracer concentrations, potential tracer fluorescence from the gas phase and temperature-dependent fluorescence cross sections. The goal of this study is the application of Mie/LIF imaging to evaporating sprays in order to evaluate potential errors and to evaluate if additional information about the evaporation history of the spray can be gained. Three fluorescing tracers with different volatilities were applied in ethanol: acetone and toluene with boiling points at 56°C and 110°C resp. and coumarin 47 as a solid tracer where no evaporation is expected.

2. Background

Before applying the combined LIF/Mie technique in a spray flame several experiments were performed in order to determine correct experimental parameters in terms of laser wavelength and intensity, detection bandpass, and tracer concentration.

- The absorption and emission spectra of the tracers must be known in order to select the excitation wavelength and the appropriate filters for signal detection
- The range of tracer concentrations that ensure a cubic dependence of the LIF signal on the droplet diameter and the squared dependence of the Mie scattered light on the droplet diameter must be determined
- The linearity of the LIF signal vs. tracer concentration must be investigated. The goal is to ensure, that the LIF signal stays proportional to the number of tracer molecules even during evaporation-induced enrichment
- The dependence of the LIF signal on laser intensity must be assessed. The goal is to find a regime, where the LIF signal is proportional to the laser fluence, like it is for the Mie scattered light. In this case, the LIF/Mie ratio is independent of the laser fluence

In evaporating sprays differences in evaporation and transport properties between fuel and tracer can cause enrichment and depletion of tracer concentrations which affects the accuracy of LIF/Mie dropsizing. On the other hand, if the effect is quantified, temporal variation in the concentration of tracers with different volatility yields information about the evaporation process.

Tracer	acetone	toluene	coumarin 47
Chem. formula	<chem>CH3COCH3</chem>	<chem>C6H5CH3</chem>	<chem>C14H17NO2</chem>
Boiling point [°C]	56.2	110	n/a
Melting point [°C]	−95	−95	73
Fluorescence range for excitation at 248 nm	330 nm – 530 nm	280 – 330 nm	420 – 520 nm
Fluorescence quantum yield, excitation at 248 nm	~ 0.001	0.17	> 0.9
Absorption cross section at 248 nm [l/mol cm]	12	120	2.1×10^4
Molecular mass [amu]	58.08	92.14	231.29

Table 1: Physical and chemical properties of the three tracers at room temperature. For acetone and toluene the spectroscopic data is taken in an atmosphere of N_2 , for coumarin the spectroscopic data is for a solution in ethanol [5-7].

If the tracer is more volatile than the fuel (i.e. acetone in ethanol) the tracer concentration decreases during the evaporation. The tracer-LIF signal which is proportional to the concentration then underpredicts the actual droplet volume D^3 . Since the Mie intensity is not affected, the LIF/Mie ratio (apparent SMD) is smaller than the SMD. If the tracer is less volatile (tolu-

ene in ethanol) or has a negligible vapor pressure (coumarin 47 in ethanol) the tracer accumulates during evaporation yielding an apparent SMD larger than the actual SMD.

These effects, however, only occur if the evaporation is slower than the transport processes within the liquid phase. This applies to the spray flame we investigate in this work. In fast evaporating sprays (i.e. Diesel sprays) the tracer accumulation and depletion may be partially prevented. In this case, the LIF/Mie ratio represents the actual SMD. It should be noted, that several fluorescent tracers are strongly temperature-dependent [8]. This effect is not further discussed here, since T -variations in our experiment are moderate (exit temperature: 40°C, fuel boiling point: 78°C).

For our measurements we excite acetone, toluene and coumarin 47 at 248 nm (KrF excimer laser). The absorption cross sections at 248 nm, the fluorescence range for excitation at 248 nm and other physical and chemical properties of these tracers are shown in Table 1. The fluorescence emission is well separated from the excitation wavelength. Scattered laser light can therefore be efficiently suppressed by longpass filters when measuring the fluorescence.

3. Experimental

The experimental setup is shown in figure 1. The beam of a KrF excimer laser (Lambda Physik EMG 150 TMSC, 18 ns pulse duration) is focused with a cylindrical lens into a light-sheet 3 mm wide and 70 mm high. The laser fluence is varied up to 4.5 MW/cm² to investigate linearity, all other measurements are carried out at 70 kW/cm². The light passes through a spray flame where ethanol (0.47 g/s) is atomized by a pressure atomizer (Delavan 67700-5) [9] within an air coflow (0.64 m/s). At the same position for calibration and for the investigation of signal dependence on droplet size and tracer concentration a monodisperse droplet generator with 25 – 75 µm droplet diameter or a nebulizer ($D < 10$ µm) can be installed. The signal light is imaged (Nikon, UV lens, $f = 105$ mm, $f_{\#} = 2$) onto two image intensified CCD cameras (LaVision, StreakStar, 100 ns gate) with one pixel representing an area of 250 x 250 µm². For the detection of the acetone, toluene and coumarin LIF, combinations of Schott WG280 + BG1, WG280 + UG5 and GG375 + UG5 filters are applied, resp.

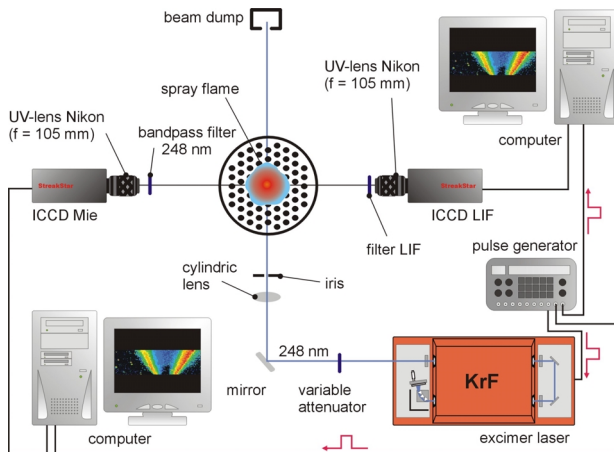


Figure 1: Experimental setup for LIF/Mie imaging in the spray flame and the investigation of the dependence of LIF and Mie signals on droplet diameter, laser fluence and tracer concentration. The droplets were produced either by a pressure atomizer, a monodisperse droplet generator or a nebulizer.

In the spray flame the area of interest is 69 x 61 mm², 30 mm above the nozzle. LIF and Mie signal images are carefully mapped and 1000 single images are taken for each tracer. Scattering off metal surfaces and chemoluminescence are negligible. A significant interference originates from LIF signals of polycyclic aromatic hydrocarbons (PAH), that are generated in the flame. Their relative average contribution to the tracer LIF signal is strongly dependent on the spatial position. Within the area in spray cone, where most of the liquid mass is found the relative PAH-LIF contribution to the tracer LIF signal is below 20 %. These signals spectrally overlap with tracer LIF and cannot be completely suppressed. Most of this additional signal contribution occurs in regions far away from the droplets in the gas phase. The simultaneously acquired Mie images are used to determine the area where no droplets are present. LIF

signal from this area is discarded. There remains an additional contribution from the immediate surrounding of the droplets. This is eliminated by subtracting an average PAH LIF signal contribution (LIF image measured without tracer in the system). After this correction, LIF and Mie intensities are averaged and the ratio is calculated as a relative SMD which is then calibrated by measurements in monodisperse droplets from the piezo-driven droplet generator.

4. Results

4.1. Dependence of LIF and Mie Signal on the droplet diameter

The left diagram in Figure 2 shows the toluene LIF and the Mie scattering signal per droplet in dependence on the droplet diameter. The LIF data are fitted ($I_{LIF} = C_{LIF} D^{n_{LIF}}$) with the exponent $n_{LIF} = 3.0 \pm 0.2$. For the Mie data a similar equation ($I_{Mie} = C_{Mie} D^{n_{Mie}}$) yields $n_{Mie} = 2.0 \pm 0.28$. The error bars are caused by fluctuations of the laser intensity at the place of the droplet chain. The right diagram in Figure 2 shows the ratio of LIF and Mie signals as a function of droplet diameter. It is well described by a linear regression.

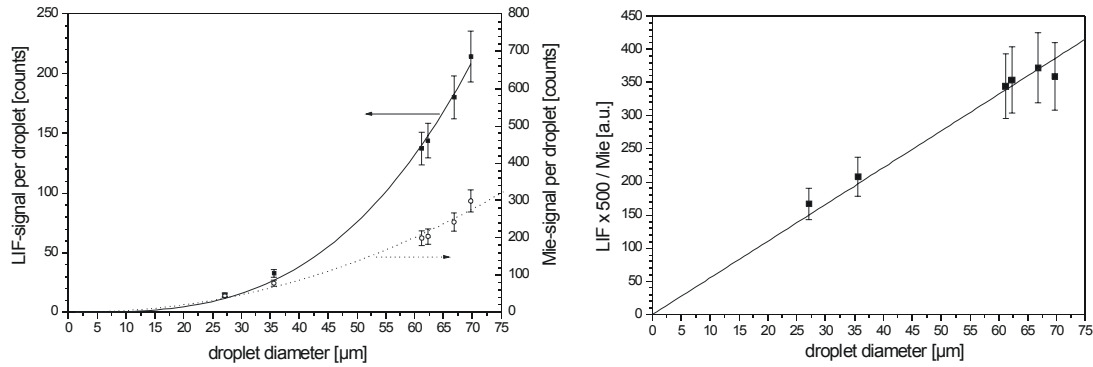


Figure 2: Left: Toluene LIF- and Mie scattering intensity in dependence on the droplet diameter in a monodisperse droplet chain. Right: LIF/Mie signal ratio. The toluene concentration was 0.019 mol/l, the laser intensity was 70 kW/cm².

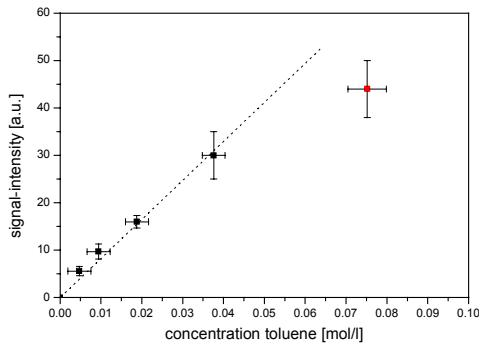


Figure 3: Toluene-LIF-intensity dependent of the toluene concentration. Laser intensity: 0.075 MW/cm². The spray was produced by a nebulizer and consisted of droplets smaller than 10 μm.

4.2. Dependence of tracer LIF on the tracer concentration

The tracer concentration influences the linearity of $LIF \sim D^3$ at high concentrations due to self-quenching and attenuation of laser light in the droplet. Choosing a low enough tracer concentration is therefore crucial especially in evaporating sprays. For small concentrations up to 0.04 mol/l toluene the LIF signal depends linearly on the concentration (Figure 3). For higher concentrations self-quenching effects cause a deviation of the linear dependence. The deviation of the linear dependence seems to occur at smaller concentrations already, but this is an artefact caused by concentration inaccuracies. For the measurements in the spray flame the concentration of 0.018 mol/l has been selected. Even if during the evaporation process in the flame a complete toluene enrichment occurred, a droplet could lose half of its mass be-

fore the onset of significant self quenching. Similar experiments with coumarin and acetone showed a linearity up to 1.9×10^{-5} and 0.8 mol/l resp. For the spray-flame measurements concentrations of $1.73 \times 10^{-6} \text{ mol/l}$ (coumarin) and 0.68 mol/l (acetone) were chosen.

4.3. Dependence of the LIF signal in the laser intensity

Figure 4 shows the dependence of toluene LIF on laser fluence. Below 0.5 MW/cm^2 a linear relation is found for all three tracers. For the measurements in the spray flame the laser intensity was set to $0.05 - 0.1 \text{ MW/cm}^2$.

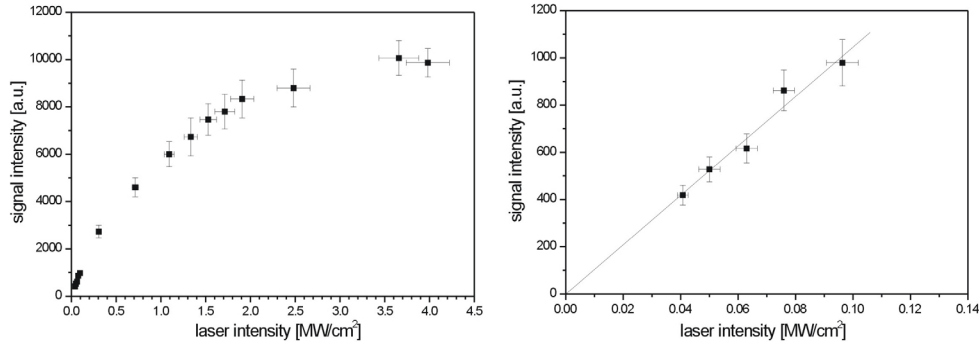


Figure 4: Toluene LIF as a function of laser intensity.

5. LIF/Mie dropsize imaging in the spray flame

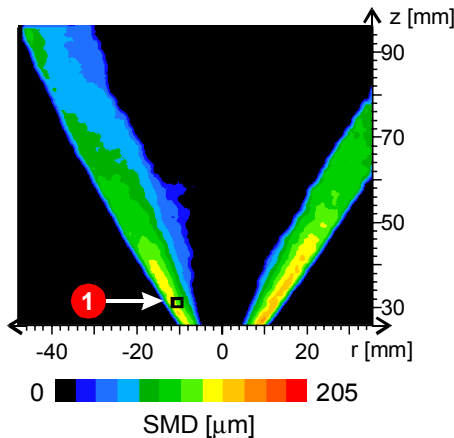


Figure 5: Two-dimensional distribution of the Sauter mean diameter in the reacting spray based on the assumption of invariant toluene concentration in the spray. The nozzle position is at the position $r = 0$ 26 mm outside the image ($65 \times 75 \text{ mm}^2$).

Figure 5 shows the droplet size (SMD) distribution in the spray flame based on toluene as tracer. Here, no tracer enrichment is taken into account. In the next paragraph these effects will be discussed. The maximum SMD is ca. $150 \mu\text{m}$ close to the nozzle on the outer side of the hollow cone spray. With increasing distance from the nozzle and towards the burner axis the SMD decreases rapidly. The LIF/Mie drop sizing technique requires that the signal is averaged over several droplets. Therefore, only pixels were taken into account, that detected a minimum of 64 times (out of 1000) liquid phase signal. These are within the spray cone, where nearly all the liquid mass is located.

With toluene and acetone as tracer, LIF/Mie ratios show systematic errors compared to the true SMD caused by tracer enrichment or depletion. The boiling point of the fuel (ethanol, 78°C) is in between those of toluene (110°C) and acetone (56°C). Furthermore the diffusion coefficients of toluene and acetone in ethanol are similar (acetone: $K_1 = 3.54 \times 10^{-3} [\text{cm/s}]^{0.5}$, toluene: $K_1 = 3.04 \times 10^{-3} [\text{cm/s}]^{0.5}$ [10]). Therefore, their transport (neglecting the convective transport) in the droplet is expected to be similar. It is therefore a reasonable assumption that the true SMD is in between the apparent SMD obtained from toluene and acetone as LIF/Mie measurements.

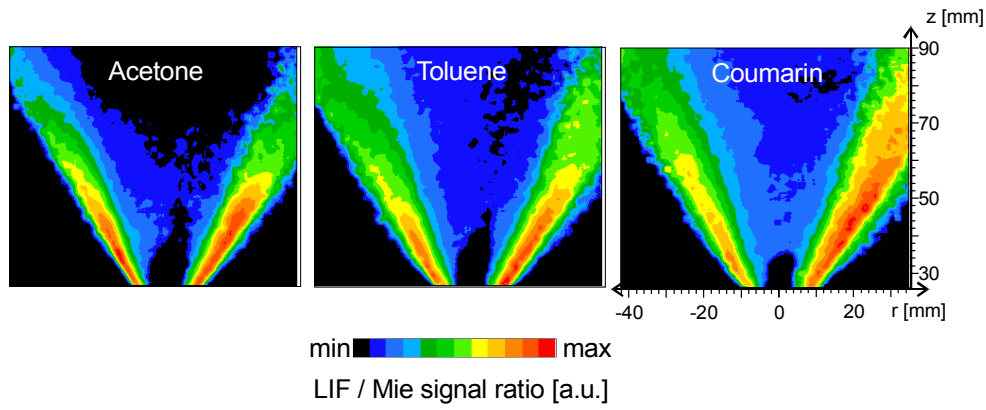


Figure 6: LIF/Mie-ratios for acetone, toluene und coumarin as tracers. At 27.5 mm above the nozzle the maximal values were normalized to each other.

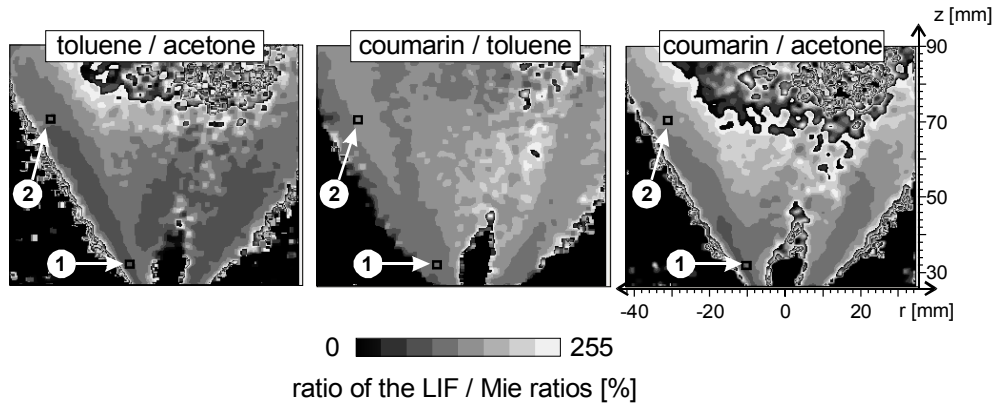


Figure 7: Ratio of the LIF/Mie-ratios for acetone, toluene und coumarin.

Figure 6 shows the relative apparent SMD distributions for all three tracers. 27 mm above the burner (lower end of the images) the distributions are normalized. For all tracers the highest values are found close to the nozzle. With increasing distance from the nozzle and increasing distance to the spray cone the values decrease significantly. Acetone shows the strongest decrease, whereas coumarin shows the smallest decrease. Acetone and toluene show only small variations. The true SMD is expected to lie in between the apparent SMD based on acetone and toluene LIF. Therefore, on the left side of fig. 7, the ratio of these two apparent SMD-distributions is shown. Of main interest is the spray cone where most of the droplets are found. At the lower end of the figure, the apparent SMDs are normalized to each other and their ratio at pos. “1” is close to 100%. Due to tracer enrichment the ratio changes and at position “2” reaches a value of 120%. This means that the relative influence of the evaporation on the SMD-values measured with toluene or acetone on the edge of the hollow cone spray is restricted to 20% and the accuracy of SMD values shown in fig. 5 is better than 20%. Inside the spray cone, where the temperatures are high caused by recirculation of hot gases and where mainly small droplets are found, the differences between the apparent SMD-values are significantly higher.

6. Comparison with a simple evaporation model

For coumarin complete enrichment is expected if the evaporation is in equilibrium. Therefore, the LIF-signal of coumarin represents the initial droplet volume. The middle and right panel in fig. 7 show the ratios of the apparent SMD values of coumarin/toluene and coumarin/acetone. Their relative variation from position “1” to position “2” will now be compared with the simple D^2 evaporation model (eq. 3). Within this model the evaporation of a droplet with the

diameter D at a fixed temperature T is described. This diameter is considered to be the SMD of our spray, $D = \text{SMD}$.

$$D(t) = \sqrt{D_0^2 - \lambda t} \quad (3)$$

This relation holds in the so-called steady state period, where all the energy transferred to the droplet is consumed for evaporation. D_0 represents the diameter at the beginning of the evaporation process, λ is the evaporation constant, which depends on temperature. We assume here, that the droplets are exposed to a homogenous temperature field at $T = 950$ K and that the evaporation process is already in the steady state period. For this temperature the value of the evaporation constant is $\lambda = -0.19 \text{ mm}^2/\text{s}$ [11].

For coumarin in a slow evaporating system no evaporation occurs and the LIF / Mie ratio represents the initial droplet volume divided by the actual droplet surface. Within the D^2 law this ratio is given by the following expression and denoted with $D_A(t)$:

$$D_A(t) = \frac{D_0^3}{D^2(t)} = \frac{D_0^3}{(D_0^2 - \lambda t)} \quad (4)$$

The true diameter of the evaporating droplet is given by $D(t)$ (cf. eq. (3)). The ratio of the LIF/Mie ratio for the completely enriching tracer and the real diameter ($D_A(t) / D(t)$) is given by the following equation and shown in Figure for different initial diameters D_0 .

$$\frac{D_A(t)}{D(t)} = \frac{D_0^3}{(D_0^2 - \lambda t)^{3/2}} \quad (5)$$

It takes approx. 9 ms for a droplet to move from pos. 1 to pos. 2 (Figure 7). ($D_A(t) / D(t)$) increases with time strongly depends on the initial droplet size D_0 . For an initial diameter of 200 μm there is only a change of a few percent. For an initial droplet size of 40 μm the change is more than one order of magnitude.

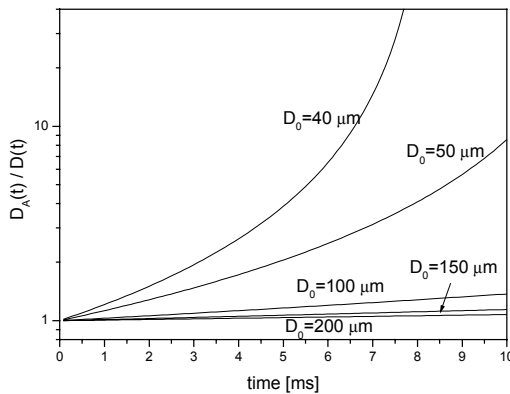


Figure 8: Ratio of apparent droplet diameter $D_A(t)$ (for complete tracer-enrichment) and real droplet diameter $D(t)$ (tracer evaporating like the fuel) depending on the evaporation time for different initial droplet diameter D_0 .

According to fig. 5 the SMD at pos. 1 is approx. 100 μm . Figure 8 shows, that the ratio ($D_A(t) / D(t)$) for a droplet with an initial diameter of 100 μm has increased by 32 % after 9 ms. This can be compared with the relative increase from position 1 to position 2 in fig. 7. The coumarin/acetone ratio increases by 40%. It has to be taken in account, that the model compares a completely enriching tracer (i.e. coumarin) with one that evaporates like the fuel whereas acetone evaporates even faster than the fuel. The coumarin/toluene ratio in contrast shows an increase of approximately 20%, which is too small compared to the model prediction. In this case, however, the experiment compares a completely enriching tracer with a one that also shows (weaker) enrichment. Therefore, the model predicts the trend of the experimental observation correctly. This indicates within the model assumptions that the evaporation process is slow enough to allow enrichment processes of the tracers in ethanol. For the following reasons the model should be considered as an approximation rather than a detailed calculation:

- The temperature is fixed to 950 K. So far, no temperature data of this flame exists.
- The droplet velocity was assumed to be 5 m/s for all droplet sizes. But the droplet velocity decreases with increasing distance from the nozzle due to friction and gravitation effects. Therefore, droplets with different sizes have different velocities.

- Steady state evaporation was assumed
- The experimental droplet size distribution characterized by the measured SMD was compared with a monodisperse droplet in the model. In order to describe the evaporation of a polydisperse spray it would be better to consider the droplet size distribution rather than the SMD

Based on these assumptions, from the comparison of LIF/Mie measurements with coumarin and toluene the evaporation along the direction of the flow (between pos. 1 and 2, cf. fig 9) can be estimated.

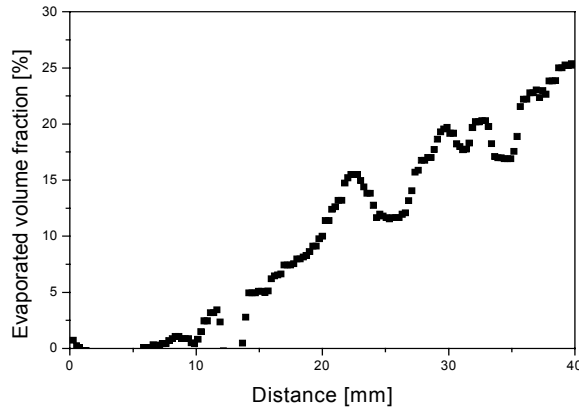


Figure 9: Fraction of evaporated fuel (by liquid volume) along the spray cone between pos 1 and 2 (cf. fig. 7). Obtained from the ratio of coumarin and toluene LIF/Mie ratio images.

7. Conclusions

LIF / Mie dropsizing is a non-intrusive laser-based measurement technique for the planar detection of the Sauter mean diameter (SMD) of a spray. While it is well studied in non evaporating sprays, systematic errors occur due to tracer enrichment and depletion of tracers during evaporation. By comparing the results obtained with tracers with different volatilities these systematic errors were specified in an ethanol spray flame with maximum SMD of $\sim 150 \mu\text{m}$. Within the spray cone where the major part of the liquid is located the apparent SMD values for the two tracers applied varied by 20% within the observed region.

Tracers with negligible vapor pressure (i.e. laser dyes) give LIF signals that represent the initial liquid volume. They allow to assess the spray evaporation with a simple evaporation model. It was found that the evaporation process is slow compared with the transport inside of the droplet which allows complete tracer enrichment.

8. References

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