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# Quantitative nitric oxide measurements by means of laser-induced fluorescence in a heavy-duty Diesel engine

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## Abstract

Quantitative in-cylinder laser-induced fluorescence measurements of nitric oxide in a heavy-duty Diesel engine are presented. Special attention is paid to experimental techniques to assess the attenuation of the laser beam and the fluorescence signal by the cylinder contents. This attenuation can be considerable at certain stages in the combustion stroke. The temperature and pressure dependence of the fluorescence signal is described in various models. In this study, LIFsim was used. Finally, calibration was realized by concentration measurements in the exhaust gas.

## 1. Introduction

In order to improve the exhaust emissions of Diesel engines in terms of particulate matter and oxides of nitrogen (NO<sub>x</sub>), knowledge of their respective formation processes is indispensable. Laser techniques offer robust diagnostics for measurements at high spatial and temporal resolution, withstanding the extreme combustion conditions where other methods may fail.

In this paper, we present quantitative laser-induced fluorescence (LIF) measurements of nitric oxide (NO), conducted in an optically accessible Diesel engine. In order to interpret the raw NO LIF signal in terms of NO number densities or concentrations, a number of effects need to be taken into account. The LIF signal is linearly proportional to the number of illuminated NO radicals, and, in the non-saturated case, also to the laser intensity. In diesel combustion, the attenuation of the laser beam may be considerable; the main causes are absorption (or scattering) by fuel droplets or vapour, aromatic fuel compounds, soot particles, H<sub>2</sub>O, and CO<sub>2</sub>. Considering the significant attenuation of (UV) light in IC engines (e.g. [1–3]), this explains the need of measurements on the local (i.e. spatially resolved) laser beam transmission. Similar measurements are needed for the fluorescence transmission. Furthermore, the strong temperature and pressure dependence of the fluorescence efficiency needs to be accounted for. Over the last two decades, various models were developed to describe this dependence [4–6]. In the analysis presented here, we used LIFsim [6] to predict the fluorescence behaviour under engine conditions, using recorded pressure traces and the average gas

temperature (calculated from the pressure assuming ideal gases) as input data.

The spatially resolved laser beam transmission can be obtained by the bidirectional laser scattering method (e.g. [7]). In this case, NO radicals are (quasi) simultaneously illuminated by two laser beams, traversing the same probe volume but in opposite directions. Recording the fluorescence (or any other scattered light) from each of the two laser pulses separately, the *local* attenuation coefficient can be retrieved from the ratio of the two intensity distributions [7, 8]. Recent modifications to the algorithm, necessary to cope with the extreme noise amplification that is inherent to this technique, will be published elsewhere [8].

UV absorption by CO<sub>2</sub> is one of the major contributors to the attenuation of the fluorescence signal. Deviations of the observed NO LIF spectrum to the theoretical spectrum perfectly match the well-known absorption spectrum of CO<sub>2</sub> [9]. By fitting this spectrum to the normalized NO LIF spectrum, the fluorescence attenuation caused by CO<sub>2</sub> is readily obtained [8]. Possible additional (broadband) attenuation, caused by in-cylinder soot, does not influence this method, affecting all NO LIF peaks in the same way. It is not at all straightforward to measure soot-based attenuation of the fluorescence, and it is not included in our analysis. Attenuation by soot deposits on the detection window, however, has been taken into account. It is reflected in the declining trend in (raw) NO LIF intensity (each time measured at a fixed crank angle) during a measurement session. This trend is caused by increasing window obscuration, and can be used to correct for that effect [8].

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## 2. Specific objectives

The aim of this paper is to present the various data processing steps that are needed for quantitative NO LIF measurements. Some of these steps involve experimental techniques, while others are based on a LIF model, also requiring experimental data for input. We will discuss the significance of each of these processing steps on the final, quantitative NO data.

## 3. Experimental Setup

### 3.1. The engine

The research engine in Nijmegen is a heavy-duty, six-cylinder truck engine (DAF trucks, NL). The measurement cylinder corresponds to the modern DAF 95XF engine type. A schematic representation is given in Figure 1. It is equipped with a number of quartz windows for optical access: there are three windows in the cylinder wall, giving a view to the uppermost part of the combustion chamber. A slot machined into the piston crown prevents complete blocking of one of the side windows even at Top Dead Centre (TDC). In addition, there is a window instead of one of the exhaust valves, and a large one in the piston bowl. The (cam-driven) fuel injection takes place through an eight-hole nozzle, located in the centre. The injector can be rotated, which allows measurements at different positions relative to the fuel sprays without repositioning the laser beam. The exhaust valve window can be replaced by a pressure transducer (AVL QHC32), to measure the pressure trace needed for the LIF model. No lubricants are used since they absorb the UV laser radiation; to avoid overheating the measurement cylinder is skip-fired. Steady-state conditions are mimicked by (pre-) heating the cooling water to operational temperatures.

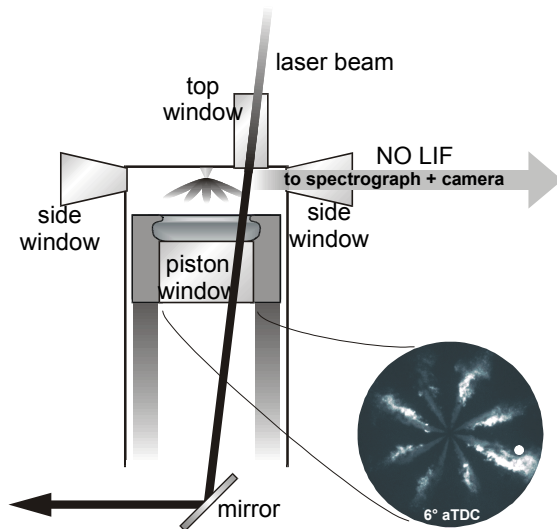


Figure 1: Schematic representation of the measurement cylinder. The laser probe location is indicated as a white dot in the piston window view, lower right corner.

Further details of the modified cylinder and some engine characteristics can be found in reference [10], but will be published in more detail elsewhere, including the high-pressure fuel injection system (1200 bar) that was installed recently [11].

The NO concentration in the exhaust gas is determined by an exhaust gas analyzer (SIGNAL Instruments, NOX analyzer series 4000).

During the experiments presented here, the engine was operated at 25% load. Under these conditions, 62 mg of low-sulfur Diesel fuel is injected per cycle between 5° bTDC and 6° aTDC, resulting in a gross indicated mean effective pressure (gIMEP) of 411 kPa.

### 3.2. NO LIF setup

For all measurements on NO LIF presented here the laser beam was directed through a fuel spray (marked by the white dot in the spray image in Figure 1).

NO is measured by means of laser-induced fluorescence (LIF), using laser radiation at 226.03 nm for excitation. This corresponds to the coinciding  $P_1(23.5)$ ,  $Q_1+P_{21}(14.5)$ , and  $Q_2+R_{12}(20.5)$  lines in the  $A^2\Sigma(v''=0) \leftarrow X^2\Pi(v''=0)$  transition. An unfocused (~3.5 mm diameter) 226 nm laser beam of 2.5 to 5 mJ pulse energy is directed through the combustion chamber, either entering via the top window and leaving it through the piston window, or vice versa, for the bidirectional NO LIF measurements. The laser radiation is produced by a frequency-doubled dye laser (Lambda Physik ScanMate 3 using Coumarin 47) pumped by a Nd:YAG laser (Continuum PowerLite 9010), or by a frequency-mixed dye laser (Radiant Narrowscan D, using Rhodamin 101) pumped by a Nd:YAG laser (Continuum Powerlite Precision II 8010).

The fluorescence signal (237, 248, 259, and 270 nm) is detected through the nearest side window by an intensified CCD camera (Roper Scientific, ICCD 512T, 16 bits) mounted behind a spectrograph (ARC SpectraPro 500i). In this configuration, the path lengths of the laser beam and fluorescence signal are minimal, thus minimizing their respective attenuation. The grating spectrograph allows us to obtain data at both one-dimensional spatial resolution (i.e. along the laser path) and spectral resolution. An example is shown in Figure 2. This spectral information is advantageous for three reasons. First, it allows us to subtract any possible broadband background (negligible in our measurements) caused by laser-induced incandescence of soot particles, or LIF of fuel components. Second, interference by  $O_2$  LIF can be monitored (also found to be minimal in our experiments). Third, the observed discrepancy between the measured and theoretical LIF spectrum can be used to assess the fluorescence absorption by  $CO_2$  (which can be considerable at certain crank angles, as will be shown in the next section).

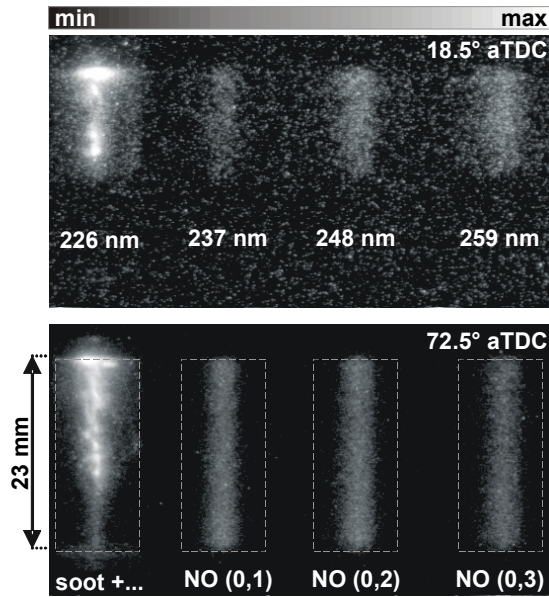


Figure 2: Two typical NO spectra, together with elastic scattering (on the far left). NO fluorescence can be seen at 237, 248, and 259 nm.

#### 4. Results and Discussion

##### 4.1. $P, T$ dependence of the LIF signal

The cylinder pressure trace is shown in Figure 3, along with the average gas temperature. The latter can easily be calculated from the cylinder pressure, if ideal gas behaviour is assumed, and heat loss and gas leakage are neglected [12]. Apart from the error induced by these assumptions, the obtained *global* temperature is by no means equal to the *local* temperature at the laser probe region (the local temperature may be substantially higher or lower), while the latter is needed for NO LIF quantification. Therefore, local temperature measurements, e.g. by two-line thermometry of NO [13], would be preferable. Although such measurements are currently in preparation, the analysis in this paper will be based on the global temperature.

Figure 4 shows the effect of the cylinder pressure and temperature on the NO fluorescence. This curve has been obtained by feeding pressure and temperature values at various crank angles to LIFsim [6]. The strong effect of quenching at high cylinder pressures (i.e. small crank angles) can clearly be seen. During expansion, the LIF signal increases drastically, by up to a factor of 20. The sensitivity of the LIF signal to temperature is not as pronounced as its pressure dependence. Two additional LIF yield curves are shown in Figure 4, for input temperatures increased by a factor of 1.5 and 2, respectively. All three curves are scaled to the same value at 130° aTDC, because this will be the calibration point. At 130° aTDC the exhaust valve opens and the LIF signal can be related to the exhaust concentration, where we assume that the contents in the probe volume are representative of the total cylinder contents at this stage in the stroke.

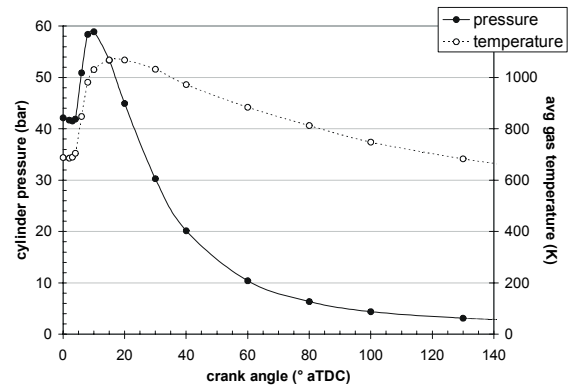


Figure 3: Cylinder pressure (measured) and average gas temperature (calculated) versus crank angle.

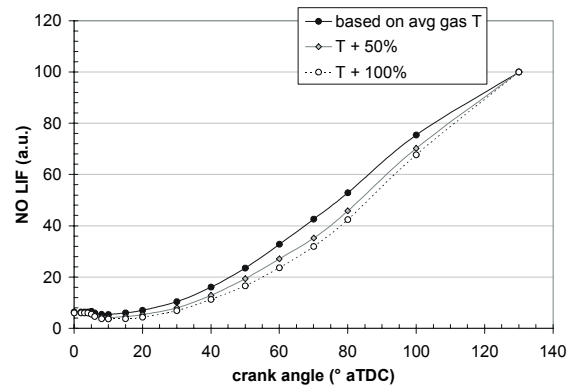


Figure 4: Fluorescence yield (arbitrary units) versus crank angle, based on the LIFsim model and the data in Figure 3. Additional curves have been added to show the sensitivity on temperature. For comparison, all curves are scaled to 100 at 130° aTDC.

##### 4.2. Laser beam and LIF transmission

Bidirectional laser scattering is, at least in principle, a powerful method to obtain spatially resolved laser beam transmission information. Together with the CO<sub>2</sub> absorption analysis of the NO LIF spectrum, assessment of (almost) all attenuation contributions involved in our measurements is now possible. Figure 5 shows the combined laser beam and fluorescence transmission, based on the results of these two methods [8]. Basically, the combined transmission is a multiplication of the laser beam transmission and the fluorescence transmission curves.

From Figure 5 it becomes clear that attenuation has a significant influence on the NO LIF measurements. Around 20° aTDC, the combined transmission is less than 30%. It should be remarked that the transmission of laser and fluorescence light is strongly dependent on the path lengths involved. In this experimental configuration, the laser beam immediately enters the

field of view, and has to travel at most 23 mm (compare to the stroke of 146 mm). The fluorescence reaches the detection window after 37 mm. Although this is slightly longer than the laser beam traveling distance, the red-shifted fluorescence is less affected by CO<sub>2</sub> absorption (which increases towards shorter wavelengths).

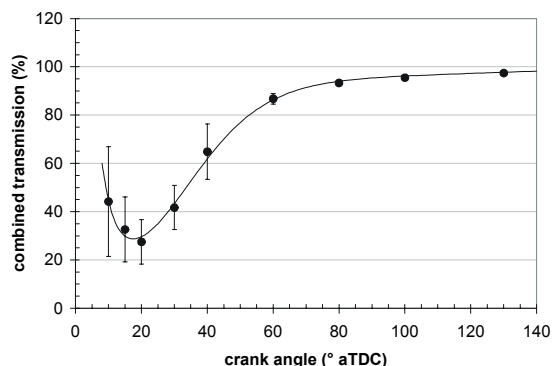


Figure 5: Product of the laser beam transmission and fluorescence transmission. The estimated error bars are based on the standard error in the fluorescence transmission, assuming similar accuracy in the laser beam transmission.

#### 4.3. Quantitative NO LIF results

The raw NO LIF images, like the examples in Figure 2, are processed by summing the pixel intensities of the three NO regions (i.e. the rectangles in the lower panel of the figure). Background intensity (and camera dark current) is corrected for by subtracting the average pixel intensities of the regions in between. In Figure 6, the obtained total LIF intensity is plotted versus crank angle. It shows that, measuring in the spray, NO LIF can be detected as early as 2° aTDC. The strong increase in the LIF signal is only partly caused by NO formation: the decrease in cylinder pressure (due to expansion) also decreases the effect of quenching, which in turn causes an increase of the NO LIF signal, even if the NO concentration would remain constant.

The curves in Figures 4 and 5 are used to process the data in Figure 6. Absolute values can be obtained by scaling the resulting curve to the calibration point at 130° aTDC. Independent measurements of the NO concentration in the exhaust gas revealed a concentration of 560 ppm. The resulting NO curve is shown in Figure 7.

Even with the corrections for laser beam and fluorescence transmission, the tiny NO peak around 2-3° aTDC remains. Interestingly, this NO signal is present before the rise in heat release at ~4° aTDC. It may disappear due to NO reburn chemistry in fuel-rich zones (the laser is probing through the fuel spray), or due to transport out of the probe volume by the swirling motion of the air, or perhaps by diffusion out of the probe volume (what we present is a *local* measurement).

After the fast rise between 10 and 20° aTDC, the NO concentration levels off, increasing only very slowly. Considering the uncertainty in the local temperature, this slow increase is not significant: if the T+50% curve from Figure 4 is used, the NO concentration remains constant (within the error bars) after 30° aTDC. The curves for T+50% and T+100% are added to Figure 7 as respectively solid and dotted lines. The fact that the NO concentration reaches a constant value indicates a (more or less) homogeneous NO distribution. This justifies our choice of calibration by the exhaust gas concentration.

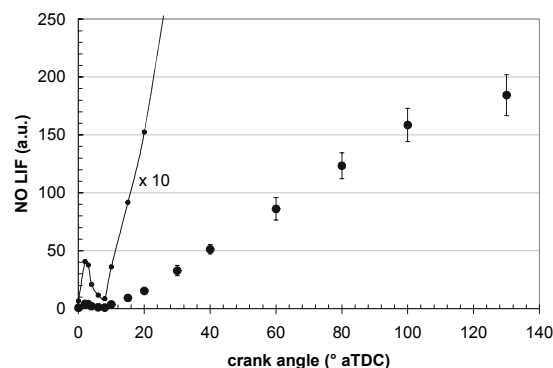


Figure 6: Total (raw) NO LIF versus crank angle. The data points correspond to averages of 15 laser shots; the error bars denote the standard error.

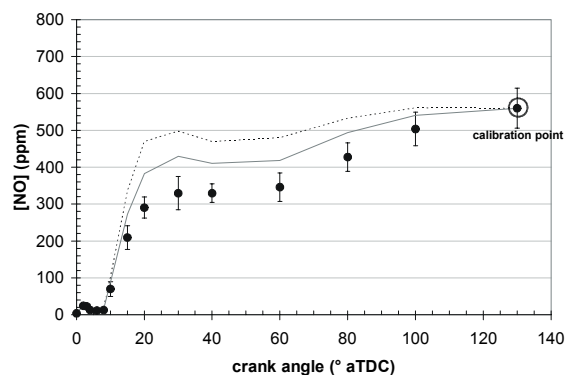


Figure 7: NO concentration during the combustion stroke, based on the data in Figure 6. The data points correspond to averages of 15 laser shots; the error bars denote the standard error. The additional lines are based on the T+50% and T+100% curves from Figure 4.

## 5. Summary and Conclusions

We have demonstrated that quantitative, in-cylinder NO measurements using laser-induced fluorescence are feasible even in the hostile combustion environment of a Diesel engine. Conversion of raw NO LIF signals into ppm's requires knowledge of a number of conditions, such as cylinder pressure, the local temperature, and the attenuation of the light involved in the measurement. In the following, we will briefly discuss the influences of these factors, in order of importance.

Of all the processing steps discussed in the previous section, the pressure dependence of the NO signal (through quenching and, to a lesser extent, line broadening) is by far the strongest effect, changing by almost a factor of twenty during the combustion stroke. Fortunately, the pressure transducer allows accurate pressure measurements, and quenching rates are relatively well-known.

The effects of laser beam and fluorescence transmission are considerable in this engine, despite minimization of the path lengths involved. Affecting the fluorescence signal by more than a factor of three, varying through the combustion stroke, attenuation effects cannot simply be neglected. Reliable methods, such as bidirectional laser scattering and NO LIF absorption by CO<sub>2</sub>, are needed to assess this attenuation.

Although the local temperature is less easy to obtain, the temperature dependence of the LIF signal is not as critical as its pressure dependence. The T+50% and T+100% curves in Figure 4 deviate from the average-gas temperature-based curve by typically 20% and 30%, respectively. In order to reduce this uncertainty, local temperature data are indispensable. Several laser-based diagnostic techniques exist to measure temperature distributions in combustion environments, two-line vibrational thermometry of NO [13] being the most obvious in this case.

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## References

- [1] J.E. Dec and R.E. Canaan, SAE Paper No. 980147 (1998)
- [2] C. Schulz, J.B. Jeffries, D.F. Davidson, J.D. Koch, and R.K. Hanson, Proc. Comb. Inst. 29, 2735-2742 (2002)
- [3] K. Verbiezen, A.J. Donkerbroek, A.P. van Vliet, W.L. Meerts, R.J.H. Klein-Douwel, N.J. Dam, and J.J. ter Meulen, Proc. ECM 2003 (2003)
- [4] J. Luque and D.R. Crosley, SRI International Report MP99-009 (1999), [www.lifbase.com](http://www.lifbase.com)
- [5] M.D. DiRosa, K.G. Klavuhn, R.K. Hanson, Comb. Sci. and Tech. 118, 257-283 (1996)
- [6] W.G. Bessler, C. Schulz, V. Sick, and J. W. Daily, 3rd Joint Meeting US Sec. Combust. Inst. (2003), [www.lifsim.com](http://www.lifsim.com)

- [7] G.G.M. Stoffels, S. Stoks, N. Dam, and J.J. ter Meulen, Appl. Opt. 39, 5547-5559 (2000)
- [8] K. Verbiezen, A.P. van Vliet, R.J.H. Klein-Douwel, A.J. Donkerbroek, W.L. Meerts, N.J. Dam, and J.J. ter Meulen, in preparation (2005)
- [9] C. Schulz, J.D. Koch, D.F. Davidson, J.B. Jeffries, and R.K. Hanson, Chem. Phys. Lett. 355, 82-88 (2002)
- [10] E.J. van den Boom, P.B. Monkhouse, C.M.I. Spanjaars, W.L. Meerts, N.J. Dam, and J.J. ter Meulen, Proc. SPIE 4430, 593-606 (2001)
- [11] K. Verbiezen, R.J.H. Klein-Douwel, A.P. van Vliet, N.J. Dam, and J.J. ter Meulen, in preparation (2005)
- [12] J.B. Heywood, Internal Combustion Engine Fundamentals, McGraw-Hill, Singapore (1988)
- [13] W.G. Bessler, F. Hildenbrand, and C. Schulz, Appl. Opt. 40, 748-756 (2001)