STUDY ON TURBULENT FLUCTUATIONS IN DIFFUSION FLAMES USING LASER INDUCED FLUORESCENCE
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STUDIE VAN TURBULENTE FLUCTUATIES
IN DIFFUSIEVLAMMEN
MET GEbruIK VAN
LASER GEïNDOUCERDE FLUORESSENTIE

Proefschrift

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Alles Ferne nahm mich in Anspruch, was in einer bloßen halben Stunde zu erreichen war und in der unerwünschten Richtung, war wie die Hinterseite des Mondes, unsichtbar, nicht vorhanden. Man meint sich für die Welt zu öffnen und zahlt dafür mit Blindheit in der Nähe. Unfaßbar ist der Hochmut, mit dem man darüber entscheidet, was einen angeht und was nicht. Alle Linien der Erfahrung sind einem vorgeschrieben, ohne daß man’s weiß, was ohne Buchstaben noch nicht zu fassen wäre, bleibt ungeschen, und der wölfische Appetit, der sich Wissbegier nennt, merkt nicht, was ihm entgeht.

(Elias Canetti, Die gerettete Zunge (1977))
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Chapter 1

Introduction

1.1 Natural gas combustion

In 1960, one of the world's largest reserves of natural gas was discovered in Slochteren, Groningen (estimated volume of producible gas $2.5 \times 10^{12}$ m$^3$). The gas was of high quality (little sulphuric acid content) and easily to be won. Many other, much smaller reserves were found later, both at land and on the continental shelf; until 1988, 98 locations had been discovered at land and 105 on the continental shelf[1]. The discovery of this valuable stock of hydrocarbon fuel changed the economic position of The Netherlands dramatically, especially after the 1973 energy crisis, when oil prices increased strongly. The government took benefit of these reserves by selling the natural gas yielding about $10^{10}$ Dutch guilders a year. About 50 % is exported. Individual users converted from use of the poisonous so-called city gas for cooking and fuel oil or coal for heating, to natural gas for both. Also in agriculture (green houses) and in industry (generation of electricity, melting of glass, in bakeries) natural gas is used extensively. In 1987, the contribution of natural gas to the total amount of energy used, was about 50 %[1]. Compared to combustion of coal or oil, combustion of natural gas is convenient and clean, because of the relatively low carbon dioxide emission and the absence of particulates produced.

Groningen natural gas mainly consists of methane and nitrogen. The usually very desirous high flame temperatures promote the production of the main pollutant originating from combustion of natural gas and air, nitric oxide (NO). This nitric oxide is poisonous, but more important, in the atmosphere it reacts to nitrous oxide and then to nitric acid, which returns to earth in the form of acid rain.

1.2 Combustion research

For efficient burning of natural gas, good mixing of fuel and air is essential. For safety reasons, the fuel and air are often supplied separately. Flames of this kind are called nonpremixed, or diffusion, flames. The mixing that is required,
is therefore to occur after the fuel exits the supply tube, by the physical process of diffusion, either laminar or turbulent. Turbulent diffusion is much faster than laminar diffusion, so turbulent mixing (diffusion) of fuel and air is most efficient. Quick oxidation of all fuel and short flames are the result, with only little time for soot formation. Because the chemical source term of the reactions giving nitric oxide depends exponentially on the temperature, the production of nitric oxide strongly depends on the peak temperatures, and the temperature fluctuations are more relevant to the NO production than the average temperature.

In a turbulent flow, the movements of gas parcels are chaotic, and, at a fixed location, the velocity fluctuates in direction and magnitude. In a flame, gradients of temperature and concentration exist, so in a turbulent flame, the concentrations and temperature will fluctuate as well. Fluctuations of temperature strongly influence the emission of NO. Combustion influences the turbulence, e.g. by thermal expansion, but also turbulence influences the rate of combustion, e.g. by fast mixing of natural gas and air or by flame extinction.

Theory to predict the properties of a chaotic, turbulent flow is not easily developed, but for a turbulent flame, with many chemical reactions going on, problems are even bigger, but also more challenging for modelling researchers. For experimental research on turbulent combustion, advanced laser diagnostic techniques had to be developed (and still are), because of the hazardous environment and because the flow and chemistry of the flame is easily disturbed by physical probes inserted in the flame. Turbulent combustion therefore is of practical interest, but also a theoretically and experimentally challenging scientific subject.

1.3 Measurements of concentration fluctuations

When turbulent flame research is considered as study of the interaction between turbulence and combustion chemistry, concentration fluctuations provide information on the turbulent mixing of species. Many (simple) combustion models are based on the assumption that chemical reactions are much faster than turbulent mixing. The models then only need to describe the mixing of one nonreactive species, or conserved scalar. This scalar usually is the mixture fraction, the normalized mass fraction of a nonreactive species issuing with the fuel. The single conserved species models, however, cannot properly describe finite-rate chemistry effects, like nonequilibrium of radical concentrations or flame extinction, or slow chemistry effects, like NO or soot formation. Therefore, models comprising detailed chemistry are currently being developed, for example in the Heat Transfer Group (Department of Applied Physics) of Delft University of Technology.

Measurements of radical concentrations are necessary to reveal the effects not predicted by the conserved scalar models. A radical particularly appropriate for this purpose is OH. It plays an important role in the breakup of methane
molecules and is one of the species that keeps the combustion reaction proceeding. It is present mainly near the reaction zone, so that it can indicate the deformation of the reaction zone due to the influence of turbulence. Also, its lifetime is short enough to follow local extinction.

Concentration measurements of radicals are possible by means of laser induced fluorescence (LIF) detection, and detection of OH is relatively easy. Laser induced fluorescence detection can be carried out in a zero-, one- or two-dimensional way. Regardless the dimensionality, LIF detection provides local, instantaneous concentration measurements. Generally, laser diagnostic techniques are well suited for experimental combustion research, because of the nonintrusive, local and instantaneous character, see also Chapter 3.

Without additional models, the conserved scalar model is not suitable to predict specific (but important) aspects of combustion, like soot and NO formation. The conserved scalar models, however, perform well in predicting main flame characteristics, like average temperatures. It is therefore interesting to carry out measurements of concentration fluctuations of an nonreactive or very slowly reacting species seeded to the natural gas flow. These measurements indicate the regions of the flame with strongest turbulent mixing of fuel and air. In addition, the influence of combustion on turbulence can be shown by comparing the mixing of this species in an isothermal jet with that in a jet flame.

1.4 Aims of this study

Even with the fastest computers of the day, the detailed chemistry modelling can only be applied to turbulent flames with simple geometry, and not to flames in industrial furnaces which generally have complex geometries. To test the models, experimental results of many different physical quantities, like concentrations of many species, temperatures and velocities, are required. These results can only be obtained under laboratory conditions. Laboratory scale turbulent flames are easily accessible for probes or laser beams, can be regulated or turned off at will, and have well-defined boundary conditions. The power of the laboratory scale turbulent flames we studied is about 25 kW, compared to flame powers used in industry often ranging from 100 kW to several MW.

In the Heat Transfer Group, several researchers collaborate to provide a description as complete as possible of turbulent co-current nonpremixed natural gas flames produced by a burner designed in the Heat Transfer Group. The co-current nonpremixed flames consist of a turbulent natural gas jet, surrounded by a turbulent primary air jet issuing from an annulus. Flame conditions are varied to study the influence of exit velocities on the flame. Variation of natural gas exit velocity and primary air exit velocity provides flexible adjustment of the mixing between natural gas and air. In addition, with a heating facility the flows can be preheated, influencing the chemical reaction rates. In total, we investigated six
flames with different conditions.

The experimental results consist of concentration measurements, which are presented in this thesis, velocity measurements, carried out and discussed by Stroomer[2], and temperature measurements for which a CARS setup is currently being constructed and which will be available in the near future. In the experimental results, emphasis is on turbulent fluctuations, expressed in standard deviations, probability density functions, and turbulence length scales. Fluctuations are also shown in this thesis by visualization of instantaneous radical concentration fields. Modelling of the flames, using detailed chemistry is carried out and discussed by Peeters[3].

The aims of this study are to provide concentration measurements, including measurements of average concentrations and of quantities describing turbulent fluctuations of the species measured. The effect of turbulence on the concentrations is studied by varying the exit velocities, and the effect of chemistry by varying the exit flow temperature. Measurement of OH was selected because turbulent fluctuations of OH show the influence of turbulence on the reaction zone and finite-rate chemistry effects, and because OH is well documented. In addition, we chose to measure NO seeded to the natural gas, to study the mixing of a conserved species by shear layer turbulence. These results uncover the qualities and shortcomings of the current models of turbulent combustion.

We aim to obtain the turbulent fluctuations, expressed in standard deviations and fluctuation distribution functions. The distribution functions of OH will exhibit effects of intermittency and of finite-rate chemistry, such as local extinction. The spatial structure of the flame under the different flame conditions will be visualized with OH, representing instantaneous cross sections of the reaction zone, and NO seeded to the natural gas, representing the shear layer mixing of fuel. Quantitative results on the spatial turbulent flame structure can be obtained from turbulence length scales of OH and NO.

Another aim is to show the influence of combustion on turbulence. This will be done by comparing the mixing of NO seeded to the fuel in an isothermal jet and in a jet flame. The difference between the length scales of reactive scalar and nonreactive scalar fluctuations will be obtained from the comparison of the length scales of OH and the NO seeded.

1.5 Outline of this thesis

This thesis is organized as follows: in Chapter 2, the general shape of the flames studied is analyzed. We analyze the different sources of turbulence in our type of flame, and the influence of these sources of turbulence on the reaction zone. The general effects of the influence of turbulence on combustion and of combustion on turbulence are described, together with the role of OH in chemical reactions and in finite-rate chemistry effects, like local extinction and superequilibrium.
1.5. Outline of this thesis

In Chapter 3, the experimental method is analyzed theoretically. For the one- and two-dimensional measurements, the fluorescence signal is not spectrally resolved. As a consequence, the influence of local temperature and composition on the fluorescence signal is stronger, complicating the relation between OH fluorescence signal and OH concentration. We develop a model to quantify these effects, resulting in an estimate of the expected biases.

In Chapter 4 we discuss the design of the burner with central fuel jet and primary air annulus, with pilot flames and heating facility. The burner was placed in a throat for secondary air. Also in Chapter 4, we present the conditions of the six different turbulent flames studied, with different combinations of natural gas and primary air velocity. Two flames were with preheated flows. Fuel exit Reynolds numbers ranged from 5000 to 10,000. Also, a special laminar premixed flat flame used to calibrate the OH concentration measurements is discussed in Chapter 4.

Experimental setups for the different measurements are discussed in Chapter 5. Concentration measurements are based on laser induced fluorescence (LIF). Species investigated are OH and NO seeded to the fuel. With spectrally resolved detection, influence of collisional energy transfer on the fluorescence signal is decreased. A setup has been constructed for spectrally resolved OH point measurements. In order to investigate the spatial structure of fluctuating concentrations, setups for broadband one- and two-dimensional detection of OH and of NO have been constructed, for quantitative analysis and visualization purposes, respectively.

Flames of the type used to calibrate the OH measurements are studied in Chapter 6, under lean and rich conditions. Results are presented of both temperature and OH concentrations, the first measured with thin thermocouples, the latter detected both spectrally selective and broadband. The results are compared with numerical simulations.

Temperatures, OH concentration fluctuations, OH visualization and turbulence length scales are discussed in Chapter 7. Visualization of local instantaneous OH fields will show the influence of large scale turbulence on the reaction zone and the existence of local extinction. Profiles of the average temperature measured with thin thermocouples will be compared with profiles of the average OH concentration, obtained from large sets of one-dimensional OH measurements. We give the shape of the probability density functions of OH, which are affected by local extinction and intermittency, and discriminate these effects by also observing the distribution functions of the peak concentration in every instantaneous profile. We will discuss the width of the OH zone, indicating the influence of velocities in the flame on the reaction zone width. We will compare the broadband detected signals of OH in a turbulent flame with the single emission-line detected signals. One-dimensional detection facilitated determination of the joint distribution functions of the local instantaneous gradient and concentration. From spatial correlation of the one-dimensionally detected measurements, we will obtain tur-
buelence length scales of OH, comprising Taylor micro scales and integral length scales.

In Chapter 8, we present results of concentration measurements of NO seeded to the fuel. In addition to the six flames, we present results of isothermal jets, with and without primary air flow. We will show that the measured concentration of NO seeded to the fuel, is not conserved in flames. Visualization of the NO seed will show the shear layer mixing in the flames. The effect of flow conditions on the mixing of the nonreactive species will be shown by profiles of average NO seed concentration and standard deviation. Distribution functions indicate the shape of the probability density function of a conserved scalar. The effect of combustion on turbulent concentration fluctuations follows from the comparison of the distribution functions in the isothermal jets and flames. We show the joint distribution of the local nonreactive scalar gradient and the concentration. The correlation between these two quantities may be used to improve the description of small scale mixing in simple conserved scalar models. Length scales of turbulence are derived from the spatial correlation of NO seed measurements, and the results will be compared with the length scale results of OH.

General conclusions on the results related to the aims mentioned above are presented in Chapter 9.
Chapter 2

Combustion theory

2.1 Introduction

The flames studied in our research are nonpremixed, or diffusion flames. In this kind of flames, the fuel and air are supplied separately. The advantage of this kind of flames is the fact that no explosive fuel-air mixtures have to be transported to the combustion chamber. Combustion is only possible in the region where the fuel and air are mixed to a ratio close to stoichiometry. The mixing is by, in our case, turbulent diffusion.

Fuel issued from a round tube into the flame chamber. The burner, studied in our research, also provided the air necessary for combustion, which issued from an annulus and which is called primary air. There is also a low velocity coflow, or secondary air flow, which is to suppress recirculation in the flame chamber. The use of a separated primary air flow, facilitated supply of preheated fuel and air flows. Because the primary air flow is also turbulent, a faster mixing between fuel and air is obtained. The burner used is discussed in greater detail in Chapter 4. Burners with turbulent air flows are also used in industry, and our burner can be compared with the so-called throughport burner discussed by Wieringa[4].

Due to the velocity difference between the fuel and primary air flow, a turbulent shear layer will form, entraining the primary air into the fuel jet. Between the primary and secondary air, there is also a turbulent shear layer. In addition to shear layer turbulence, there is turbulence originating in the burner tubes.

In this chapter we discuss the location of the reaction zone, in relation to the sources of turbulence. The flame length depends on the turbulent mixing between fuel and air, and the relation differs from that giving the flame length in the case of a fuel jet without turbulent primary air flow. We discuss the local influence of turbulence on combustion and of combustion on turbulence. The role of OH in the combustion process is analyzed, together with effects of finite-rate chemistry on OH, like local extinction and superequilibrium.
2.2 Reaction zone and shear flows

In turbulent jet flames, the most important source of turbulence is from turbulent shear layers. To understand the influence of turbulence on combustion we relate the location where the reactions take place to the region with shear layer turbulence. In our flow system with a central fuel jet and an primary air annulus jet, there is shear layer mixing fuel and primary air, and one mixing primary and secondary air, and both influence the reaction zone.

In nonpremixed flames, the combustion reactions occur in a narrow region, called the reaction zone, with a stoichiometric mixture. A mixture is stoichiometric when fuel and air are mixed on a molecular scale so that there is sufficient, but not more, air to react with all fuel. The mixture of fuel and air (or more generally, oxidizer) can be represented with the mixture fraction, $\xi$, which is unity in pure fuel and zero in pure air. In the case of combustion of Groningen natural gas and air, the stoichiometric mixture fraction is 0.070. This implies that in the region of stoichiometric mixture fraction the fuel concentration is small and we expect a large influence of the air flow on the reaction zone.

The location of the shear layers in combination with the location of the reaction zone, depends on the flow system. We observe an isothermal jet issuing in still air, the influence of a coflow on this isothermal jet and the combination of the fuel jet flow and annulus jet flow, which adds a turbulent flow and a shear layer to the flow system. We locate the reaction zone in these isothermal flows.

2.2.1 Location of the reaction zone in a jet issuing in still and coflowing air

For the following discussion, we assume, like Kuo[5], that the turbulent diffusion of fuel is equal to that of jet momentum, or, equivalently, that the Schmidt number is unity. In reality, the Schmidt number is about 0.8, but this does not change the following discussion fundamentally. With unity Schmidt number, the spread of the radial axial velocity profile is equal to that of the mixture fraction, and the decrease of the centerline values is also equal. In other words, the average mixture fraction in an isothermal jet flow is equal to the average axial velocity normalized to the average exit velocity. When $\xi$ is the average mixture fraction, $\xi_{cl}$ the average centerline mixture fraction, $U$ the average axial velocity, $U_{cl}$ the average axial centerline velocity and $U_j$ the average jet exit velocity, the mixture fraction for the isothermal case and for unity Schmidt number becomes according to Kuo[5]:

$$\frac{\xi}{\xi_{cl}} = \frac{U}{U_{cl}}, \quad \xi_{cl} = \frac{U_{cl}}{U_j}.$$  (2.1)

The entrainment of ambient fluid causes the shear layer to grow. The resulting jet spread corresponds to a decreasing centerline velocity. Rajaratnam[6] gives a phenomenological relation for the relation between the average axial centerline
velocity relative to the average jet exit velocity $U_j$ and the axial distance $x$ relative to the jet diameter $d_j$:

$$
\frac{U_{cl}}{U_j} = \frac{6.3}{x/d_j}.
$$

(2.2)

For a jet with velocity $U_j$ issuing in a coflow with velocity $U_c$, according to Rajaratnam[6], (2.2) can be replaced by:

$$
\frac{U_{cl} - U_c}{U_j - U_c} = \sqrt{\frac{U_j}{U_j - U_c}} \frac{6.3}{x/d_j} \approx \frac{6.3}{x/d_j}.
$$

(2.3)

Hence, the spread of a jet with velocity $U_j$ issuing in a coflow with velocity $U_c$ is like that of a jet with a velocity $U_j - U_c$ issuing in still fluid. A phenomenological relation of the radial dependence of the average axial velocity is given by Rajaratnam[6]:

$$
\frac{U - U_c}{U_{cl} - U_c} = \frac{1}{2} \left(1 + \cos \frac{\pi r}{2r_s}\right),
$$

(2.4)

where $r_s$ is the radial location of the inflection point in the radial profile of the axial velocity, where $(U - U_c)/U_{cl} = 0.5$; this is also the radial location with largest shear stresses. The width of the jet is proportional to the axial distance: $r_s/x = \text{constant}$. Rajaratnam gives relations for the spread of the shear layer between the central jet and the coflowing fluid, which is a function of the ratio $U_j/U_c$, and also for the boundaries of the shear layer. The radial location where the mixture fraction is stoichiometric ($\xi = \xi_{st} = 0.07$) is $r_{st}$. Because $r_s$ is related to the centerline velocity and $r_{st}$ to the absolute value of the mixture fraction, the relation between $r_{st}$ and $r_s$ depends on the axial distance $x$. Replacing in (2.1) $U$ and $U_{cl}$ for $U - U_c$ and $U_{cl} - U_c$, respectively, and using (2.3) and (2.4), we find for the radial location of the reaction zone:

$$
r_{st} = \frac{2r_s}{\pi} \arccos \left( \frac{2\xi_{st}}{(U_{cl} - U_c)/(U_j - U_c) - 1} \right) = \frac{2r_s}{\pi} \arccos \left( \frac{x\xi_{st}}{3.15d} - 1 \right).
$$

(2.5)

The ratio $r_{st}/r_s$ decreases with $x$ from 1.6 at $x/d_j = 10$, to unity at $x/d_j = 45$ and to zero at $x/d_j = 90$. It should be noted that these numerical values are derived from not very accurate phenomenological relations of isothermal turbulent flows. The ratio $r_{st}/r_s$ decreases monotonically with $x$, and $r_s$ increases linearly with $x$. This gives a cigar-shaped contour of stoichiometric mixture fraction, shown in Figure 2.1. From these considerations, we can conclude that for small $x$, the reaction zone is located outside the surface of strongest shear stresses, close to the outer boundary of the shear layer. The value of $r_s$ increases faster with $x$ than $r_{st}$, so for increasing $x$, stoichiometric surface moves into the shear layer. Hence, the shear stresses at the reaction zone increase. From (2.5) it follows that
at approximately $x/d_j = 45$, $r_{st}/r_s$ equals unity, and the reaction zone is at the location with largest shear stresses.

If we consider the surface of stoichiometry in a flame instead of in an isothermal flow, we should also include density and viscosity as function of the location. However, our purpose is to qualitatively position the reaction zone in relation to the shear layers in our flame. We therefore neglect these density and viscosity effects.

We now show that a widening of the jet, this is an increase in $r_s$, for not very large axial distance, will also cause an increase of $r_{st}$. For a developed jet, the centerline excess velocity is inversely proportional to the value of $r_s$. For not too large $x$, $(U_{cl} - U_c)/(U_j - U_c)$ is still quite high, and an increase in $r_s$ results in an increase in $r_{st}$. For example, according to (2.3), at $x/d_j = 12.5$, $(U_{cl} - U_c)/(U_j - U_c) = 0.5$. A $20\%$ decrease in $(U_{cl} - U_c)/(U_j - U_c)$ from 0.5 to 0.4, gives a $20\%$ increase in $r_s$ and, according to (2.5), a $15\%$ increase in $r_{st}$. When the centerline excess velocity becomes smaller, for larger axial distance, a change in $r_s$ gives a much smaller change in the location of the reaction zone, $r_{st}$. For example, at $x/d_j = 50$, $(U_{cl} - U_c)/(U_j - U_c) = 0.125$. A $20\%$ decrease in $(U_{cl} - U_c)/(U_j - U_c)$ from 0.125 to 0.100, gives a decrease in $r_{st}$ of only $3\%$. A larger entrainment of air gives a decrease of the centerline mixture fraction, but also a widening of the jet. The first effect decreases $r_{st}$, the latter increases $r_{st}$, but the above examples show that for the large part of the flame, the net effect is an increase of $r_{st}$.

### 2.2.2 Three-flow system

We now discuss the sources of turbulence in our three flow system, with a fuel jet flow, a primary air annulus jet flow and a secondary air coflow. Following the case of a fuel jet issuing in coflowing air, we determined the location of the reaction zone by the mixing of fuel and primary air. The reaction zone has a shape qualitatively similar to that in the previous two-flow case. In the three-flow case, however, there are more sources of turbulence that interact with the reaction zone. Schematically, the three-flow system with the shear flows is shown in Figure 2.1. A detailed discussion of the burner configuration can be found in Chapter 4. Basically, the burner consists of a fuel jet, 6 mm in diameter and of an annulus with an inner diameter of 15 mm and an outer diameter of 45 mm. Pilot flames are close to the fuel exit on a ring 7 mm in diameter, located on the burner rim. Fuel jet velocities ($U_j$) of the flames studied varied from 11.0 to 50.4 m/s, primary air velocities ($U_a$) from 2.2 to 20.4 m/s. (In these velocities, the acceleration due to preheating of the flows is included.) In all flames, the secondary air coflow was 0.3 m/s. The flame that is used as a standard flame, was operated with an average fuel exit velocity of 21.9 m/s, an average primary
2.2. Reaction zone and shear flows

Figure 2.1: Schematic and time average representation of the shear layers in the fuel jet/primary air annulus jet/secondary air coflow system. $U_j$, $U_a$ and $U_c$ are the average exit velocity of central jet, annulus and coflow respectively. The broken lines indicate the boundaries of the shear layers of the central fuel jet and of the annulus jet, obtained from relations given by Rajaratnam[6], the solid line indicates the location of the reaction zone, given by (2.5). The length of the velocity vectors give the ratio of the average exit velocities; the scale in radial and axial direction is equal. Region denoted (a) is in the shear layer of the fuel jet but outside that of the annulus jet, region (b) inside both shear layers and region (c) inside the annulus jet shear layer, but outside the fuel jet shear layer.

air exit velocity of 4.4 m/s (this is flame III of Table 4.2).

2.2.3 Shear layers in the three-flow system

First we discuss the sources of turbulence and after that we analyze which of them interact with the reaction zone. The shear layers in the three-flow system are shown in Figure 2.1, where the broken lines indicate the boundaries of the shear layers. The solid line indicates the surface of stoichiometric mixture fraction. Close to the burner nozzle, there is a potential core in the fuel jet flow, where there is no influence of shear layers between fuel flow and primary air flow or between primary air flow and secondary air coflow. No air is entrained in the fuel inside the potential core. The turbulence in this potential core is from the turbulent pipe flow only. The potential core is shown in Figure 2.1, but is very small. In the "standard flame" the potential core extends to approximately $x = 30$ mm, or $x/d_j = 5$. Right behind the burner rim, there is wake turbulence. The
shear layer between the fuel flow and the primary air flow is an important source of turbulence in a turbulent jet, because of the large velocity difference. We have seen before, however, that close to the nozzle, the reaction zone is located so close to the outer boundary of this shear layer, that turbulence originating in this shear layer does not strongly affect the reaction zone. Part of the fuel jet is inside the potential core of the primary air annulus jet flow. In this region, which is region (a) in Figure 2.1, the fuel issues in an air flow with a turbulence originating in the burner annulus. Outside the potential core of the annulus jet flow, there is a region with both the shear layer mixing fuel and primary air, and the one mixing primary and secondary air. This is region (b) in Figure 2.1. The part of the shear layer from the annulus jet flow outside the shear layer from the fuel jet flow, region (c) in Figure 2.1, is of minor importance, because on average, the reaction zone is not in this region.

2.2.4 Location of the reaction zone in the three-flow system

The average location of the reaction zone, given by (2.5), is shown as a solid line in Figure 2.1. The fact that the reaction zone in a natural gas flame is at a (low) mixture fraction of 0.07, makes that for small \( x \) the reaction zone is close to the outer boundary of the shear layer of the fuel jet, where shear stresses are small. Consequently, close to the nozzle, the turbulence in the vicinity of the reaction zone is not from the shear layer but from the turbulent annulus flow and from the wake behind the burner rim. The location of the reaction zone makes that the turbulence of the primary air flow is of great influence. For increasing \( x \), the reaction zone separates from the outer boundary of the central shear layer and enters the region with larger shear stresses. In addition, the influence of the shear layer between primary and secondary air is noticed from a distance of approximately 80 mm, or \( x/d_j = 133 \).

We carried out experiments in flames with primary air velocity large compared to the fuel jet velocity. When the primary air exit velocity is increased, the difference of the average fuel velocity and the average primary air velocity is decreased. In the case of low-turbulence fuel and primary air flows, the decrease in the initial velocity difference would decrease the shear stresses, resulting in less intense turbulent mixing of fuel and primary air. In our case, however, the fuel and primary air flows are turbulent at the burner exit, and despite the smaller average velocity difference, the local instantaneous shear stresses need not be smaller because of the higher level of turbulence in the primary air flow. In addition, the increased primary air velocity will increase the shear stresses between the primary and secondary air, giving a faster mixing of secondary air with the fuel jet for larger axial distance.
2.2. Reaction zone and shear flows

2.2.5 Flame length

For a jet issuing in still air, the relation between nonpremixed flame length and fuel flow rate is given by several authors, see for example Kuo[5] or Glassman[7]. In our three-flow system, the primary air flow also has the character of a turbulent jet and affects the flame length, as will be shown in the following.

In the case of a two-flow system and a laminar nonpremixed flame, the flame length is determined by equating the convection time necessary to bring a fuel element to the flame tip and the diffusion time necessary to bring an oxidizer element to the jet axis. It is found that the flame length only depends on the jet diameter, \( d_j \), fuel jet exit velocity, \( U_j \), and the molecular diffusion coefficient, \( \mathcal{D} \), see Kuo[5]:

\[
L_f \propto \frac{d_j^2 U_j}{\mathcal{D}} \propto \frac{\phi_{vj}}{\mathcal{D}},
\]

(2.6)

where \( \phi_{vj} \) is the volumetric fuel jet flow rate. When the Schmidt number \( Sc = \nu / \mathcal{D} \) is constant, see Kuo[5], \( \mathcal{D} \) in (2.6) can be replaced by the viscosity \( \nu \).

In the case of a turbulent flame, the viscosity \( \nu \) is replaced by the turbulent eddy viscosity, \( \nu_t \). The turbulent eddy viscosity is proportional to a macroscopic length scale of turbulence, which on its turn is proportional to the jet diameter, and to the root mean square value of the velocity fluctuations, which is proportional to the jet exit velocity. Substitution of the turbulent eddy viscosity with this proportionality in (2.6), gives a length of a turbulent nonpremixed flame proportional only to the jet diameter. Hence, turbulent flame lengths are independent of fuel jet flow rates or Reynolds numbers. The turbulent flame length appears to be smaller than the maximum laminar flame length[5].

In our three-flow system, the turbulent primary air flow is of great influence on the turbulent eddy viscosity. This turbulent eddy viscosity is therefore in the order of magnitude of \( U_a d_a \), where \( d_a \) is the hydraulic diameter of the annulus and \( U_a \) the annulus exit velocity. Equating the time for an air element to reach the jet axis to the time for a fuel element to reach the flame tip, yields a flame length in the three-flow system:

\[
L_f \propto \frac{d_j^2 U_j}{\nu_t} \propto \frac{U_j d_j}{U_a d_a}.
\]

(2.7)

Increased primary air flow rate increases turbulent diffusion of the air towards the fuel and therefore decreases the flame length.

In the case of a preheated jet, the density of the issuing fluid is changed. A jet with a density different from the density of the environment has a different development, see Pitts[8], and can be interpreted as if the flow is from a tube with different exit diameter, see Thring and Newby[9]. When our fuel and primary air flows are preheated, fuel jet flow experiences an "ambient" flow of
preheated primary air, so the fuel jet development is not changed by preheating. The preheated primary air flow, however, has a density different from the cold secondary air. In (2.7), the hydraulic diameter of the primary air annulus, \( d_a \), has to be replaced by an effective annulus hydraulic diameter, \( d_a^* \), given by:

\[
 d_a^* = \sqrt{\frac{\rho_a}{\rho_c}} d_a,
\]

(2.8)

where \( \rho_a \) is the density of the preheated primary air from the annulus, and \( \rho_c \) the density of the cold secondary air.

### 2.3 Interaction between reactions and turbulence in a jet flame

In the previous section the average location of the reaction zone was discussed in relation to the different sources of turbulence. We now investigate the interaction between turbulence and chemistry on a more detailed scale. First, we discuss the local effect of turbulence on combustion. After that, we discuss the effect of combustion on turbulence through the effect of temperature on the density and viscosity. Finally, we focus on the relation between turbulence and finite-rate chemistry involving OH.

#### 2.3.1 Local influence of turbulence on the reaction zone

We have shown in Section 2.2 that close to the nozzle, the average location of the reaction zone is close to the outer boundary of the fuel jet shear layer and that in our case of a three-flow system turbulent annulus flow influences the reaction zone, see Figure 2.1.

For a two-flow system of a fuel jet in coflowing air, both Chen and Goss[10] and Roquemore et al.[11] visualized the central fuel jet and the reaction zone, using reactive Mie scattering (RMS) and laser induced fluorescence (LIF) of OH[10], and reactive Mie scattering and flame luminosity detection[11], respectively. In both studies methane flames are used, with a stoichiometric mixture fraction (0.055) different from that of natural gas (0.07). Jet diameters \( d_j \) are 5 mm[10] and 10 mm[11]. The average fuel exit velocities \( (U_j = 29 \text{ m/s}[10], \text{ and } U_j < 17 \text{ m/s}[11]) \), and exit Reynolds numbers \( (Re_j \approx 10^4[10] \text{ and } Re_j < 1.3 \times 10^4[11]) \) are comparable with those in our experiments, see Table 4.2. The 17 m/s velocity flame presented by Roquemore et al.[11] was lifted and is therefore different from the attached flames close to the nozzle and probably also for larger \( x \).

Chen and Goss[10] show that the OH structure is not wrapped around large-scale eddies for \( x/d_j < 60[10] \) but that only some of the turbulent eddies displace the OH structure. Displacement of the reaction zone is accompanied by
2.3. Interaction between reactions and turbulence in a jet flame

stretching of the reaction zone. Most of the turbulent eddies identified by Chen and Goss in the fuel flow appear not to affect the OH structure, because the separation between these eddies and the OH zone, and therefore between the eddies and the reaction zone, is too large. The flame surface fluctuates at a frequency lower than that of vortex passage\cite{10}. With increased fuel jet velocity, more eddies displace and stretch the OH zone and the probability increases that eddies penetrate this zone. This penetration results in local extinction evidenced by holes in the OH zone\cite{10} or by holes in the luminous flame zone\cite{11}. Roquemore \textit{et al.}\cite{11} show that in the attached flames ($U_j < 12$ m/s) vortices in the fuel flow have developed from coherent vortices close to the nozzle into small-scale, unorganized vortex structures more downstream, and that the flame surface is not convoluted by these vortices, but only penetrated by some of them.

Velocities and temperatures have been measured by Sislian\cite{12} in a jet diffusion flame of methane and argon in still air ($\xi_{st} = 0.079$, $U_j = 17.2$ m/s, $Re_j = 7.85 \times 10^3$, $d_j = 10$ mm). They show that especially for small $x$, the radial location of maximum turbulent kinetic energy is significantly smaller than the radial distance of maximum average temperature. With increasing axial distance, the maximum become wider and the high temperature regions overlap with the regions of large turbulent kinetic energy.

To stimulate the interaction between OH concentration and turbulence, Stärner and coworkers (see for example Stärner \textit{et al.}\cite{13}) diluted the fuel by nitrogen and thereby increased the stoichiometric mixture fraction up to about 0.5 to have the reaction zone inside the shear layer and for small $x$ have it coincide with the region of largest shear stresses.

2.3.2 Influence of combustion on the turbulent flow

We now observe the influence of combustion on turbulence. The combustion influences turbulence through increased viscosity and decreased density in the regions of the flame with high temperature. Decrease of the level of turbulence in the flame is due to the increased viscosity, especially near the reaction zone where temperatures are highest. This effect is discussed by Takagi \textit{et al.}\cite{14}, Sislian \textit{et al.}\cite{12} and Roquemore \textit{et al.}\cite{11}.

Sislian \textit{et al.}\cite{12} have carried out carefully designed experiments, comparing isothermal jet flows (with a mixture of helium and air issuing in air) and jet flames (with a mixture of methane and argon issuing in air), where the exit density and viscosity of the flame jet flow was 1.0 and 1.3 times the density and viscosity of the isothermal jet flow, respectively. The exit Reynolds numbers of the flame jet flow and of the isothermal jet flow were 7.85 and $10.2 \times 10^3$, respectively. They used laser Doppler anemometry (LDA) to measure velocities
and properties of turbulence like turbulent kinetic energy and normal and shear stresses, and thermocouples to measure average temperatures.

Close to the nozzle, at $x/d_j = 5$, Sislian et al.[12] found that at $r = 0$ $\sqrt{k}/\langle u \rangle = 1.1$ % in the flame, and $\sqrt{k}/\langle u \rangle = 9.1$ % in the isothermal jet, where $k$ is the turbulent kinetic energy and $\langle u \rangle$ the average axial velocity. We thus see, that close to the nozzle and in the central portion of the jet, the turbulence is very low, compared to that in the isothermal jet. Also close to the nozzle, the off-axis maximum turbulent kinetic energy in the flame is about half that of the isothermal flow. The increased viscosity near the reaction zone prevents mixing of the jet flame with ambient air[11,12]. In the flame, the flows are accelerated by thermal expansion (decreased density). For $x/d_j > 15$, the turbulent kinetic energy of the flame, relative to the average axial velocity, becomes larger than the relative turbulent kinetic energy in the isothermal flow. The increase in turbulence due to the larger velocity gradients in the flame outweighs the decreased turbulence due to larger viscosity in the flame for larger axial distance[12].

Roquemore et al.[11] found that natural convection along the hot flame surface induces an extra instability. In our three-flow system, this natural convection flow is overruled by the forced convection flow from the annulus, so this kind of instability is not expected in our type of flames.

### 2.3.3 Role of OH in turbulent combustion

Both in hydrogen combustion and in hydrocarbon combustion, hydroxyl, or OH, is an important species, serving as a so-called chain carrier. Other chain carriers are O and H and they form a radical pool supplied by fast two-body reactions, see Williams[15] and Barlow et al.[16]. Fast reactions are chain branching reactions:

$$H + O_2 \rightleftharpoons OH + O,$$

and

$$O + H_2 \rightleftharpoons OH + H,$$

and chain propagating reactions, like:

$$OH + H_2 \rightleftharpoons H_2O + H,$$

$$H + HO_2 \rightleftharpoons OH + OH,$$

and

$$CH_4 + OH \rightleftharpoons CH_3 + H_2O.$$

The fact that the two-body reactions are fast is used in the so-called partial equilibrium models, where only the fast two-body reactions are assumed to be in equilibrium, see for example Correa and Gulati[17]. In the chain branching reactions, more radicals are produced than consumed, in the propagating reactions
the number of radicals remains equal. Chain branching and chain propagating reactions therefore do not deplete the radical pool.

On the other hand, chain terminating or recombination reactions do deplete the radical pool. These reactions do not have radicals as products. An example is the reaction between two CH₃ radicals that produce ethane. Chain termination involving OH is usually by slow, three-body reactions, like[16]:

\[
H + OH + M = H₂O + M.
\]

These recombination reactions are rather slow, because of the low probability of a collision between two reactants and an inert species at the same instant.

Relating the radical concentrations to the mixture fraction, the radicals have maximum concentrations near or at stoichiometry, where the temperature is highest. A model that is often used to describe the reaction zone in a turbulent nonpremixed flame, is the laminar flamelet model developed by Peters, see for example Peters[18]. In this model, the flame is locally observed as a laminar counterflow diffusion flame, of which the location displaced and stretched by the turbulence field. The model usually involves complete chemistry. This model predicts that OH peaks very close to but at the lean side of stoichiometry, see Barlow et al.[16]. Other radicals like H and CH₃ have a maximum mole fraction at the rich side of stoichiometry, O radicals at the lean side.

2.3.4 Damköhler number of chemical reactions involving OH

The parameter used to express the interaction between turbulence and chemistry is the Damköhler number, \( Da \). It is equal to the ratio of a time scale of turbulence, \( \tau_t \), and one of chemical reactions, \( \tau_{ch} \), see for example Barlow et al.[16]:

\[
Da = \frac{\tau_t}{\tau_{ch}}. \tag{2.9}
\]

Different choices can be made for \( \tau_t \) and the different chemical reactions have different time scales \( \tau_{ch} \). When \( Da \gg 1 \), the chemistry is so fast that it is not influenced by turbulence, when \( Da \ll 1 \), there is interaction between turbulence and chemistry. When reactions are fast and \( Da \) is large, the reactions take place in narrow regions and a turbulent flame can be modelled with strained laminar diffusion flames, or flamelets, that follow instantaneously the stoichiometric mixture fraction in the flame and that are stretched by the turbulent eddies. When chemical reactions are slow compared to turbulent mixing, \( Da \) is low and there is time for the turbulence to mix the reactants and products. In this case, according to Borghi[19] the reaction zone consists of (partially) premixed flamelets. As was shown in Section 2.3.3, OH takes part in both fast, two-body, reactions and in slow, three-body reactions. Because of the largely different chemistry time scales, (at least) two different Damköhler numbers are necessary, \( Da_2 \) for the
fast two-body, chain branching and chain propagating reactions, and $Da_3$ for the slow three-body, recombination reactions. In an atmospheric pressure flame, the chemistry time scales of the three-body reactions are two orders of magnitude larger than those of the two-body reactions[16,20], so the reaction zones of the two-body reactions may be thin compared to the turbulent length scales, whereas the three-body reaction zones are thick, resulting in distributed reaction zones of three-body reactions[17]. In our burner there is a facility to preheat fuel and air. This will decrease chemistry time scales, both of the two-body reactions and of the three-body reactions.

Barlow et al.[16] define the turbulence time scale as the large-eddy turnover time:

$$\tau_t = \frac{w}{U}, \quad (2.10)$$

where $w$ is the width of the jet at half the centerline velocity, $U$ is a characteristic jet excess velocity. Another, more locally defined, turbulence time scale is suggested by Pope[20], derived from the scalar dissipation rate, $\chi$. The scalar dissipation rate is proportional to the local strain rate in the flow, see Peters[18], which is a parameter of turbulence. A turbulence time scale can be defined as:

$$\tau_t = \frac{1}{\chi}, \quad (2.11)$$

where $\chi$ is the scalar dissipation rate, related to the mixture fraction $\xi$ and the diffusion coefficient $\mathcal{D}$ as:

$$\chi = \mathcal{D}(\nabla \xi)^2. \quad (2.12)$$

Effects of finite-rate chemistry are local flame extinction and concentrations that do not reach equilibrium, can be related to the Damköhler numbers $Da_2$ and $Da_3$, respectively. This is discussed in the following sections.

### 2.3.5 Local extinction

According to Correa and Gulati[17] local extinction occurs when the two-body, chain branching and propagating reactions are affected by turbulence or, equivalently, when $Da_2$ in the order of unity. The lower the Damköhler number, the stronger the interaction of turbulence with the two-body reactions. When eddies stretch the reaction zone, it becomes narrower and the heat production becomes smaller relative to the heat losses. Consequently, temperature decreases, and when temperature is lower than a certain value, the flame extinguishes. Peters[18] states that this moment of extinction corresponds to a critical value of the scalar dissipation rate, given by (2.12). This scalar dissipation rate is proportional to the local velocity gradient or strain rate[18].

Assuming that a laminar flamelet model can be used in the part of our nonpremixed flames close to the nozzle, the chemistry time scale of the fast two-body reactions in this region can be estimated from the strain rate at which
2.3. Interaction between reactions and turbulence in a jet flame

A laminar counterflow diffusion flame extinguishes. The effect of preheating the flows on local extinction can also be estimated by investigating the extinction of a preheated laminar diffusion flame.

Flame extinction occurs when \( Da_2 \approx 1 \), so at local extinction the two-body chemistry time scale is comparable to the turbulence time scale. The inverse of the strain rate is a time scale characterizing the flow field and in a turbulent flame this time scale can be used as a turbulence time scale. It follows from \( Da_2 \approx 1 \) that the time scale of the two-body chemistry is in the same order of magnitude as the time scale of the flow field at extinction of the laminar counterflow diffusion flame. From numerical simulations of a strained laminar diffusion flame, the strain rate, or scalar dissipation rate, at which the flame extinguishes, can be calculated. The strain rate, \( a \), of the laminar diffusion flame is defined as the absolute value of the velocity gradient perpendicular to the reaction zone, at a location just to the lean side of the reaction zone. See for the configuration of the fuel flowing from a porous cylinder in a direction opposed to a parallel air flow Chelliah et al.[21].

Barlow et al.[16] found in a hydrogen/air flame a two-body chemistry time scale which was about 3 times smaller than the inverse of the strain rate at flamelet extinction, so the time scales are in the same order of magnitude. Numerical simulation of strained laminar diffusion flames, carried out by Peeters[3], showed that the flame extinguishes at a critical strain rate of about 400 \( s^{-1} \) when fuel (methane) and oxidizer (air) are at room temperature at the flow exits. This critical strain rate corresponds to a scalar dissipation rate at stoichiometry of 15 \( s^{-1} \). By preheating the fuel and air to 673 K, the flamelet extinguishes at a much larger strain rate of 1975 \( s^{-1} \), corresponding to a scalar dissipation rate of about 75 \( s^{-1} \). The velocity gradients in the preheated flame therefore must be about 5 times larger to cause local extinction. The chemistry time scale of two-body reactions is consequently 5 times smaller when the flows are preheated to 673 K. Two-body chemistry time scales (defined as the inverse of the strain rate) of the initially cold and of the preheated natural gas flames are thus in the order of magnitude of 2.5 ms and 500 \( \mu s \), respectively. Influence of flow temperature on laminar flamelets is also reported by Darabiha and Candel[22], who preheated the air to a temperature of 1000 K. For a hydrogen/air flame, Barlow et al.[16] found a critical strain rate of about 15000 \( s^{-1} \) and a two-body chemistry time scale of 20 \( \mu s \) for hydrogen/air combustion. Using (2.10), the turbulence time scales in the flames we studied are in the order of magnitude of 1 ms at \( x = 50 \) mm, so local extinction is expected in some of these flames.

With increasing \( x \), according to (2.10) the turbulence time scales increase and so do \( Da_2 \) and \( Da_3 \). This increasing turbulence time scale also follows from (2.11), for the scalar dissipation rate decreases with \( x \). On the other hand, the fuel and air will be mixed with hot combustion products, increasing the temperatures and decreasing the chemistry time scales. Hence, the Damköhler numbers are expected to increase with \( x \). Dilution of the reactants with combustion products
will increase the chemistry time scales, but this effect is expected to be of minor importance. Chen and Goss[10] found an increasing probability of holes in the OH structure with increasing $z^\dagger$. According to Williams[15], it is not clear yet whether the extinct flamelets re-ignite and, if so, in what way. Borghi[19] suggests local extinction may be followed by mixing of products, fuel and air and later by re-ignition of a partially premixed flamelet.

2.3.6 Superequilibrium

The OH concentration will be larger than that at thermodynamical equilibrium (OH concentration is superequilibrium), due to fast production of OH and the slow recombination of the radicals, where "fast" and "slow" are relative to the turbulence time scales[16]. The level of superequilibrium can be defined as the ratio of the actual instantaneous concentration and the equilibrium concentration at the same value of the mixture fraction in an adiabatic flame[16]. Measurements of Barlow et al.[16] were conditional on the mixture fraction, so they were able to measure the level of superequilibrium as a function of the mixture fraction. Drake et al.[23] measured distribution functions of OH concentration fluctuations and of the mixture fraction fluctuations separately. From every mixture fraction value, the equilibrium OH concentration was calculated, yielding distribution functions of equilibrium OH concentration fluctuations. They defined the level of superequilibrium on the maximum instantaneous values, so this was conditional on the stoichiometric mixture fraction. Drake et al. also defined an average level of superequilibrium, by comparing the average OH concentration at a certain location with the average equilibrium value obtained from the distribution functions of the mixture fraction fluctuations.

The level of superequilibrium depends on the time scale of three-body recombination reactions in relation to the time scale of turbulence. According to Correa and Gulati[17], high levels of radical concentration superequilibrium are found when the recombination reactions are slow compared to the turbulence time scales, or equivalently, when the Damköhler number of the recombining reactions, $Da_3$ is not much larger than unity. With increasing axial distance, the turbulence time scale increases and chemistry time scales decreases due to the premixing with hot combustion products, so $Da_3$ decreases. This results in a decreased level of superequilibrium with increasing axial distance. Drake et al.[23] have shown for a nonpremixed hydrogen/air flame, that the maximum instantaneous concentration remained constant with $x$ in the largest Reynolds number flame ($Re_j = 8.5 \times 10^3$), whereas in the flame with $Re_j = 1.6 \times 10^3$ the maximum instantaneous concentration decreased with a factor of about 2 from $x/d_1 = 25$ to 200. Hence, the instantaneous level of superequilibrium decreased

---

† This probability was defined as the number of one-dimensional OH images that showed extinction and was therefore weighed with the size of the holes. A decreasing number of holes of increasing size may give increasing probability, so the increasing hole-probability is not necessarily inconsistent with the above reasoning.
with \( x \) in the flame with \( \text{Re}_j = 1.6 \times 10^3 \) and lower, but remained constant with \( x \) in the \( \text{Re}_j = 8.5 \times 10^3 \) flame. Apparently, in the larger Reynolds number flame, the strain rates remain so large at large \( x \), that instantaneous concentrations are never at equilibrium. The average level of superequilibrium, however, decreases with axial distance and becomes unity, because at the radial location of maximum average OH concentration the larger concentrations become less probable with increasing \( x \) due to intermittency.

Three-body recombination reactions are slower than two-body chain branching reactions, so the Damköhler number of the recombination reactions \( (Da_3) \) is smaller than that of the chain branching reactions \( (Da_2) \). In a flame with local extinction, \( Da_2 \) is near unity and consequently, \( Da_3 \) is in the order of magnitude of unity or smaller. One will therefore always find superequilibrium radical concentrations in flames with extinction. The lower \( Da_2 \), the lower \( Da_3 \) needs to be, so stronger extinction implies a higher level of superequilibrium. This implication, however, is based on global parameters and not on local, instantaneous properties.

### 2.4 Relation between concentration, mass- and mole fraction

In the discussion of the concentration measurements, we sometimes need to compare our concentration measurements with reported mass- or mole fractions. It is therefore necessary to give the relations between these quantities.

The concentration of the species \( s \) is related to the mole fraction \( X_s \) or the mass fraction \( Y_s \) by:

\[
c_s = \frac{N_A X_s \rho}{M} = \frac{N_A Y_s \rho}{M_s}, \tag{2.13}
\]

where \( M \) is the local mole mass and \( M_s \) is the mole mass of species \( s \). \( M \) is related to \( M_s \) by

\[
\frac{X_s}{M} = \frac{Y_s}{M_s}, \tag{2.14}
\]

or, because \( \sum_s X_s = 1 \):

\[
M = \frac{1}{\sum_s Y_s / M_s}. \tag{2.15}
\]

Using the ideal gas law, the density can be expressed in pressure and temperature, by:

\[
\rho = \frac{M_p}{RT}, \tag{2.16}
\]

where \( p \) is the pressure, \( T \) the temperature and \( R \) the molar gas constant. Turbulent mixing of one fluid with another, both with different \( M_s \), makes that
\( M \) depends on the local instantaneous state of mixing. Relation (2.13) can be rewritten:

\[
c_s = \frac{N_{AP}}{RT} X_s. \tag{2.17}
\]

When there are fluctuations in density, (2.13) becomes:

\[
\langle c_s \rangle = \frac{N_A}{M_s} \langle \rho Y_s \rangle. \tag{2.18}
\]

It should be noted that:

\[
\langle \rho Y_s \rangle < \langle \rho \rangle \langle Y_s \rangle. \tag{2.19}
\]

When there are fluctuations in density, but not in pressure or temperature, the ratio between concentration and mole fraction is constant, and the average concentration is:

\[
\langle c_s \rangle = \frac{N_{AP}}{RT} \langle X_s \rangle, \tag{2.20}
\]

so the average concentration is proportional to the average mole fraction.
Chapter 3

Laser induced fluorescence

3.1 Introduction

Measurement of short living reaction intermediates, like radicals, requires a measuring technique that is both local and instantaneous. This is also essential when turbulent fluctuations are to be resolved. Any technique that changes the environment of the reaction intermediates also affects the reactions which determine the amount of the species under investigation, and thus affects the amount itself. For example, with a suction probe a sample with the species investigated is carried to a measuring device, and consequently the temperature of the sample is changed.

We thus see that physical probes usually are not the proper means for measurements of (turbulent fluctuations of) reacting species. Laser techniques provide nonintrusive, fast response measurements, where several physical phenomena can be used to measure different quantities. The Doppler effect and Mie scattering is used for velocity measurements (laser Doppler anemometry or LDA), Raman scattering is used for concentration measurements of major species (spontaneous Raman spectroscopy) and for temperature measurements (coherent anti-Stokes Raman spectroscopy or CARS), fluorescence is used for concentration measurements of short-living species with small concentrations (laser induced fluorescence spectroscopy or LIFS). These techniques are still being developed and refined to cope with problems specific for these laser-diagnostic techniques, see Eckbreth[24].

The measuring volume is usually determined by the size of the focussed laser beam, which is in the order of magnitude of 0.5 mm. The response time depends on the technique, but can be as short as 1 ns. It is assumed that the laser light does not change temperature or chemistry, which is the case in most applications. Species selective concentration measurements are possible, by using absorption and emission of laser light (LIFS) or spectral selective scattering (Raman scattering). Many of these techniques also allow for instantaneous one- or two-dimensional measurements, with which spatial structures become apparent.

Reaction intermediates exist in small amounts, and a sensitive technique like laser induced fluorescence spectroscopy, is often the only method to carry
out instantaneous and spatially resolved measurements. The most difficult part of laser induced fluorescence measurements is relating the signal to the concentration, as will be discussed theoretically in this chapter. In spite of the rather large systematic error associated with the method, instantaneous one- and two-dimensional detection of species using laser induced fluorescence, is a very powerful tool when it comes to understanding turbulent combustion; not for all purposes absolute and accurate concentration measurements, which are generally zero-dimensional, are essential. The significance of so-called semi-quantitative concentration measurements is outlined by Crosley[25].

The experiments discussed in this thesis are based on fluorescence. According to Eckbreth[24], in this context fluorescence is to be considered as "the sequence of an optical absorption process followed by a spontaneous emission event". After photon absorption, the molecule is brought into an excited electronic state; in the spontaneous emission process, the molecule returns to the electronic ground state again. For fluorescence, the states before and after spontaneous emission should have the same electronic spin. If this is not the case, the process is called phosphorescence, with life times (in the order of magnitude of 100 µs) much longer than those of fluorescence (ranging from about 100 ps to 10 µs)[24]. In the spontaneous emission process, transitions are allowed to energy levels other than the level occupied before absorption. As a result, the emitted light has more than one spectral component and light is emitted with wavelengths different from the wavelength of light absorbed. An example from every-day life, is fluorescent ink, that absorbs light in the ultraviolet range and emits light in the visible range, making the ink very bright.

For a discussion of fluorescence, we need to discuss very briefly spectroscopy of the molecules investigated and the energy transfer processes that occur. To suppress the influence of collisional energy transfer processes that interfere with fluorescence, spectral selective detection is required. However, a spectroscope, necessary for this spectral selective detection, cannot be combined with one- or two-dimensional detection, which is necessary to reveal the spatial flame structure. Therefore, we investigate in this chapter the difference between single emission-line detection and broadband detection. For this purpose, we study the rates of the collisional energy transfer processes that are of influence. A less detailed discussion of this influence can be found in De Vries et al.[26]. We conclude with an estimate of the errors that arise from broadband detection.

3.2 Spectroscopy

Study of fluorescence is to be accompanied by detailed knowledge of spectroscopy. In this study, the emphasis is on two-atom molecules, OH and NO. For theoretical study of spectroscopy of two-atom molecules, we refer to Herzberg[27], for an overview of this theory to Eckbreth[24] and for molecular constants of many two-
atom molecules to Huber and Herzberg[28]. Detailed information of the rotational spectra of OH and NO is given by Dieke and Crosswhite[29] and by Engleman et al.[30], respectively. Throughout the thesis, we follow the notation of Dieke and Crosswhite[29], which is also used by Engleman et al.[30]. For the moment, it is sufficient to know that the energy of a two-atom molecule consists of electronic energy states, vibrational energy states and rotational energy states:

\[ E_{\text{tot}}(e, v, J) = E_{\text{el}}(e) + E_{\text{vib}}(v) + E_{\text{rot}}(J), \]  

(3.1)

where \( E_{\text{tot}} \) is the total energy of the molecule, \( E_{\text{el}}(e) \) the electronic energy, \( E_{\text{vib}}(v) \) the vibrational energy and \( E_{\text{rot}}(J) \) the rotational energy. The electronic ground state is designated by \( e = 0 \), the first excited state with \( e = 1 \), etcetera, \( v \) is the vibrational quantum number, \( J \) the rotational quantum number. The energy of the electronic ground state is zero by definition. The energy is usually expressed as wavenumber \( \nu \) (unity generally cm\(^{-1}\)):

\[ E = h \nu, \]  

(3.2)

where the wavenumber is the reciprocal of the wavelength: \( \nu = \lambda^{-1} \). Analogous to (3.1), we can define \( \nu_{\text{el}}, \nu_{\text{vib}} \) and \( \nu_{\text{rot}} \).

According to Herzberg[27], the vibrational state can be described by:

\[ E_{\text{vib}}(v) = h c \left( \omega_e(v + \frac{1}{2}) - \omega_e x_e(v + \frac{1}{2})^2 + \ldots \right), \]  

(3.3)

where \( \omega_e \) and \( \omega_e x_e \) are coefficients which depend on the electronic state \( e \), and which can be found in Huber and Herzberg[28], \( v \) is the vibrational quantum number, \( h \) is the Planck constant and \( c \) the velocity of light. For many applications, the first order approximation of the vibrational energy is sufficiently accurate. For the electronic ground state we use \( v'' \), for the first excited state \( v' \).

The rotational energy states are given by[27]:

\[ E_{\text{rot}}(J) = h c \left( B_v J(J + 1) - D_v J^2(J + 1)^2 + \ldots \right), \]  

(3.4)

where the coefficients \( B_v \) and \( D_v \) depend on the vibrational level. The rotational energy levels are not equidistant. \( J \) is the rotational quantum number. For the electronic ground state, we use \( J'' \), for the first excited state we use \( J' \). The rotational energy is given by the length of the angular momentum vector. This angular momentum also has a direction, so every rotational state has a degeneracy \( g_{\text{rot}} \):

\[ g_{\text{rot}} = 2J + 1. \]  

(3.5)

The total angular momentum, giving the rotational quantum number \( J \), is the vector sum of the angular momenta of molecular rotation, electron orbital motion and electron spin. Coupling between these angular momenta is possible in different ways, with the so-called Hund's cases as extremes. The main effect of the
Table 3.1: Energy of the first excited electronic state, and coefficients of vibrational energy, $\hbar \omega_e$ (J) and of rotational energy $\hbar c B_v$ (J) (for $v = 0$), for both the electronic ground state and the first excited electronic state. Between brackets are the more commonly used values in cm$^{-1}$.

<table>
<thead>
<tr>
<th>species</th>
<th>electronic</th>
<th>vibrational</th>
<th>rotational</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$E_{el,1}$ (10$^{-19}$ J)</td>
<td>$\hbar \omega_e$ (10$^{-20}$ J)</td>
<td>$\hbar c B_v$ (10$^{-23}$ J)</td>
</tr>
<tr>
<td></td>
<td>ground state</td>
<td>1st excited state</td>
<td>ground state</td>
</tr>
<tr>
<td>OH</td>
<td>6.50 (32684)</td>
<td>7.43 (3737.8)</td>
<td>6.32 (3178.8)</td>
</tr>
<tr>
<td>NO</td>
<td>8.73 (43906)</td>
<td>3.79 (1904.4)</td>
<td>4.72 (2374.3)</td>
</tr>
</tbody>
</table>

Differences in coupling, is effect of the spin state on the energy, see Herzberg[27]. When this influence of spin state is large, there is no close relation between the energies with a given value of $J$ and different spin quantum numbers, and the energies of a specific spin state are grouped (Hund’s case a). When the influence of spin is small, the energy for a given value of $J$ is only slightly corrected for different spin states (Hund’s case b). The coupling often depends on the value of $J$, like in the case of the electronic ground state of OH[29] and NO[30], so (3.4) is usually to be corrected, see Dieke and Crosswhite[29], Engleman et al.[30] and Reisel et al.[31]. Under some conditions, the different energies corresponding to the different spin states are almost independent of $J$ and can be included in the electronic energy[27]. See for details in the case of OH and NO, Dieke and Crosswhite[29] and Engleman et al.[30], respectively.

The electronic ground states of both OH and NO are called $X^2\Pi$, the first excited electronic states $A^2\Sigma$. $X$ is used for the electronic ground state, $A$ for the first excited electronic state which is allowed by spin conservation[24], $B$ for the second, etcetera. $\Pi$ and $\Sigma$ give the total electronic orbital angular momentum quantum number $\Lambda$: $\Sigma, \Pi, \Delta, \ldots$ for $\Lambda = 0, 1, 2, \ldots$. The superscript 2 indicates that two electronic spin states are possible: with electronic spin quantum numbers $\Sigma = \pm \frac{1}{2}$ and $-\frac{1}{2}$. To discriminate both electronic spin states, states are designated by $X^2\Pi_{1/2}$ and $X^2\Pi_{3/2}$, see Dieke and Crosswhite[29] and Reisel et al.[31].

In Table 3.1 we give for OH and NO, the energy level of the first excited state, and the coefficients $\omega_e$ and $B_v$ (for $v = 0$), which give the separation of the vibrational and rotational energies, respectively, both for the electronic ground state and the electronic excited state. The values are obtained from Huber and Herzberg[28]. We find that the energy difference between the electronic states is largest, and between the rotational states smallest. We also see that the rotational levels of NO are much closer than those of OH.
3.3 Boltzmann distribution

The molecules are in an energy state of which the statistics are determined by the Boltzmann distribution. The Boltzmann factor $F_B$ consists of three components: the distribution over the electronic states, over the vibrational states and over the rotational states. The Boltzmann factor is the fraction of molecules in a given electronic, vibrational and rotational state, and is given for example by Lucht et al.[32] by:

$$F_B = F_{B,el}F_{B,vib}F_{B,rot},$$  \hspace{1cm} (3.6)

where $F_{B,el}$ is the Boltzmann factor of the electronic state observed, $F_{B,vib}$ the Boltzmann factor of the vibrational state and $F_{B,rot}$ the Boltzmann factor of the rotational state.

In our case, we are not interested in the Boltzmann distribution of the excited electronic states. In the temperature range that is of our interest, the number of molecules in the excited electronic states is negligible. We therefore only need to pay attention to the energy levels corresponding to different electron spin states, which are a result of spin splitting and $\Lambda$-doubling of the electronic ground state, see Lucht et al.[32].

In the case of the $X^2\Pi$ states (ground states) of OH and NO this spin splitting can be included in the electronic energy. For NO this splitting is much larger compared to the rotational energy spacing, and therefore cannot be neglected. For NO the energy of the $^2\Pi_{3/2}$ state is larger than that of the $^2\Pi_{1/2}$ by the amount of $E_{\text{split}}$, for corresponding levels of $J$, see Herzberg[27]. Therefore, the Boltzmann factor of the $X^2\Pi_{1/2}$ and $X^2\Pi_{3/2}$ states are different:

$$F_{B,el,2} = \frac{g_{el,2}e^{-E_{\text{split}}/kT}}{g_{el,1} + g_{el,2}e^{-E_{\text{split}}/kT}},$$ \hspace{1cm} (3.7)

for the $X^2\Pi_{3/2}$ state, and

$$F_{B,el,1} = \frac{g_{el,1}}{g_{el,1} + g_{el,2}e^{-E_{\text{split}}/kT}},$$ \hspace{1cm} (3.8)

for the $X^2\Pi_{1/2}$ state. $T$ is the absolute temperature, $k$ the Boltzmann constant. For OH, the energy of the $^2\Pi_{3/2}$ state is smaller than that of the $^2\Pi_{1/2}$ state, and the indices 1 and 2 of (3.7) and (3.8) have to be exchanged. $g_{el,1}$ and $g_{el,2}$ are the electronic degeneracies of both spin states and are equal, see Lucht et al.[32].

The fraction in the vibrational state is given by:

$$F_{B,vib} = \frac{e^{-E_{\text{vib}}(v)/kT}}{\sum_v e^{-E_{\text{vib}}(v)/kT}},$$  \hspace{1cm} (3.9)

Because of the degeneracy of the rotational energy states, $F_{B,rot}$ has a maximum. For not too low temperatures and with the first order approximation
of the rotational energy, the summation in the denominator can be approximated by an integration, which results in [27]:

\[ F_{B,\text{rot}} = \frac{hcB_e}{kT}(2J + 1)e^{-E_{\text{rot}}(J)/kT}. \] (3.10)

The rotational level \( J \) for which the Boltzmann factor is least sensitive to the temperature follows from \( \partial F_{B}/\partial T = 0 \).

The energy \( E_{el}(1) \) is so large, that even at flame temperatures the fraction of molecules in the first excited state is negligible, and equivalently, they always return to the electronic ground state when they are excited to the first excited electronic state. The vibrational energy separation is much smaller than the electronic energy separation, and at higher temperatures, the levels with \( v = 1 \) and higher become populated; at room temperature only the lowest state is populated. The fraction of OH molecules in the states with \( v \geq 1 \) is still less than 7% at a temperature of 2000 K, but the fraction of NO molecules in these states is about 25% at 2000 K. For rotational distribution, the value of \( J \) with the largest population increases with temperature.

3.4 Quenching, vibrational and rotational relaxation

In addition to absorption, stimulated and spontaneous emission, three other kinds of energy transfer are important in this context: quenching, vibrational relaxation and rotational relaxation. These are all inelastic collisional processes. Excitation of molecules violates the Boltzmann distribution of the molecules over the different energy states, because the population of the initial level of the excitation process becomes smaller, and the of the final level becomes larger than according to the Boltzmann distribution. Rotational relaxation tends to re-establish the Boltzmann distribution among the rotational states, vibrational relaxation among the vibrational states and quenching among the electronic states.

In our research the OH and NO was excited from \( v'' = 0 \) in the electronic ground state to \( v' = 0 \) in the first excited state. Because the state with \( v = 0 \) has the largest population or, analogous, the largest probability to be populated, vibrational relaxation in the electronic excited state does not occur.

We therefore focus on quenching and rotational relaxation, which both affect the population of the energy level connected by the laser radiation. First, we designate two rotational energy levels in the excited electronic state by \( j \) and \( k \), and one in the electronic ground state by \( l \), see Figure 3.1. The rotational relaxation rate from level \( j \) to \( k \), \( R_{jk} \), and the quenching rate between \( k \) and \( l \), \( Q_{kl} \), are given by:

\[ R_{jk} = \sum_s N X_s(v)\sigma_{r,jk,s} \] (3.11)

and

\[ Q_{kl} = \sum_s N X_s(v)\sigma_{q,kl,s}. \] (3.12)
respectively, where \( s \) indicates the species by which the excited molecule is relaxed or quenched. \( \sigma_{i,jk,s} \) and \( \sigma_{a,kl,s} \) are the cross sections of rotational relaxation and quenching, which depend on the species \( s \) and the initial and final states. \( N \) is the total number of molecules per unit volume, related to the Avogadro number \( N_A \), the pressure \( p \), the gas constant \( R \) and the temperature \( T \):

\[
N = \frac{N_A p}{RT}.
\] (3.13)

The average molecular velocity \( \langle v \rangle \) is according to Lucht et al.\cite{33} given by:

\[
\langle v \rangle = \sqrt{\frac{8kT}{\pi \mu_{\text{red}}}},
\] (3.14)

where \( \mu_{\text{red}} \) is the reduced molecular mass of the species under investigation and the species it collides with. Substitution of (3.13) and (3.14) in (3.11) and (3.12) shows that \( Q_{kl} \) and \( R_{jk} \) are proportional to \( T^{-1/2} \).

### 3.5 Transitions during laser excitation

We observe the excitation of a species by a short, intense laser pulse. We designate the initial rotational energy level in the electronic ground state with \( i \), and the final level in the first excited state with \( j \). Level \( i \) is adjacent to other rotational levels of which one specific is called \( l \), and level \( j \) is adjacent to levels of which one specific is called \( k \), see Figure 3.1. The absorption rate is \( b_{ij} \), determined by the Einstein coefficient \( B_{ij} \) and the laser intensity per unit bandwidth, \( I_\nu \)\cite{24}:

\[
b_{ij} = B_{ij} I_\nu / c.
\] (3.15)

Due to absorption, the Boltzmann distribution in the electronic ground state is violated, and the population of level \( i \), \( N_i \), has become too small compared to that of adjacent levels, whereas that of level \( j \), \( N_j \), has become too large. Therefore, by rotational relaxation molecules in level \( j \) are transferred to other levels, to a specific level \( k \) with rate \( R_{jk} \); also by rotational relaxation molecules are transferred to level \( i \) from other levels, from a specific level \( l \) with a rate \( R_{li} \). Molecules are excited, so there is quenching from the directly populated level \( j \) and the other, indirectly populated levels, to levels in the electronic ground state. The quenching rate from level \( j \) or \( k \) to all levels in the electronic ground state, is \( \sum_j Q_{jl} \) and \( \sum_k Q_{kl} \), respectively. In addition, spontaneous emission from the directly and indirectly populated levels occurs, and stimulated emission from level \( j \) back to the initial level \( i \).

Eckbreth\cite{24} gives orders of magnitude of the spontaneous emission rates \( A \), and quenching rates \( Q \), at atmospheric pressure (summed over all transitions): for OH, \( A \approx 1.5 \times 10^6 \text{ s}^{-1} \) and \( Q \approx 6 \times 10^8 \text{ s}^{-1} \), and for NO, \( A \approx 3 \times 10^6 \text{ s}^{-1} \).
Figure 3.1: Transitions between rotational levels in the electronic ground state \((i\) and \(l\)) and the first excited electronic state \((j, k, p\) and \(q\)), where \(B\) designates absorption and stimulated emission, \(A\) spontaneous emission, \(Q\) quenching and \(R\) rotational relaxation.

and \(Q \approx 5 \times 10^9\) s\(^{-1}\). Quenching is much faster than spontaneous emission in atmospheric pressure flames, and dominates the downward transfer. Due to the large quenching rate, the actual lifetime of the fluorescence is about 1000 times smaller than in the case when there is no quenching. To a good approximation, we may therefore say that there is no fluorescence when laser excitation has ended. The rotational relaxation rates are in the order of magnitude of 10 times larger than the quenching rates, see Crosley[34] or Chan and Daily[35].

Rate equations can be constructed by equating the time derivative of the population of a certain level and the rates increasing and decreasing the population, see for example Eckbreth[24].

### 3.5.1 Balanced cross rate model

When the laser induced processes are sufficiently fast (i.e. the laser intensity is sufficiently high), the populations of the levels \(i\) and \(j\) quickly reach a dynamical equilibrium during the laser pulse, see Lucht et al.[36]. The balanced cross rate model, as proposed by Lucht et al. is that the rotational relaxation rate from the overpopulated level \(j\) is balanced by the rotational relaxation rate to the underpopulated level \(i\). As a consequence:

\[
N_i + N_j = N_i^0, \tag{3.16}
\]

where \(N_i^0\) is the population of level \(i\) before excitation, which is related to the total number of molecules through the Boltzmann factor. Using realistic values
for all parameters and experimentally determined laser pulse shape, Lucht et al.[36] found that the sum \( N_i + N_j \) varied with about 20%. If the balanced cross rate model holds, the population of level \( j \) is:

\[
N_j = N_i^0 \frac{B_{ij}}{B_{ij} + B_{ji}} \frac{1}{1 + I_{\nu, sat}/I_\nu}.
\]  

(3.17)

The saturation intensity \( I_{\nu, sat} \) depends on quenching and rotational relaxation, see for example Eckbreth[24] or Lucht et al.[36]. The population of level \( i \) before excitation, \( N_i^0 \), is related to the total number of molecules of the species \( s \) under investigation per volume unit, \( N_s \), by:

\[
N_s = N_i^0/F_{B,i},
\]  

(3.18)

where \( F_{B,i} \) is the Boltzmann factor of level \( i \). The quantity \( N_s \) is equivalent to the concentration of species \( s \), used in the following chapters, which is designated with \( c_s(\vec{r}, t) \).

3.5.2 Saturation

There are two limiting regimes of excitation: the linear regime and the saturated regime. The fluorescence is in the linear regime when the laser intensity is small. As follows from (3.17), with \( I_\nu/I_{\nu, sat} \ll 1 \), the population \( N_j \), and consequently the fluorescence signal, is proportional to the laser intensity, and inversely proportional to the quenching rate, see for example Eckbreth[24]. When the laser intensity is very large, \( I_\nu/I_{\nu, sat} \gg 1 \), the fluorescence is saturated. In that case, the laser-coupled processes, absorption and stimulated emission dominate the energy transfer and are so fast that the quenching does not significantly decrease the population of the level \( j \). When \( I_\nu/I_{\nu, sat} \gg 1 \), \( N_j \) becomes independent of quenching, rotational relaxation and laser intensity, as follows from (3.17):

\[
N_j = N_i^0 \frac{B_{ij}}{B_{ij} + B_{ji}}.
\]  

(3.19)

In the single emission-line detection scheme, together with saturated fluorescence and balanced cross rate, the multi-level model, as shown in Figure 3.1, reduces to a two-level model, consisting of levels \( i \) and \( j \), and the fluorescence signal becomes independent of rotational relaxation and quenching, see Lucht et al.[36]. In the case of broadband detection, however, the rotational relaxation from level \( j \) to other levels, and the quenching from indirectly populated levels, enters the relation between the fluorescence signal and the total number of molecules. This is shown in the following, where we assume balanced cross rate.

3.6 Fluorescence signal

The fluorescence signal \( S_{F, M} \) of a transition from a level \( k \) to a level \( l \) is proportional to the number of excited molecules in level \( k \), \( N_k \), the Einstein coefficient
of spontaneous emission $A_{kl}$, the energy per photon $h\nu$, the solid angle covered by the detecting lens $\Omega/4\pi$, and the efficiency of the optics $\eta$:

$$S_{F,kl} = \frac{\Omega}{4\pi} \eta h\nu N_k A_{kl}. \tag{3.20}$$

Level $k$ is in the electronic excited state and can be, but is not necessarily populated by the laser radiation directly. When $k = j$, the population of the excited level is given by (3.17) and (3.18). In the following sections we analyse the fluorescence signal when only a single transition from level $j$ is detected, and when all transitions from level $j$ and all indirectly populated levels are detected simultaneously.

### 3.6.1 Single emission-line detection

In this case we detect a transition from the level directly populated by the laser radiation, $j$, to a level in the electronic ground state $l$, different from the initial level $i$, see Figure 3.1. The resulting single emission-line detected fluorescence signal is proportional to:

$$S_{F,il} \propto N_j A_{jl}. \tag{3.21}$$

When fluorescence is saturated, according to (3.17), $N_j$, and thus the fluorescence signal, is independent of relaxation or quenching. The parameters $B_{ij}$, $B_{ji}$, $\Omega$, $\eta$, $\nu$, and $F_{B,i}$ are known constants or can be measured, so the fluorescence signal can be related to the concentration with (3.17) and (3.18).

### 3.6.2 Broadband detection

When rotational levels are not resolved in the detection, transitions are detected both from the level directly populated by the laser radiation, $j$, and from levels indirectly populated by rotational relaxation. In the case of saturation of the two-level system $(i,j)$, the population $N_i$ is known, and given by (3.18) and (3.19). The broadband detected signal is then:

$$S_{F,bb} \propto \sum_i \left( N_j A_{ji} + \sum_k A_{kl} N_k \right). \tag{3.22}$$

In the following we show that in this case the fluorescence signal is still proportional to the population $N_j$, but that that this signal also depends on rotational relaxation and quenching.

### 3.6.3 Indirect population of energy levels by rotational relaxation

We assume saturated fluorescence and validity of the balanced cross rate model. In this case the population of level $N_j$ is known. Using Figure 3.1, the rate equation of the indirectly populated level $k$ is given by:

$$\frac{dN_k}{dt} = R_{jk} N_j + \sum_p R_{pk} N_p - \sum_q R_{kj} N_k - \sum_l Q_{kl} N_k. \tag{3.23}$$
3.7. Influence of rotational relaxation and quenching on fluorescence signal of OH

Because both level $p$ and level $k$ are populated by rotational relaxation and $N_j$ is much larger than $N_k$ and $N_p$, we may assume that:

$$R_{jk} N_j \gg \sum_p R_{pk} N_p - \sum_q R_{kq} N_k.$$  \hspace{1cm} (3.24)

Then integration is possible when we assume that the laser intensity is so large that $N_j$ quickly reaches a constant value, see Lucht et al.[36]:

$$N_k = \frac{R_{jk}}{\sum_i Q_{kl}} N_j \left[ 1 - \exp \left( -\sum_k Q_{kl} \cdot t \right) \right].$$  \hspace{1cm} (3.25)

Signals are integrated during the gate time $t_G$ of the camera. When $(\sum_k Q_{kl})^{-1}$ is much shorter than $t_G$, the time dependent factor in (3.25) can be approximated by unity and $N_k$ becomes constant in time, which is assumed here. Then one obtains:

$$S_F \propto \left( \sum_i A_{ji} + \sum_k \frac{R_{jk} \sum_i A_{ki}}{\sum_i Q_{kl}} \right) N_j,$$  \hspace{1cm} (3.26)

which is proportional to $N_j$ and, with (3.18) and (3.19) to $N_s$, but also dependent on relaxation and quenching. If the short quenching time approximation cannot be made, the second term in right hand side of (3.26) has to be multiplied by an approximately constant factor $f(t_G)$. We thus find that the fluorescence signal is still proportional to the population $N_j$, but that the relation contains the unknown relaxation and quenching rates.

3.7 Influence of rotational relaxation and quenching on fluorescence signal of OH

As is shown in (3.26), the fluorescence signal depends on rotational relaxation and quenching, which in turn depend on the composition and temperature. In the case of NO, we can analyze the influence of relaxation and quenching by checking whether the total flux of excited NO is conserved, for it is assumed only to be transported. OH, however, is a reacting species. To study the influence of relaxation and quenching in the case of OH, we therefore modelled the broadband detected fluorescence signal of OH, using the relations (3.11)–(3.14) and (3.26).

The OH fluorescence signal is calibrated by comparing the signal in the turbulent flame with that in a reference flame. Because in the turbulent flame composition and temperature, and consequently quenching and rotational relaxation rates, depend on the mixture fraction, the difference between the concentration and its estimate from laser induced fluorescence measurements, also depends on the mixture fraction.
3.7.1 Simplified broadband detected fluorescence model

We simplified (3.26) by assuming that the spontaneous emission Einstein coefficients, the quenching rates and the rotational relaxation rates do not depend on the specific rotational level, and we define $A$, $Q$ and $R$ by:

\[ A \equiv \sum_i A_{ki}, \]
\[ R \equiv \sum_k R_{jk}, \]
and
\[ Q \equiv \sum_i Q_{ki}, \]

These relaxation and quenching rates are related to cross sections which are not line specific and which are given by:

\[ \sigma_{r,s} \equiv \sum_k \sigma_{r,jk,s}, \]

and

\[ \sigma_{q,s} \equiv \sum_i \sigma_{q,ki,s}. \]  \hspace{1cm} (3.28)

We then obtain a relation between the total population of the indirectly populated levels, $\sum_k N_k$, and the population of the directly populated level $j$, $N_j$, also given by Crosley[34]:

\[ \sum_k N_k = N_j \frac{R}{Q}. \]  \hspace{1cm} (3.29)

The ratio $R/Q$ is called the branching ratio, see Chan and Daily[35], and gives the degree of thermalization of the excited state prior to decay[34]; the branching ratio is in the order of magnitude of 10. The broadband signal then becomes:

\[ S_F \propto N_j A \left( 1 + \frac{R}{Q} \right). \]  \hspace{1cm} (3.30)

$N_j$ is related to the total number density of the species, by (3.17) and (3.18). The fluorescence signal obtained in the turbulent flame (as function of the mixture fraction) will generally differ from that obtained in the reference flame, because the composition and temperature, and thus the rotational relaxation and quenching, of both flames are not equal.

3.7.2 Calculated composition of reaction zone

We now present results of the estimate of these errors, using (3.30) and calculations of major species mole fractions ($O_2$, $N_2$, $H_2O$ and $CO_2$) and temperature. We now assume that in a turbulent nonpremixed flame the temperature and composition are represented by the mixture fraction and that the nonpremixed flame
Figure 3.2: Mole fraction $X$ of OH in a nonpremixed flame sheet at equilibrium, as function of the mixture fraction $\xi$, $\xi_{m} = 0.07$.

sheet is at equilibrium. For major species concentrations this is a reasonable assumption. To represent the turbulent flame composition and temperature we use calculations of a nonpremixed flame sheet at equilibrium. The calculations have been carried out by Peeters[3]. The OH mole fraction is shown in Figure 3.2. It should be noted that in an actual flame, the OH is not at equilibrium, but only the major species and temperature influence rotational relaxation and quenching. The major species mole fractions and temperature are shown in Figures 3.3 and 3.4, for the region with significant OH, $0.04 < \xi < 0.10$, where the stoichiometric mixture fraction is 0.07.

The reference flame is a laminar one-dimensional premixed flame, of which the major species mole fraction and temperature have also been calculated by Peeters[3], with use of the computer code PREMIX[37]. The reference signal is from the post flame zone of the flat flame ($\Phi = 0.88$), where the calculated mole fractions of $N_2$, $H_2O$, $CO_2$ and $O_2$ are $0.72$, $0.17$, $0.084$ and $0.023$, respectively, and the calculated temperature is 1800 K.

3.7.3 Used value of Einstein coefficient

The value of $A$ used was $(1.49 \pm 0.12) \times 10^6$ s$^{-1}$, obtained by averaging the Einstein coefficients of rotational levels with quantum numbers $N = 1$ to 12, given by Dimpf and Kinsey[38], which is almost equal to the value given by Eckbreth[24]. The reciprocal value $A^{-1}$ gives a fluorescence lifetime, $(671\pm54)$ ns, slightly smaller than the value given by Dimpf and Kinsey[38] and Crosley[34], $(686\pm14)$ ns. The larger error of the first value is due to the difference in Einstein
coefficients of the different rotational levels, whereas the latter value is obtained from one specific rotational level.

3.7.4 Used values of quenching cross section

Much literature is available on the quenching cross sections of OH [39]–[44] and on the rotational relaxation cross sections of OH [33,35], [44]–[46] The values, however, are not always unambiguous for the same conditions, and in addition, the temperatures for which the values are obtained are also widely spread. Copeland et al.[41] showed that the quenching cross section depends on the rotational quantum number, and Copeland et al.[42] showed that the quenching cross section also depends on the temperature. These results were obtained at room temperature, and will therefore not be used here. This temperature dependence is extrapolated by Garland and Crosley[43] to flame temperatures for all species of our interest. Table 3.2 summarizes the values given by Garland and Crosley[43] for temperatures between 1500 and 2500 K, which are the values used in our calculations.

3.7.5 Used values of rotational relaxation cross section

Lengel and Crosley[45] showed that the rotational relaxation cross section of N₂ depends on the rotational level, and Chan and Daily[35] found phenomenological relations of the rotational relaxation cross section of post flame gases as function of temperature and rotational quantum number. Campbell[46], however, inter-
Figure 3.4: Temperature $T$ in a nonpremixed flame sheet at equilibrium, as function of the mixture fraction $\xi$, $\xi_{st} = 0.07$.

Table 3.2: Quenching cross sections $\sigma_q,s$ for species $O_2$, $N_2$, $CO_2$ and $H_2O$, obtained from Garland and Crosley[43], for temperatures 1500, 2000 and 2500 K. The estimated accuracy of the values is 30%.

<table>
<thead>
<tr>
<th>$T$ (K)</th>
<th>$\sigma_q,s$ ($10^{-20}$ m$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$O_2$</td>
</tr>
<tr>
<td>1500</td>
<td>6.5</td>
</tr>
<tr>
<td>2000</td>
<td>4.9</td>
</tr>
<tr>
<td>2500</td>
<td>3.8</td>
</tr>
</tbody>
</table>

Interpreted these values as being relative, which were normalized to a cross section which was experimentally determined.

As our estimate of the influence of rotational relaxation and quenching on the broadband detected fluorescence is not carried out rotational level specific, we will use average rotational relaxation cross section values. No value for the rotational relaxation cross section of $O_2$ was found. Water vapour has a very large rotational relaxation cross section and is also present in large amounts in the region of the mixture fraction with significant OH. Nitrogen has a smaller cross section, but is present in much larger amounts. The relaxation cross section is therefore dominated by water vapour and nitrogen. Neglect of the relaxation by $O_2$ is expected to give an error very small compared to errors from uncertain-
Table 3.3: Rotational relaxation cross sections $\sigma_{r,s}$ for species $N_2$, $CO_2$ and $H_2O$, obtained from different authors, together with the average value chosen by us, for a temperature of about 2000 K. The estimated accuracy of the values is 30 %.

| reference | $\sigma_{r,s}$ $(10^{-20} m^2)$ |
|-----------|----------|----------|----------|
|           | $N_2$    | $CO_2$   | $H_2O$   |
| [35]      | 60       | 100      | 300      |
| [46]      | 75       | 75       | 200      |
| [44]      | 70       | -        | -        |
| [33]      | -        | -        | 170      |
| used in this study | $68 \pm 23$ | $88 \pm 25$ | $250 \pm 75$ |

The temperature dependence is obtained from the relations given by Chan and Daily[35]. The effective rotational relaxation cross section calculated with these relations is about $190 \times 10^{-20} m^2$ at 2000 K, which is about a factor 2 larger than when obtained from the species selective cross sections, with the same composition. We therefore use the relations of Chan and Daily[35] in a relative manner like Campbell[46], to represent the temperature dependence of the rotational relaxation cross section. In the range $0.04 < \xi < 0.10$, this correction for different temperatures is smaller than 12 %.

3.7.6 Estimated rotational relaxation and quenching rates

The total quenching and rotational relaxation rate are given in Figures 3.5. For the reference flame, the rotational relaxation and quenching rates estimated are $6.5 \times 10^9$ s$^{-1}$ and $4.5 \times 10^8$ s$^{-1}$, respectively.

3.7.7 Estimated broadband detected fluorescence signal

With the estimates of $R$ and $Q$, we can calculate the fluorescence signal, given by (3.30). In Chapter 5 we show that complete saturation is not reached, and therefore we present estimated signals both for saturated, where $N_j$ is independent of quenching and relaxation, and linear fluorescence, where $N_j$ is inversely proportional to the quenching rate. The accuracy of the prediction is about 40 %, using the accuracies of $A$ and of the cross sections. Relative to the signal from the reference flame per unit OH concentration, the estimated fluorescence signal from the flame sheet per unit OH concentration is shown in Figure 3.6, both for linear and saturated fluorescence. We also included the Boltzmann factor, given in (3.18), but the ratio of the Boltzmann factors in both flames does not deviate
more than 1.3 % from unity. We see that at stoichiometry, the fluorescence signal in the turbulent flame is 35 % larger than in the reference flame in the case of saturated fluorescence, and about 80 % larger in the case of linear fluorescence. At the edges of the range of $\xi$ observed, the difference is very small. The influence of quenching is weaker in the saturated case. In Figure 3.7 we show the real OH mole fraction, $X_{OH}$, and the one measured with broadband detected fluorescence and calibrated with a laminar one dimensional premixed reference flame, $\tilde{X}_{OH}$, for the case of saturated fluorescence ($\tilde{X}_{OH|sat}$) and for the case of linear fluorescence ($\tilde{X}_{OH|lin}$).

The influence of temperature on the rotational relaxation cross sections, and thus on the fluorescence signal is rather small. The temperature dependence of the number of molecules given by (3.13), and of the molecular velocity, given by (3.14), are equal for both the quenching rate and the rotational relaxation rate, and they therefore cancel. The fluorescence signal temperature dependence is therefore mainly due to the temperature dependence of the quenching cross sections, especially of water vapour, which is the dominant quencher. The quenching cross sections are smaller at higher temperatures, and consequently the quenching rates decrease with temperature faster than the rotational relaxation rates. Thus, the fluorescence signal increases with temperature, in the saturated fluorescence case slower than in the linear fluorescence case. When the stoichiometric temperature, shown in Figure 3.4, is decreased with 20 % (from 2208 to 1766 K), the saturated fluorescence signal decreases with 28 % and the linear fluorescence signal decreases with 49 %. As is shown in Figures 3.2 and 3.4, the gradients in the OH mole fraction or concentration are much larger than in the temperature. The influence of turbulent fluctuations of the temperature on the OH fluorescence signal is therefore very small compared to the influence of turbulent OH concentration fluctuations on the OH fluorescence signal.
Figure 3.6: Estimated broadband detected fluorescence signal per unit OH concentration in the equilibrium flame zone relative to that in the reference flame, for saturated fluorescence (— —) and for linear fluorescence (—— —).

Figure 3.7: OH mole fraction in nonpremixed flame sheet $X_{\text{OH}}$ (—) and the OH mole fraction measured with broadband fluorescence and calibrated with a reference flame for saturated fluorescence $\tilde{X}_{\text{OH}}|_{\text{sat}}$ (——) and for linear fluorescence $\tilde{X}_{\text{OH}}|_{\text{lin}}$ (—— —).
3.7.8 Discussion

In the previous section we have seen that the broadband detected OH profiles are systematically affected (biased) because of the influence of temperature and composition on the rotational relaxation rate and quenching rate. We expect that for the level of saturation in our experiments, the stoichiometric OH fluorescence signal is overestimated by about 50%. The bias is rather large and we consider our measurements as semi-quantitative as described by Crosley[25]. Because of the rather large uncertainties in the model presented above, we also check the influence of broadband detection experimentally, by comparing single emission-line detected fluorescence with broadband detected fluorescence, both in the laminar premixed flat flame (Chapter 6) and in the turbulent diffusion flame (Chapter 7). There is also evidence that Garland and Crosley[43] overpredict the temperature dependence of the quenching cross sections, see Barlow et al.[47], so the systematic difference between the fluorescence signals from the turbulent flame and the reference flame, and hence the bias of the turbulent flame OH measurements, may be smaller than predicted by our model.

In a turbulent nonpremixed flame, the OH concentration gradients are very large near the reaction zone, and the OH concentration fluctuations are therefore dominated by fluctuations in the location of the stoichiometric surface. This implies that the systematically affected OH broadband detected fluorescence profile follows these fluctuations in the stoichiometric surface location. We expect that the quenching and relaxation rates relative to mixture fraction are not strongly influenced by the flame conditions (i.e. exit velocities). Therefore the broadband detected fluorescence results obtained in different flames can be compared to study the influence of flame conditions on OH concentration fluctuations. The maximum temperature is not the same under all flame conditions, and in Chapter 7 we make use of the above mentioned influence of temperature on the biased OH concentration measurements.

The main reason for the use of broadband detection, was because we wanted to study the spatial structure of OH concentrations under different flame conditions. For visualization and determination of length scales of turbulence, one- or two-dimensional detection is necessary, and single emission-line detection methods cannot be used. Fortunately, these applications do not require absolute values of the concentration fluctuations.
Chapter 4

Burner setup and flame conditions

4.1 Introduction
The burner consists of a round fuel tube and an annulus for primary air supply. Pilot flames fix the flame to the nozzle. The flame environment is confined by a glass chamber, in which a secondary air flow prevents large-scale recirculation. A burner with a similar three-flow configuration was used by Driscoll et al.[48]. The primary air issues from a rather narrow annulus, so this air flow also has the character of a jet and turbulent mixing of fuel and air is enhanced. The configuration is comparable to the "throughport" burner configuration in glass melting furnaces, investigated by Wierenga[4].

4.2 Burner design
The burner is 1 m long. In the first 0.94 m, the fuel tube is 8 mm in diameter, the annulus inner diameter 30 mm, and the outer diameter 45 mm. The hydraulic diameter, relative to the length of the burner is therefore 118 and 63 for the central tube and the annulus, respectively. Near the nozzle, the inner diameter of the annulus decreases (with an angle of 7°), ending with a diameter of 15 mm. In the 8 mm fuel tube, an insert is placed, locating the pilot flames close around the fuel exit. Due to this insert, the exit diameter of the fuel tube is 6.0 mm, a value that is used throughout this thesis as fuel jet exit diameter $d_j$. The hydraulic diameter of the annulus at the exit, $d_a$, is 30 mm. The nozzle has a 9 mm wide rim between the fuel exit and the annulus exit. The recirculation behind this rim gives an extra flame stabilization, in addition to stabilization by the pilot flames. The pilot flames are discussed in the next section. For a schematic drawing of the burner nozzle, see Figure 4.1.

4.3 Pilot flames
With respect to the pilot flames, the nozzle design was based on that of Stårner and Bilger[49]. Following Stårner and Bilger, the pilot flames are fuelled by a
Figure 4.1: Cross sectional detail of burner nozzle in cross section, with central fuel exit and primary air flow annulus. Around the fuel nozzle, 12 holes provide pilot flames, premixed in a small chamber.
mixture of acetylene, hydrogen and air, where the ratio of the flow rates of acetylene and hydrogen was chosen to give the same C to H ratio as in the natural gas. This in order not to change the chemistry of the turbulent flame. On the 9 mm wide rim around the fuel exit, 12 holes (0.5 mm in diameter, located on a ring 7 mm in diameter) supply the pilot flames. The pilot flames are rich (equivalence ratio $\Phi = 1.4$), because stoichiometric flames appeared to blow off. The flow rate of the pilot flames was chosen to give a total power equal to one percent of that of flame III (see Table 4.2). The cold flow exit velocity is 12 m/s.

4.4 Flow configuration

The burner is placed in a throat providing a secondary air supply. Schematically the total burner setup is shown in Figure 4.2.

The throat is supplied by a horizontal air flow, which is redirected by a skewed plate and a blanket (Viledon, type PA 560 G). A contraction with a ratio of the surfaces of roughly 4 to 1, gives an additional flow straightening. At the blanket, the surface is square (1.2 m $\times$ 1.2 m), but gradually transforms into an octagonal shape (0.57 m from side to side). The octagonal cross section was chosen to give a better approximation of the circular wall used in the simulations. The height of the throat is 1.2 m. The throat fits to a glass flame chamber, 0.9 m high. The eight sides of the flame chamber are covered with Pyrex windows and two movable $200 \times 200$ mm$^2$ quartz windows (Heraeus Syprasil 3, 6 mm thick), which pass the ultraviolet laser beam and emitted fluorescence light.

4.5 Heating facility

In order to be able to influence the chemistry of combustion (and NO production), we were able to change flame temperatures significantly by preheating the fuel and primary air flows. For this purpose we constructed a heating facility, with which primary air was preheated, to a maximum temperature of about 773 K. By conduction, fuel is also preheated, to about the same temperature. Heating is by means of an electrical heating element (Sylvania model 63007, maximum power 18 kW), with a microprocessor controlled temperature stabilization (Watlow manufactured).

4.6 Natural gas composition

The turbulent nonpremixed flames are fuelled by natural gas, with an average composition given in Table 4.1, obtained from Geerssen[50]. Table 4.1 indicates that Groningen natural gas mainly consists of methane and nitrogen.
Figure 4.2: Burner setup with burner placed in a throat for secondary air. The flame is confined by a glass flame chamber.

Table 4.1: Average composition of Groningen natural gas, obtained from [50].

<table>
<thead>
<tr>
<th>component</th>
<th>formula</th>
<th>volume fraction (%)</th>
<th>mole fraction (%)</th>
<th>mass fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>methane</td>
<td>CH₄</td>
<td>81.30</td>
<td>81.29</td>
<td>69.97</td>
</tr>
<tr>
<td>ethane</td>
<td>C₂H₆</td>
<td>2.85</td>
<td>2.87</td>
<td>4.63</td>
</tr>
<tr>
<td>propane</td>
<td>C₃H₈</td>
<td>0.37</td>
<td>0.38</td>
<td>0.90</td>
</tr>
<tr>
<td>butane</td>
<td>C₄H₁₀</td>
<td>0.14</td>
<td>0.15</td>
<td>0.47</td>
</tr>
<tr>
<td>pentane</td>
<td>C₅H₁₂</td>
<td>0.04</td>
<td>0.04</td>
<td>0.16</td>
</tr>
<tr>
<td>hexane</td>
<td>C₆H₁₄</td>
<td>0.05</td>
<td>0.05</td>
<td>0.23</td>
</tr>
<tr>
<td>nitrogen</td>
<td>N₂</td>
<td>14.35</td>
<td>14.32</td>
<td>21.52</td>
</tr>
<tr>
<td>oxygen</td>
<td>O₂</td>
<td>0.01</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>carbon dioxide</td>
<td>CO₂</td>
<td>0.89</td>
<td>0.89</td>
<td>2.10</td>
</tr>
</tbody>
</table>
4.7 Flow conditions

The maximum fuel flow rate is determined by the pressure of the natural gas supply network and limits the power of the flame to about 25 kW. The maximum natural gas flow rate is about $6 \times 10^{-4}$ m$^3$/s (or 2.2 m$^3$/hr), the maximum primary air flow rate $1.3 \times 10^{-2}$ m$^3$/s (or 46 m$^3$/hr), and the maximum secondary air flow rate $2.6 \times 10^{-1}$ m$^3$/s (or 9.4 m$^3$/hr). In all flame settings used, the primary air flow provided a sufficient amount of oxygen to burn all fuel. Due to turbulent mixing of primary and secondary air, however, the secondary air will also participate in the combustion.

Without preheating, the exit temperature of fuel and primary air is room temperature. The secondary air is taken from outside the building, and during the measurements, the temperature was 288 K.

We investigate different combinations of fuel and primary air flow rate, and initial fuel and primary air temperature. The flame settings and the corresponding names are given in Table 4.2. The average exit velocities of fuel jet, primary air annulus jet and secondary air coflow are designated $U_f$, $U_a$ and $U_c$, respectively, the exit temperatures $T_f$, $T_a$ and $T_c$, respectively, and the exit Reynolds numbers $Re_f$, $Re_a$ and $Re_c$, respectively.

During the measurements, the flow rate settings of fuel and primary air showed a slight drift, giving an accuracy of the velocity and the flow rate of about 4%. The secondary air coflow velocity was always $(0.3\pm0.1)$ m/s, where the error is due to nonhomogeneity of the secondary air flow.

We have studied three flames with a constant ratio of fuel and primary air flow rates, with increasing fuel and primary air flow rates. This ratio is chosen so that there is a primary air excess of 18%. In order of increasing fuel and primary air velocity, the flames are named I, II and III. In a fourth flame, flame IV, the fuel flow rate is equal to that of flame III, but the primary air flow rate is 2.5 times larger, giving a primary air excess of 114%.

Flames V and VI are preheated flames. Mass flow rates of flame V are equal to those of flame III, of flame VI equal to those of flame IV. In the preheated flames, the air flow is heated up to 773 K at the base of the burner. Due to heat losses to ambient air and to the fuel, the air exit temperature is 654 K in flame V and 688 K in flame VI. The exit temperatures have been measured with a thin thermocouple. Fuel is heated inside the burner and because the fuel tube is inside the annulus tube, heat losses in the fuel flow are smaller than in the air flow. Fuel temperature is 675 K in flame V and 664 K in flame VI. Because of the higher primary air velocity of flame VI, the air exit temperature is higher and the fuel exit temperature is lower than of flame V.

The Reynolds numbers of the fuel and primary air flows, based on the hydraulic diameter of fuel nozzle and primary air annulus, the average exit velocities and the kinematic viscosities are shown in Table 4.2. The velocity and the kinematic viscosity are at the exit temperature. The kinematic viscosity of
Table 4.2: Flow conditions for six different flames. \( U \) is the average exit velocity, \( Re \) the exit Reynolds number, \( T \) the temperature. Subscripts \( j \) and \( a \) denote the fuel nozzle and the primary air annulus exit respectively. The accuracy of the fuel and primary air velocities is about 5\%, of the secondary air velocity about 30\%.

<table>
<thead>
<tr>
<th>flame</th>
<th>( U_j ) (m/s)</th>
<th>( T_j ) (K)</th>
<th>( Re_j ) (10(^3))</th>
<th>( U_a ) (m/s)</th>
<th>( T_a ) (K)</th>
<th>( Re_a ) (10(^3))</th>
<th>( U_c ) (m/s)</th>
<th>( T_c ) (K)</th>
<th>( Re_c ) (10(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>11.0</td>
<td>295</td>
<td>4.9</td>
<td>2.2</td>
<td>295</td>
<td>4.4</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
<tr>
<td>II</td>
<td>15.4</td>
<td>295</td>
<td>6.8</td>
<td>3.1</td>
<td>295</td>
<td>6.2</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
<tr>
<td>III</td>
<td>21.9</td>
<td>295</td>
<td>9.7</td>
<td>4.4</td>
<td>295</td>
<td>8.8</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
<tr>
<td>IV</td>
<td>21.9</td>
<td>295</td>
<td>9.7</td>
<td>8.0</td>
<td>295</td>
<td>16.0</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
<tr>
<td>V</td>
<td>50.4</td>
<td>675</td>
<td>5.3</td>
<td>11.2</td>
<td>654</td>
<td>5.5</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
<tr>
<td>VI</td>
<td>49.6</td>
<td>664</td>
<td>5.3</td>
<td>20.4</td>
<td>688</td>
<td>9.3</td>
<td>0.3</td>
<td>288</td>
<td>12</td>
</tr>
</tbody>
</table>

fuel and air at 673 K are about 5 times larger than at 300 K. The acceleration due to thermal expansion increases the exit velocities with a factor of 2.4, so the net effect of the preheating is a decrease of the exit Reynolds numbers by about 50\%.

4.8 Flat flame burner

The flat flame burner used to give a reference flame for calibration of OH concentrations, manufactured by McKenna, is used by many researchers, see for example Lucht[51], Lucht et al.[52] and Kaiser et al.[53]. The burner consists of a porous disk, 61 mm in diameter, cooled by water flowing through spiral-shaped tube embedded in the porous disk. Because of the large diameter, to a good approximation the flame is flat near the centerline axis. The flame is shielded by a nitrogen flow, issuing from a ring with an inner diameter of 63 mm and an outer diameter of 73 mm, to suppress entrainment of ambient air. The burner has an internal chamber for mixing of fuel and air.

Fuel is methane (99.5\%), oxidizer is air. The flows are regulated using flow meters and mercury manometers for methane and air separately. For calibration of the OH measurements, a lean flame was used, with an equivalence ratio \( \Phi \) of 0.88. The axial OH and temperature profiles of this flame and of a rich flame, \( \Phi = 1.17 \) have been measured and are discussed in Chapter 6. The total flow rates of the lean and the rich flame are \( 3.96 \times 10^{-4} \text{ m}^3/\text{s} \) and \( 3.82 \times 10^{-4} \text{ m}^3/\text{s} \), respectively.
Chapter 5

Experimental setups for laser induced fluorescence

5.1 Introduction

Five different setups used for laser induced fluorescence experiments are discussed in this chapter. Both OH and NO have been measured, the first being related to the reaction zone, the latter being a nonreactive scalar indicating the fuel mixing. Both OH and NO measurements have been conducted with one- and two-dimensional detection. In addition, OH has also been measured with single emission-line detection, to eliminate the influence of quenching and rotational relaxation. In Chapter 3, it has been shown that with saturated fluorescence, balanced cross rate and single emission-line detection, the influence of quenching and rotational relaxation is small. For the influence of rotational relaxation and quenching on the broadband detected fluorescence of OH, also see Chapter 3.

The burner setup is discussed in Chapter 4, together with the flames investigated. The laser induced fluorescence setups have been designed in a way that the fluorescence from the McKenna burner is measured with exactly the same optical configuration as the turbulent flame fluorescence, so that turbulent flame fluorescence signals can be calibrated.

The setups for detection of OH and NO, and for one- and two-dimensional detection only differ in details. Not every setup is therefore treated separately, but we discuss the main parts and indicate the differences for the different purposes.

In this Chapter, we first outline literature on OH and conserved scalar measurements in turbulent nonpremixed flames. We discuss the calibration procedure for the OH measurements. We explain the laser configurations used, for OH and NO excitation. Excitation spectra are given, together with the line selection for the case of OH and NO excitation. This line selection is primarily based on the Boltzmann distribution of the rotational levels, which requires a more detailed discussion for the case of NO. The detection part of all five setups is discussed. Finally, experimentally obtained saturation curves are presented for the different setups.
5.2 Reported OH measurements

A wide variety of measurements of OH concentrations, both of the average concentration and of the concentration fluctuations, can be found in literature. Basically, OH can be measured in a zero-, one- and two-dimensional way; three-dimensional detection is still in a premature stage at this time. Average radical concentrations, higher statistical moments and concentration distributions can be obtained from point measurements. Point measurements are necessary when the fluorescence signal is to be spectrally resolved with a spectroscope. A spectrally resolved fluorescence signal can be transformed into a concentration with smaller bias than the broadband detected signal, as is discussed in Chapter 3. For many purposes the larger bias in one- and two-dimensional measurements is accepted because of the extra information on the spatial structure that is revealed. In this study, the emphasis is on one- and two-dimensional measurements to represent the spatial flame structure.

Point measurements of OH in nonpremixed, piloted methane flames are reported by Barlow et al.[47], [54]–[56] and by Stårner et al.[57]. In these experiments, fuel is either pure or nitrogen-diluted methane. The OH concentration is related to the mixture fraction obtained, because OH measurements have been combined with major species and temperature measurements. Distribution functions of OH fluctuations in a nonpremixed hydrogen flame are given by Drake et al.[23]. In relation to the Damköhler number, OH concentrations in a nonpremixed argon diluted hydrogen flame can be found in Barlow et al.[16].

One-dimensional detection of OH in a premixed methane-air flame has first been reported by Aldén et al.[58]. The detector used was an intensified diode array camera, originally designed to serve as a detector on a spectroscope. The technique is also used by Stepowski et al.[59], to detect OH in hydrogen-air nonpremixed flames and, in a somewhat different setup, to measure temperature profiles from Rayleigh scattering signals, and by Stepowski and Cabot[60] to measure mixture fraction profiles, also from Rayleigh scattering signals.

Two-dimensional detection of OH is reported by Dyer and Crosley[61]. An introduction in "two-dimensional combustion diagnostics" is given by Hanson[62] and by Hanson et al.[63]. Equipment requirements can be found in Kychakoff et al.[64]. OH concentration measurements in axial and radial cross sections of a nonpremixed hydrogen-air flame are presented by Seitzman et al.[65, 66].

5.3 Reported strategies for conserved scalar measurements

In combustion diagnostics, there is a desire for measuring the mixture fraction, because many (simple) combustion models are based on the transport of a conserved scalar like the mixture fraction.
Point measurements of the mixture fraction are possible when major species concentrations and density are measured with combined spontaneous Raman spectroscopy and Rayleigh scattering. The most accurate method is to derive the mixture fraction from the fraction of C, H and O atoms in the major species. The major species that are used are CH\textsubscript{4}, O\textsubscript{2}, H\textsubscript{2}O, CO\textsubscript{2}, CO, and H\textsubscript{2}[56]. This method is successfully applied and also combined with other methods, like laser induced fluorescence detection of OH, see for example Barlow et al.[56]. The detection of so many species is done with several detectors on a spectroscope, giving zero-dimensional results. For two-dimensional measurements of the mixture fraction in a turbulent flame, it is hardly possible to detect all major species mentioned above. With loss of accuracy, however, it is possible to derive the mixture fraction in a nonpremixed flame from two out of the three quantities fuel, oxygen and temperature, see Stårner et al.[67]. This requires two cameras with appropriate filters for detection of the species (using Raman scattering) and/or temperature (using Rayleigh scattering).

An alternative for this double Raman or combined Raman/Rayleigh method, is one- or two-dimensional detection of Mie scattering from macropscopic particles which are seeded to the fuel flow. Differential diffusion of fluid and seed particles limits the detection to large-scale structures[67]. In addition, maintaining a constant seed concentration is difficult in practice.

Differential diffusion is eliminated when a gas is used as seed, and also the seed concentration is more easily controlled. With our experimental facilities, seeding gas concentrations can be measured instantaneously with laser induced fluorescence. Biacetyl can be used as a seeding gas, see Lozano et al.[68], but this gas is thermally instable at elevated temperatures, see Lee et al.[69], so it is only applicable to cold flows. Another species that can be measured relatively easily, is NO. According to DiRosa et al.[70], NO is "an appealing seed species", proposed by several authors, like Stårner et al.[67], DiRosa et al.[70], McMillin et al.[71], and Lee et al.[69]. It is a species that is relatively thermally stable[69] and chemically stable over a wide range of conditions[70]. However, NO is toxic and one of the main combustion pollutants. In addition, it may react to NO\textsubscript{2} in the presence of O\textsubscript{2}[70]. In flames, quenching is the main cause that makes that fluorescence of NO is not conserved. The major species, although not with the largest quenching cross sections, are the main cause of quenching[71], see also Section 8.2. Quenching by carbon dioxide and by water is dominant. The wide temperature range in which the NO is to be detected in flames, makes that the Boltzmann fractions will change throughout the flame. This effect can be minimized with the proper choice of an absorption line, but is never absent, see Section 5.7.3.

Another more fundamental problem is the fact that fluorescence signal is proportional to the concentration, and that the instantaneous temperature is to be known to convert this concentration to a mass or mole fraction. The mixture fraction on which the combustion models are based, is the normalized
mass fraction of nonreactive species issuing with the fuel.

Notwithstanding these complications, we show in Chapter 8 that one- and two-dimensional detection of NO seeded to the fuel in both isothermal jets and flames provides information on the shear layer mixing in the flames, on the influence of combustion on turbulence and yields length scales of turbulence.

5.4 Calibration method

To put the turbulent flame measurements to an absolute basis, the fluorescence signal is compared with that from a reference flame, following Lucht et al. [72], Drake et al. [23] and Schefer et al. [73]. This reference flame is placed in the setup at the location of the turbulent flame, preserving optics alignment and guaranteeing a constant relation between concentration and signal. The reference flame is a laminar premixed flat methane/air flame of a McKenna porous plug burner (see Section 4.8). Turbulent flame measurements are preceded by a calibration measurement. The OH concentration at a specific location in the reference flame is obtained from full chemistry, kinetic model calculations, using the PREMIX computer code developed by Kee et al. [37]. The chemistry model that has been used in the calculations was developed by Warnatz [74]. The calculations have been carried out by Peeters and are discussed in greater detail in [3]. To test the calculations, we have investigated this burner under lean and rich conditions and we have compared measured temperatures and relative OH concentration profiles with the calculations, of which the results are presented in Chapter 6. This results in an estimation of the calibration error.

The concentration was measured in the lean flame ($\Phi = 0.88$) at 25 mm above the burner surface, where the calculated OH concentration is $2.13 \times 10^{15}$ cm$^{-3}$, see Chapter 6. All turbulent flame measurements were carried out on the same day, without changing laser or detection alignment. Therefore, these turbulent flame measurements have been calibrated with the same reference flame measurement, and a calibration error is systematic in character.

5.5 Laser configurations

All three setups for OH measurements (single emission-line detection point measurements, and broadband detected one- and two-dimensional measurements) used the same laser configuration, differing from the laser configuration in both setups for NO measurements (broadband detected one- and two-dimensional measurements). The laser configuration is determined by the excitation wavelength of the species.

The heart of the laser system is a Nd:YAG laser, with a repetition rate of 10 Hz, a pulse duration of 5 ns, and an energy per pulse of 900 mJ at a wavelength
of 1064 nm (manufactured by Quantel International, currently Continuum, model YG-680). For the two-dimensional detection measurements, the laser was run at a repetition rate of 8.3 Hz. For our applications, the fundamental Nd:YAG laser light was doubled, giving pulses of 400 mJ at 532 nm. With this wavelength a dye laser section (Quantel International, TDL 60) was pumped, with a grating tuned oscillator and two amplifiers. The second amplifier was designed to give a cylindrical beam, 5 mm in diameter. We used Rhodamine dyes. The fundamental dye laser light can be doubled, mixed with the fundamental Nd:YAG laser light or mixed after being doubled, giving wavelengths in different ranges, where mixing-after-doubling gives the smallest wavelength light.

The wavelengths of OH lines are given in the extensive report of Dieke and Crosswhite[29], those of NO in Engleman et al.[30]. Both in the case of OH and NO, we used excitation in the $A^2\Sigma, v' = 0 \leftarrow X^2\Pi, v'' = 0$ band. This means excitation from the lowest vibrational state in the electronic ground state to the lowest vibrational state in the first excited electronic state. For OH, this results in a wavelength of about 309 nm, for NO in a wavelength of about 226 nm. For the laser configuration this has some consequences. The 309 nm wavelength is obtained by doubling the fundamental dye laser wavelength (from Rhodamine 590), whereas a wavelength of 226 nm is obtained from mixing the doubled dye laser light (from a mixture of Rhodamine 610 and 640) with the fundamental Nd:YAG laser light (1064 nm). Each KDP crystal used for the doubling or mixing is tuned with a feedback system, a so-called tracking system, which is sensitive to alignment. In the mixing-after-doubling configuration, two of these tracking systems are used, which makes stable tracking difficult. Laser light has been obtained with an energy of about 17 mJ per pulse at 309 nm, and of 3 mJ per pulse at 226 nm.

5.6 Laser beam

5.6.1 Laser beam traverse

By means of a periscope with two high energy laser (HEL) mirrors, the laser beam is brought to an adjustable height above either the burner nozzle of the nonpremixed flame burner or the burner surface of the McKenna burner. The HEL mirrors were manufactured by Melles Griot, 12 mm in diameter, and not designed for a specific polarization. The combination of two HEL mirrors reduce the laser energy with about 25 % at 309 nm, and 43 % at 226 nm, giving a laser pulse energy of about 12 mJ and 1.7 mJ after the mirrors for the case of OH and NO, respectively. The upper mirror was traversable over a range of about 1 m and was stepping motor driven.
Figure 5.1: Radial profile of the fluorescence signal (—) and of the scattering signal (— —), with a laser wavelength of about 309 nm. Note that the profiles are affected by spherical aberration and yield too large values.

5.6.2 Fluorescence volume

For the zero- and one-dimensional detection, the laser beam is focussed with a spherical, 0.5 m focal length, 25 mm diameter, UV grade fused silica lens (Melles Griot manufactured). The beam waist coincides with the center line of the burners.

The waist diameter was measured by projecting scattering of the laser beam at a vertically mounted diode array camera. Projection was by means of a single spherical lens (focal length 100 mm, diameter 50 mm). Because of saturation of the fluorescence, the region from which significant fluorescence arises is larger than the laser beam diameter. We therefore also measured the diameter of the volume with significant fluorescence. This was done for the case of OH, with a wavelength of about 309 nm, for NO the values may be slightly different. The profiles are shown in Figure 5.1. The values obtained are 0.7 and 1.7 mm for the laser beam diameter and the fluorescence volume diameter, respectively. Both are the full width at half maximum. We see that the fluorescence volume diameter is about 2.5 times larger than the laser beam diameter. For these measurements, a single lens has been used for the projection, with a rather large diameter to focal length ratio or aperture. After these beam waist measurements, it was discovered that all measurements with this single lens projection suffered severely from spherical aberration, giving broader peaks and characteristic "shoulders" in the profiles. These were absent when a UV Nikkor camera lens was used, and
for this reason we used the UV Nikkor camera lens in the broadband detected experiments. The beam waist values obtained are therefore expected to the too large because of the spherical aberration. They are also significantly larger than the values of the laser beam diameter and of the fluorescence volume, reported by Salmon and Laurendeau[75] and by Lucht[51]¹. They reported values 374 μm[75] and 360 μm[51] as laser beam diameter, and 636 μm[75] and (500±100) μm[51] as fluorescence volume diameter, also using 500 mm focal length focussing lenses. Despite this expected overestimation of our laser beam diameter and fluorescence volume diameter, we know that the fluorescence measuring volume diameter is about 2.5 times larger than the laser beam diameter. A laser beam diameter of about 0.3 mm and a 2.5 times larger fluorescence measuring volume depth of 0.75 mm seem to be more reasonable. This fluorescence volume diameter gives the spatial resolution of the measurements, and it is not used in the measurements itself.

5.6.3 Laser sheet
To be able to detect OH or NO in a two-dimensional plane, a laser sheet was constructed. A spherical lens has been used to focus the laser beam and a cylindrical lens to diverge the laser light into a laser sheet. The thickness of the sheet is determined by a spherical focussing lens. A quartz lens with a focal length of 1.5 m has been used to produce a laser sheet with almost constant thickness of about 0.5 mm in the region of interest. A negative cylinder lens with a focal length of −12.7 mm expanded the laser beam in the vertical direction, and was placed 0.70 m away from the jet axis. The divergence of the laser sheet resulted in a laser intensity decreasing with the distance from the cylindrical lens, but in an only slightly decreasing OH or NO fluorescence signal. The decrease was only observed in the average images, and is not significant in the instantaneous images presented here. The size of the laser sheet in axial direction was about 15 cm at the jet axis.

5.7 Excitation spectra
By scanning the laser wavelength and detecting the broadband fluorescence signal, excitation spectra with the rotational line structure of OH or NO are obtained. For OH, excitation spectra have been obtained in the various flames given in Table 4.2 and in the laminar premixed flat flame; for NO, spectra have been obtained from NO seeded to these flames. The dye laser wavelength is controlled by a personal computer. The wavelength of this system is set by comparing the

¹ Salmon and Laurendeau[75] and Lucht[51] used 50 mm diameter, 200 mm and 300 mm focal length lenses for projection, which have a smaller aperture and are therefore less sensitive to spherical aberration.
Figure 5.2: Part of the rotational line structure of $A^2 \Sigma, v' = 0 \leftarrow X^2 \Pi, v'' = 0$ OH, obtained from an excitation scan of the laser. $\lambda_l$ is the laser wavelength. The $Q_1(8)$ line used in the experiments.

lines in the spectra with those reported by Dieke and Crosswhite [29] and Engleman et al. [30] for OH and NO, respectively. The rotational line structure of NO is much denser than that of OH, so identification of every line in the spectrum of NO by comparing with the lines of Engleman et al. [30] is more difficult, but is possible when mapping the bandheads. Based on the excitation spectra, the excitation lines are selected. It is important that the line is identified exactly, because the Boltzmann distribution determines the temperature sensitivity of the different lines, see Section 3.3. In addition, excitation spectra are used to select a line that is well separated from other lines.

5.7.1 OH excitation spectrum

In Figure 5.2 a part of the rotational lines in the $A^2 \Sigma, v' = 0 \leftarrow X^2 \Pi, v'' = 0$ excitation spectrum is shown, obtained in the laminar premixed flat flame, but equal spectra were obtained in the turbulent flames. In Figure 5.2, we also identify the lines of the $Q_1$ branch. Comparison with Dieke and Crosswhite [29], showed that all lines found in the spectrum were reported to originate from OH. We therefore know that no fluorescence from other species interferes with the OH fluorescence. However, in the bright yellow parts flame I ($x \geq 200$ mm), soot scattering or PAH fluorescence interfered with the OH fluorescence, as discussed in Chapter 7.
5.7. Excitation spectra

Figure 5.3: Part of the rotational line structure of $A^2\Sigma, v' = 0 \leftarrow X^2\Pi, v'' = 0$ NO, obtained from an excitation scan of the laser. The average temperature was about 1200 K. $\lambda_1$ is the laser wavelength. Indicated is the $Q_1(19)$ line used in the experiments.

Absorption line selection

The excitation line we have chosen is $Q_1(8)$ at 309.329 nm. This transition gives large signal, is separated from neighbouring transitions, and is relatively insensitive to temperature: for the rotational level in the electronic ground state with $J'' = 8\frac{1}{2}$, connected to the $Q_1(8)$ line, the Boltzmann factor changes only 10% for a temperature range of 1000 to 2600 K, see Lucht et al. [72].

5.7.2 NO excitation spectrum

In Figure 5.3, the excitation spectra of the $A^2\Sigma - X^2\Pi$ band of NO is shown for an average temperature of 1200 K. For this purpose NO was seeded to a diffusion flame. The spectrum of NO is very dense compared to the line width, so that it is generally not possible to select one single transition. The selected absorption line, $Q_1(19)$, indicated in the figure, is relatively well separated from other lines. The arguments for the selection of this line are given in Section 5.7.3.

5.7.3 Absorption line selection

The fluorescence signal is proportional to the product of the NO concentration and the Boltzmann factor of the initial energy level, see Chapter 3; the Boltzmann factor is temperature dependent. In the experiments where NO was seeded to the fuel, the fluid containing NO will pass a wide temperature range; from
room temperature at the burner exit to flame temperatures at about 2000 K. As will be shown in the following, it is not possible to have a constant Boltzmann factor in the complete temperature range. A careful selection of the absorption line is therefore necessary to minimize the influence of temperature on the Boltzmann factor in this range.

To calculate the Boltzmann factor, defined \( F_B \), we used values for the different energy level constants, given by Huber and Hertzberg\[28\]. The formula used for the calculation has a general form given in Section 3.3.

In principle an absorption line from two sets of lines can be selected: from the \( X^2\Pi_{1/2} \) state and from the \( X^2\Pi_{3/2} \) state, see Reisel et al.\[31\]. Figure 5.4 shows the Boltzmann factors as function of temperature for the \( X^2\Pi_{3/2} \) state for the rotational energy levels \( J'' = N'' - \frac{1}{2} \), \( N'' = 5, 10, 15, 20 \) and 25, and in Figure 5.5 for the \( X^2\Pi_{1/2} \) state for the rotational energy levels \( J'' = N'' + \frac{1}{2} \), \( N'' = 5, 10, 15, 20 \) and 25.

For small rotational levels, the \( F_B \) quickly decreases with increasing temperature. For \( N'' = 5 \), at 300 K \( F_B \) is more than 8 times larger than at 2000 K for the \( X^2\Pi_{1/2} \) state, and more than 5 times larger for the \( X^2\Pi_{3/2} \) state. For larger values of \( N'' \), the maximum shifts to higher temperatures. At room temperature, \( F_B \) decreases with increasing \( N'' \), whereas for high temperatures it increases. Due to the higher energy level of state \( X^2\Pi_{3/2} \), the population of this level is generally smaller and so is \( F_B \).

From Figures 5.4 and 5.5 it is apparent that small values of \( N'' \) are not appropriate, because \( F_B \) changes much with temperature. When \( N'' \) is very large, \( F_B \) becomes very small for low temperatures. Therefore, the best choice is \( N'' \approx 20 \) in the \( X^2\Pi_{1/2} \) state. From the excitation spectrum we have chosen \( Q_1(19) \), for it combined a large Boltzmann factor over a wide temperature range with a relatively good separation from other lines.

### 5.8 Detection

We now discuss the different detection schemes for single emission-line detection of OH, one- and two-dimensional broadband detection of OH, one- and two-dimensional broadband detection of NO. Some parts of the setups are characteristic of the species, like the filters used, others are characteristic of the dimension of the measurements, like the type of camera or data storage.

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2. Engleman et al.\[30\] is not correct in designating the level with \( J'' = N'' - \frac{1}{2} \) the \( f_1 \) electronic ground state, for that does not match with the value of \( J' \) in the \( F_1 \) first excited electronic state. We therefore follow Reisel et al.\[31\], who are correct on this point.
5.8. Detection

Figure 5.4: Boltzmann factor for $N'' = 5 (-)$, 10 ($-$), 15 ($-$ $-$), 20 ($-$ $-$) and 25 ($-$ $-$), for the $X^2\Pi_{1/2}$ state, for temperatures up to 2000 K.

Figure 5.5: Boltzmann factor for $K = 5 (-)$, 10 ($-$), 15 ($-$ $-$), 20 ($-$ $-$) and 25 ($-$ $-$), for the $X^2\Pi_{3/2}$ state, for temperatures up to 2000 K.
5.8.1 Single emission-line detection of OH

For single emission-line detection of OH, the fluorescence spectrum, which consists of many lines (see Chapter 3) has to be spectrally resolved, so that one emission-line can be selected. For the balanced cross rate model to be useful, the emission should be from a spontaneous emission transition from the rotational level directly populated by the laser excitation. Excitation of the $Q_1(8)$ line, gives the following lines that can be used for detection: $R_1(7)$, $Q_1(8)$, $P_1(9)$, $Q_{12}(8)$, $P_{12}(9)$ and $O_{12}(10)$. The line $Q_1(8)$ cannot be used, because the wavelength is the excitation wavelength, so scattering signals can interfere. The satellite branches $Q_{12}(8)$, $P_{12}(9)$ and $O_{12}(10)$ are very weak and are therefore also not appropriate.

To resolve the fluorescence signals spectrally, we used a double, 0.85 m spectrooscope (Specx 1404), with two 1800 lines/mm ruled gratings. At the exit, a diode array camera (O-SMA, Princeton instruments) detected a part of the fluorescence spectrum with a range of 5 nm. The diode array camera consisted of 700 connected diodes with 25 $\mu$m separation. The limit of the spectrooscope resolution based on the resolution of the camera, is 7.4 pm. A finite entrance slit width decreases the resolution, for example with a slit width of 200 $\mu$m, the resolution was 60 pm.

As outlined by Lucht[51] and Salmon and Laurendeau[76], it is possible to reject in one direction the nonsaturated wings of the fluorescence volume with the entrance slit of the spectroscope. For this, the image of the measuring volume was rotated 90°, so that the image was parallel to the entrance slit. Combined with a periscope for traversing the measuring volume, a system with five, 50 mm diameter flat mirrors facilitated image rotation. The measuring volume was traversable in the flame from $x = 100$ mm to $x = 250$ mm, and measurements were obtained at $x = 100, 150, 200$ and 250 mm. The fluorescence signal was first collimated with a traversable spherical 500 mm focal length, 50 mm diameter, UV grade fused silica lens (manufactured by Melles Griot). After the periscope and image rotator, the beam was focussed at the entry slit, again with a 500 mm focal length, 50 mm diameter lens. The aperture of this lens matched with the aperture of the spectrooscope of $f/8$, so the beam was covered by the first mirror in the spectrocope completely. The magnification factor was unity. Because of the image rotation, the entry slit width determined the height of the measuring volume, $H$, see Figure 5.6. In the experiments, the entry slit width was set at 200 $\mu$m, so $H = 200$ $\mu$m. As a result, only the central part of the fluorescence volume contributed to the detected signal, because the fluorescence volume diameter was about 0.75 mm. The depth of the measuring volume, $D$, was determined by the fluorescence volume diameter, estimated in Section 5.5, so $D \approx 0.75$ mm. The length of the measuring volume was equal to the height of the spectrocope entry slit, in the experiments set at the smallest value, 2 mm, so $L = 2$ mm.

The diode array camera was enhanced by means of a so-called multichannel plate, or MCP. A gate pulse generator facilitated triggering of the camera and
enabling the camera for only a short time. The smallest gate time was 6 ns, larger than the laser pulse duration. The complete laser pulse was therefore detected. To reduce jitter, the gate time was set at 40 ns. The complete setup of single emission-line detection is shown schematically in Figure 5.7.

A typical OH fluorescence spectrum, obtained in a flat flame, after excitation of the selected \( Q_{1}(8) \) line in the \( A^{2}Σ, ν' = 0 ← X^{2}Π, ν'' = 0 \) band, is shown in Figure 5.8. We see the main lines \( R_{1}(7) \), \( Q_{1}(8) \) and \( P_{1}(9) \), and the satellite line \( P_{12}(9) \) line. Note that \( Q_{1}(8) \) emission is at the excitation wavelength and at this wavelength the signal also contains scattered light detected. The satellite line \( Q_{12}(8) \) is not resolved from the \( Q_{1}(8) \) line. The satellite \( O_{12}(10) \) line is at 316.50 nm and is not covered. All other lines are due to emission from rotational levels populated by rotational relaxation.

### 5.8.2 One-dimensional, broadband detection of OH

One-dimensional detection is necessarily associated with broadband detection, because a spectroscopic for wavelength selection cannot be used and interference filters bandwidth is far too large. The consequences of broadband detection on the relation between the OH concentration and the fluorescence signal are discussed in Chapter 3.

The camera used for one-dimensional detection was the same diode array camera as used for the detection of the fluorescence spectra, discussed in Section 5.8.1, with same gating time of 40 ns. The method was adopted from Aldén
et al.[58]. The laser beam and diode array camera were traversable through the flame, ranging $x = 50$ mm to about 1 m, which is larger than the flame length. Measurements were obtained at $x = 50, 100, 150, 200, 250$ and 300 mm. For larger $x$, soot strongly interfered with the OH measurements in most flames. The setup is shown schematically in Figure 5.9.

The fluorescence originating from the one-dimensional laser beam was projected at the diode array by means of a UV Nikkor 105 mm, $f/4.5$ camera lens. The aperture used was $f/8$. The field of view has been measured by projecting an object of known size at the diode array. The diodes in the array measure 25 $\mu$m to 2.5 mm, but due to the multichannel plate, the resolution along the diode array is three times smaller, 75 $\mu$m. The image reduction factor was 6.11, so the diode separation projected at the measuring volume, was 0.15 mm. The height of the diodes projected at the laser beam was much larger than the laser beam diameter, so the measuring volume was not clipped by the diodes. The measuring volume had the shape of a pill box with a height of 0.15 mm and a diameter equal to that of the fluorescence volume, which was estimated to be 0.75 mm.

Like the excitation line, the fluorescence detected was in the (0,0) vibrational band. A Schott UG11 filter has been used to pass only the (0,0) vibrational band. This filter also blocked very strong scattering from the fundamental dye laser light which was dumped outside the laser. UV light with the wavelength of the laser passes the UG11 filter. Rayleigh scattering, however, has not been detected because of the horizontal polarization of the (electric field vector of the) laser light. Laser light scattered in the room gave a background signal which was
independent of the presence of a flame, constant in time and almost constant along the diode array. The scattered light signal has been suppressed as much as possible by shielding. The average background was estimated from the interpolation of the signals at the edges of the instantaneous one-dimensional images and subtracted in the data processing algorithms.

The camera was controlled by a personal computer and the measurements have been stored on the personal computer’s hard disk. Data processing was on a Hewlett-Packard 730 workstation.

5.8.3 Two-dimensional, broadband detection of OH

Fluorescence originating from the laser sheet was detected with a gated and intensified Proxitronic Nanocam CCD camera. Gating time was 100 μs. A UV Nikkor 105 mm camera lens projected the fluorescence from the laser sheet at the image intensifier. The aperture used was f/8. By recording the image of a ruler we have determined the field of view to be 100 mm horizontally and 78 mm vertically, with a pixel size projected at the measuring volume of 0.14 × 0.14 mm².

An electronic system was built to synchronize camera, video recorder and laser pulse. A trigger pulse was extracted from the video signal produced by the camera. The frequency of this pulse was divided by 3 and with that signal the laser and the camera were triggered. The camera was in the store mode, so it
Figure 5.9: Experimental setup for one-dimensional broadband detection, where the diode array camera is used as one-dimensional detector. Projection at the diode array camera was by means of a UV Nikkor camera lens.

produced video frames at 25 Hz. Both the firing of the laser and the refreshing of the frame were triggered with a 8.3 Hz repetition rate pulse. When stored on video tape, three consecutive images are identical. Off line, one of every three images on video tape has been digitized with a 128 grey scales frame grabber in a Sun Sparc workstation and stored on a 1 Gbyte hard disk. No attempts have been made to calibrate the measurements. The two-dimensional detection setup is shown schematically in Figure 5.10.

One-dimensional OH concentration measurements detected with a diode array camera are more appropriate for quantitative interpretation than the two dimensional measurements. Nearly in real-time, the one-dimensional diode array signals are digitized and stored on a micro computer hard disk, whereas the two dimensional video images signals are stored on video tape and digitized off-line. This video tape storage introduces slight nonlinearity effects and extra noise. In addition, the dynamic range of the video frame grabber is 128 grey values (7 bits), compared to roughly 16 thousand (14 bits) of the one-dimensional digitizer. A third advantage is the huge memory occupation of two-dimensional images: a set of 5000 one-dimensional measurements takes 10 Mbytes of memory, whereas as set of 5000 two-dimensional images takes about 2 Gbytes. For statistical analysis, these large numbers of samples are required. We will therefore use the one-dimensional measurements to quantify the observations made on the two-dimensional images. The advantages of one-dimensional detection over point measurements is the revealed spatial structure and the possibility of instanta-
Figure 5.10: Experimental setup for two-dimensional broadband detection. With a negative cylinder lens a laser sheet is constructed. Fluorescence is detected by means of a CCD camera and stored on video tape. Off-line, data were processed.

5.8.4 One- and two-dimensional, broadband detection of NO

The setups for one- and two-dimensional detection of NO were almost equal to those for one- and two-dimensional detection of OH, except for the laser configuration and high energy mirrors, as discussed in Section 5.5. On the detection side of the setups, a filter has been used to suppress all soot scattering and laser induced fluorescence of PAH, which is for a wavelength of 226 nm much stronger than the fluorescence of NO. For this purpose, an interference bandpass filter (custom made by Acton Research) was used that transmitted only the (0,1) transition (this is the transition to the $v'' = 1$ band in the electronic ground state). The center of the band pass filter was at 241.5 nm, the bandwidth was 18.0 nm. The laser wavelength at about 226 nm was blocked by this filter, so no elastic scattering contributed to the signal detected, and also PAH fluorescence was completely suppressed. This filter was used in both the one- and two-dimensional setup.

Timing of the NO measurements (pulse duration and repetition rate) is equal to the timing of the OH measurements. In the one-dimensionally detected NO measurements, the image reduction factor was 6.20, giving a diode separation projected at the measuring volume of 0.15 mm. In the two-dimensional measurements, the pixel size projected at the measuring volume was $87 \times 87 \, \mu m^2$. The field of view is 63 mm horizontally and 50 mm vertically. The one-dimensional measurements have been obtained at $x = 50, 100, 150, 200$ and 250 mm, and when the signal was sufficiently high, at $x = 300$ mm. The two-dimensional mea-
measurements covered three regions, \( x = 25-75 \text{ mm}, 75-125 \text{ mm} \) and 125-175 mm.

In contrast to the OH concentration, the maximum signal of the NO seed decreases with axial distance (approximately with \( 1/x \) in isothermal jets, faster in the flames). In the one-dimensional measurements, the dynamic range of the camera was sufficient to have nonsaturated camera signal at \( x = 50 \text{ mm} \) and a sufficiently large signal to noise ratio at \( x = 250 \text{ mm} \). In the one-dimensionally detected measurements, the (nonlinear) camera gain therefore has been kept constant. In the two-dimensionally detected measurements, it was adjusted at every station to match the maximum concentration with the maximum signal, in order to have maximum signal to noise ratio.

The seeding flow rate has been matched to the natural gas flow rate, to have the same NO concentration in the fuel flow of all flames, except in the preheated flames. To have a significantly large signal for larger \( x \) and to have NO concentrations much larger than those produced in the flame, the NO seeding volume fraction in the natural gas flow was 0.74%.

### 5.9 Saturation

In Chapter 3 we have shown that with saturated fluorescence the bias in the broadband detected signal is smaller than in the case of linear fluorescence. It is therefore important to know the degree of saturation. From (3.17) and (3.26) we find that for the fluorescence signal the only parameter that depends on the laser intensity is the population of the rotational level in the first excited electronic state directly populated by excitation. Therefore, the broadband detected fluorescence signal can be used to determine the degree of saturation. This is an important observation, because the fluorescence signal in the single emission-line detected fluorescence signal is so weak that we require the largest laser energy for a signal sufficiently large to detect.

We decreased the laser energy by detuning the doubling crystal in the Nd:YAG laser, this in order not to spoil the laser beam intensity profile. In the one-dimensional OH detection setup, we measured the fluorescence from arising from the laminar flat flame. For determination of the saturation of the one-dimensional NO detection setup, we measured fluorescence of NO seeded to flame 1 (Table 4.2) at \( x = 100 \text{ mm} \).

Lucht et al.[72] report saturation levels for OH of 90 to 95%, after excitation of the \( Q_1(8) \) line of the \( \text{A}^2\Sigma, v' = 0 \leftarrow \text{X}^2\Pi, v'' = 0 \), which is the same excitation line we have used. Their energy per pulse was 3.5 mJ at maximum and the fluorescence volume diameter 0.3 mm. This results in a laser intensity per unit bandwidth (with unit cm\(^{-1}\)) of \( 5 \times 10^{12} \text{ W/(m}^2\text{cm}^{-1}) \). The high level of saturation was reached because only the largest intensity part of the fluorescence pulse was detected, using a gate time of 2 ns. According to Lucht et al.[77], detection of the complete fluorescence pulse would require a much larger laser pulse.
Figure 5.11: Fluorescence signal as function of laser energy, for one-dimensional detection of OH, with the least squares fit saturation curve and limiting value.

Figure 5.12: Fluorescence signal as function of laser energy, for one-dimensional detection of NO with the least squares fit saturation curve and limiting value. The NO was seeded to the fuel of flame I, and the fluorescence was measured at $x = 100$ mm at centerline.
energy. In our case, we used the same excitation wavelength, our maximum laser pulse energy was 15 mJ, our fluorescence volume diameter was estimated to be 0.75 mm. The bandwidth of the laser is about 1 cm$^{-1}$. The laser intensity per unit bandwidth was about $5 \times 10^{12}$ W/(m$^2$cm$^{-1}$), a value similar to that of Lucht et al.[72]. We have detected the complete fluorescence pulse, and the resulting level of saturation in the one-dimensional measurements is therefore 70 to 75%. This is shown in Figure 5.11, where the fluorescence signal is given as function of the laser pulse energy. We have fitted a curve represented by (3.17), where the laser pulse energy is of course proportional to the laser intensity. This fit yields the limiting value.

The resulting bias of the broadband detected fluorescence signal can be found in Figure 3.6, and gives an overestimation of about 50% due to broad band detection at stoichiometry, when the temperature is the equilibrium temperature, see Chapter 3.

Results of NO fluorescence saturation have only been found with Verdieck and Bonczyk[78], who did not give the specific absorption line used. For a laser spectral intensity of $6 \times 10^{10}$ W/(m$^2$ cm$^{-1}$) they did not reach saturation. In our case, the laser pulse energy was 1.7 mJ, the fluorescence volume diameter was estimated to be 0.75 mm, giving a laser spectral intensity of about $7 \times 10^{11}$ W/(m$^2$ cm$^{-1}$). The fluorescence signal for different values of the laser energy, together with the least squares fit and limiting value, is shown in Figure 5.12. At a laser pulse energy of 1.7 mJ, the saturation level is 99.5%. The level of saturation is so high because at the location of the measurements the concentration of strong quenching molecules, like O$_2$, CO$_2$ or H$_2$O, is very small.
Chapter 6

Temperature and OH measurements in flat flames

6.1 Introduction

To calibrate the turbulent flame measurements of OH, we used a laminar premixed flat flame, produced by a McKenna burner, see also Section 4.8. To transform the flat flame fluorescence signal into an OH concentration, the OH concentration profile has been calculated using the PREMIX computer code[37], with the Warnatz chemistry model[74]. Heat losses due to radiation of CO₂ and H₂O have been included in the model, using emission coefficients obtained from Hubbard and Tien[79]. The calculations are discussed more extensively by Peeters[3]. We compare the temperature and OH concentration with the temperature measured with a thin thermocouple and the relative OH concentrations measured with laser induced fluorescence. For the lean flame, the OH concentrations are obtained both by single emission-line detection and broadband detection, for the rich flame by single emission-line detection only.

6.2 Temperature profiles

Temperature profiles have been measured in the laminar premixed flat methane/air flames, with a thin thermocouple. The thickness of the thermocouple is 50 μm, the bead diameter is 130 μm. Flames have been operated under lean and rich conditions. Equivalence ratios Φ of these flames were 0.88 and 1.17, total cold flow rates $3.96 \times 10^{-4}$ m$^3$/s and $3.82 \times 10^{-4}$ m$^3$/s, respectively.

The velocity of the gases in the flat flame is rather small, the burned gas velocity is about 0.8 m/s. Therefore, the temperature measurements have to be corrected for radiation losses. Because of the small gradients in the post-flame zone of the flat flame, conduction losses are negligible. Several radiation correction methods are available, either assuming the thermocouple to be a cylinder, when the two thermocouple wires would be connected without any thicker part, or a sphere, when in the welding process at the junction a spherical bead has formed. Real thermocouples, however, only approximately have one of those shapes. The
different assumptions result in different values of the Nusselt number $Nu$, and hence different radiation corrections, see for example Bradley and Matthews[80]. For the radiation correction, a characteristic diameter, the emissivity of the thermocouple and the conductivity and viscosity of the gas have to be known. The flue gas thermal conductivity and viscosity have been found in Geerssen[50].

The thermocouple consisted of Pt/Rh(6%)-Pt/Rh(30%). For these alloys, the emissivity is not reported, and therefore the emissivity of the thermocouple is assumed to be equal to the emissivity of a Pt/Rh(10%) wire, found in Touloukian and DeWitt[81]. It is assumed to be a linear function of the temperature. For a temperature of 1750 K, the emissivity used is 0.22. Bradley and Entwistle[82], however, showed that the emissivity increases with the ratio rhodium to platinum, giving for a temperature of 1000 K an emissivity which is 17 % larger for Pt/Rh(10%) than for pure Pt. Hence, the emissivity of our thermocouple is expected to be larger for Pt/Rh(10%), and the temperature results may yield more precise values of the emissivity.

Bradley and Matthews[80] propose a radiation correction method based on a balance between the radiative heat loss and the convective heating of a cylinder. The heat transfer to the cylinder is given by a Nusselt number, valid for $0.01 < Re < 10^4$,

$$Nu = 0.42 \times Pr^{0.2} + 0.57 \times Pr^{0.33} \times Re^{0.5}.$$  \hspace{1cm} (6.1)

The characteristic diameter used in $Re$ is the diameter of the wire.

Kaskan[83], cited by Kaiser et al.[53], also assumed heating of a cylinder and used a relation:

$$Nu = 0.8 \times Re^{0.25},$$  \hspace{1cm} (6.2)

without specifying the range of $Re$.

Kaiser et al.[53] used a radiation correction proposed by Fristrom and Westenberg[84]. This method is based on the energy balance of a sphere, and the bead diameter is used as the characteristic diameter. Convective heat transfer to the sphere has been neglected ($Re$ very small), so $Nu = 2$. When convective heat transfer to the sphere is included, according to Bird et al.[85], the relation for the $Nu$ becomes:

$$Nu = 2.0 + 0.66 \times Pr^{0.2} \times Re^{0.5},$$  \hspace{1cm} (6.3)

for $10 < Re < 10^4$. The characteristic diameter used in $Re$ is the diameter of the bead. In our case the Reynolds number is approximately 0.4. According to (6.3) $Nu$ approximates 2 for decreasing $Re$. In our case, the difference between the correction for $Nu = 2$ and $Nu$ according to (6.3) is only 9 K at 1680 K.

### 6.2.1 Lean flame

In Figure 6.1 the corrected thermocouple measurements are shown for the lean flame ($\Phi = 0.88$), using the different correction methods given above, together
6.2. Temperature profiles

Figure 6.1: Temperatures in the lean ($\Phi = 0.88$) laminar premixed flat flame, uncorrected measurements (+), measurements assuming a cylindrical thermocouple and $Nu$ given by (6.1) (□), measurements assuming a cylindrical thermocouple and $Nu$ given by (6.2) (○), measurements assuming a spherical thermocouple and $Nu = 2$ (△), measurements assuming a spherical thermocouple and $Nu$ given by (6.3) (♦), PREMIX calculation with radiative heat losses (—) and PREMIX calculation of the maximum temperature in the absence of radiative heat losses (— —), and measurement by Schoenung and Hanson[86] in a similar flame ($\Phi = 0.86$) (×).

with the temperature profile from the PREMIX model with inclusion of radiative heat losses and calculation of post-flame temperature without radiative heat losses. Note that, because of heat losses to the burner surface by conduction, this latter temperature is not the adiabatic flame temperature (about 2200 K). Peeters[3] also calculated the temperatures at $\Phi = 0.83$ and 0.93. For $x > 2$ mm, the predicted temperatures for $\Phi = 0.83$ and for $\Phi = 0.93$ are 35 K lower and higher, respectively.

The measured temperature profiles show a temperature gradient decreasing with $x$. This concave character of the measurements is not represented by the PREMIX calculations. According to Lucht et al.[52] the temperatures near the flame zone (small $x$) may be too high because of "exothermic hydrogen recombination reactions catalyzed on the surface of the thermocouple"[52]. Regardless this catalytic effect, the temperature is bounded by the maximum temperature of the flame without radiation but with conductive heat losses to the burner surface; for $\Phi = 0.88$, this is 1895 K. This maximum temperature may be affected by the presence of a thermocouple: the temperature gradient close to the burner surface
changes when a thermocouple that catalyzes combustion reactions, is inserted in the flame very close to the burner surface (x smaller than about 2 mm); in that case, the maximum temperature in the absence of radiative heat losses in the flame changes by the presence of the thermocouple. Our measurements are obtained for x > 4 mm, so in our experiments the initial temperature gradient, and thus the maximum flame temperature, is not affected by the thermocouple in our experiments.

Due to the small velocities in the flat flame, the corrections are significant, and the results depend on the correction method and on the value of the emissivity. At a temperature of 1750 K, the emissivity used is 0.22. We find that the correction method following Kaskan[83] gives the largest correction, with corrected temperatures closest to the calculations. At x = 30 mm, the radiation correction is 83 K with this method. This yields a temperature of 1796 K. With a difference with the predicted temperature of only 46 K at x = 30 mm, the agreement is rather good.

Schoenung and Hanson[86] also measured the temperature of a laminar premixed methane/air flame, produced by a stainless steel honeycomb burner. They used an electrically heated thermocouple. Temperatures were measured at 7 mm above the burner surface, for different equivalence ratios. Flow rates have not been given. At 7 mm from the burner surface they found a temperature of 1834 K in a flame with a equivalence ratio (Φ = 0.86) comparable to our lean flame. This temperature is 37 K higher than the measured temperature (corrected with the Kaskan method) and 24 K lower than the PREMIX calculation, again rather small differences.

Comparison of the calculated and measured temperatures in the post-flame zone suggests a somewhat larger correction. As stated before, the emissivity increases with the rhodium content. With a value for the emissivity of 0.3, the corrected temperature is only 16 K smaller than the predicted temperature at x = 30 mm, and at x = 4 mm, the temperature is equal to the predicted temperature of the flame without radiation. This is the largest correction possible. Comparison with emissivities given by Touloukian and DeWitt[81] shows that a value for the emissivity of 0.3 is not unrealistic. In the range of x measured, the temperatures corrected with this emissivity, deviate less than 36 K from the predicted value, which is a very good agreement. Hence a larger value of the emissivity improves the correspondence of the measurements with the predicted temperatures. Due to an uncertainty in the value of Φ of about 3 %, the uncertainty in the temperature is about 20 K in the lean flame.

6.2.2 Rich flame

Temperature measurements in the rich flame (Φ = 1.17) have also been corrected with the method of Kaskan, and results are given in Figure 6.2, together with PREMIX calculations and the predicted maximum adiabatic temperature.

In the region covered with the measurements, the corrected temperature
6.2. Temperature profiles

Figure 6.2: Temperatures in the rich ($\Phi = 1.17$) laminar premixed flat flame, measurements assuming a cylindrical thermocouple and $Nu$ given by (6.2) (○), PREMIX calculation with radiative heat losses (—) and PREMIX calculation of the maximum temperature in the absence of radiative heat losses (— —), and measurement by Schoenung and Hanson[86] in a similar flame ($\Phi = 1.18$) (×).

measurements obtained in the rich flame do not differ more than 20 K from the temperatures obtained in the lean flame, for corresponding distance from the burner surface. The temperature in the rich flame is 20 K higher at $x = 4$ mm. It decreases faster with $x$, and becomes 13.4 K lower for $x = 26$ to 30 mm. The calculations also show that in the post-flame zone the rich flame temperature is very close to the lean flame temperature: the rich flame temperature is 13 to 0.5 K higher than the lean flame temperature for the range of $x = 2$ to 30 mm. Only for smaller $x$, the temperature differences are larger, with a maximum of about 85 K near $x = 0.75$ mm. We thus see that both in the experiments and in the calculations the temperatures of the lean and rich flame are very close, but that both the experiments and calculations show a slightly faster temperature decrease with $x$ in the rich flame. The post-flame zone temperatures do not differ more than 20 K. The differences between the calculations and the measurements are therefore systematic in character, and the discussion of the difference between the calculations and measurements in the lean flame is also valid in this case.

At 7 mm from the burner surface in a flat methane/air flame with equivalence ratio ($\Phi = 1.18$) comparable to our rich flame, Schoenung and Hanson[86] found a temperature of 1851 K, close to the temperature of 1800 K measured at $x = 7$ mm in our flame, and to the predicted temperature of 1850 K.

Calculation of the temperature with values of $\Phi = 1.12$ and 1.22, yielded
at \( x = 10 \) mm a temperature that is 2 K lower and 18 K higher than for \( \Phi = 1.17 \), respectively. Due to an uncertainty in the value of \( \Phi \) of about 3\%, the uncertainty in the flame temperature is about 10 K in the rich flame.

Both in the lean and in the rich flame, the difference between the measured and calculated temperature is about 50 K, or about 3\%. This difference is attributed to a radiation correction, which is sensitive to the thermocouple emissivity. Close to the burner surface, the measured temperatures are higher than expected, which can well be explained by exothermal reactions catalyzed by the thermocouple platinum surface. For larger \( x \), the axial temperature gradient in the calculations is close to that in the experiments. In the next section we compare the relative OH concentration measurements with the predicted OH concentrations which were obtained from the PREMIX model calculations that also yielded the temperature results.

6.3 OH concentration profiles

The calculated OH concentration increases very rapidly in the reaction zone, at about \( x = 1 \) mm reaching a peak value which is more than ten times the post-flame OH concentration, which tends to equilibrium. In a flat flame without radiative heat loss, the OH concentration becomes constant. Radiative heat loss of the flame results in a temperature decreasing with increasing distance (Section 6.2) and thus in an OH concentration decreasing with increasing distance. This decrease has also been observed by Lucht et al.[52].

The OH radical concentration profiles have been measured with single emission-line detection in the lean (\( \Phi = 0.88 \)) and in the rich flame (\( \Phi = 1.17 \)), and in the lean flame also with broadband detection. We compare measured relative OH profiles with the calculations. The ratio of the OH measurements in the lean and rich flame is compared with the ratio of the calculated OH concentrations, to check whether the effect of different equivalence ratio on the measured OH concentration is equal to the effect predicted by PREMIX. In addition, we compare the single emission-line detected measurements with the broadband detected measurements.

To decrease influence of shot noise, 10 single emission-line detected measurements have been accumulated on the diode array. The OH concentration measurements of course only yield relative values, and the results were scaled so that at \( x = 25 \) mm the measured value matched the calculated concentration (the lean flat flame OH concentration at \( x = 25 \) mm is used as reference for the turbulent flame measurements). The calculated and measured concentration profiles of the lean flame are shown in Figure 6.3 and of the rich flame in Figure 6.4.
6.3. \textit{OH} concentration profiles

Figure 6.3: \textit{OH} concentrations in the lean ($\Phi = 0.88$) laminar premixed flat flame, predicted by adiabatic calculation (---), predicted by calculation with modelled radiation losses (--), measured with single emission-line detection (+) and measured with broadband detection (○). At $x = 25$ mm the relative measurements were matched with the calculations with modelled radiation.

6.3.1 Lean flame

The single emission-line detected measurements follow the predicted profile very well, although the values are somewhat low for small $x$. This is because the exact location of the reaction zone is difficult to predict in the calculations, and a somewhat better match can be obtained when the predicted profile is shifted about 0.5 mm towards the burner surface. The broadband detected \textit{OH} measurements follow the predicted profile even better. At $x = 10$ mm, the difference between the single emission-line detected measurements and the broadband detected measurements is 17\%, and smaller than 8\% at the other locations, which is a very good agreement. The close agreement between the single emission-line detected and broadband detected \textit{OH} measurements is because in the region of $x$ covered, the temperature and major species concentrations do not change much with $x$. As a result, the quenching and rotational relaxation rates are almost independent of $x$, giving a constant relation between the \textit{OH} concentration and the fluorescence signal, see also Section 3.6.2.

The maximum in the \textit{OH} concentration profile is not found experimentally, in the lean nor in the rich flame, because this maximum is too close to the burner surface: the burner blocks the fluorescence signal beam for $x$ smaller than about 3 mm.

As discussed in Section 5.4, \textit{OH} fluorescence from the flat flame is used
to put the turbulent flame measurements on an absolute scale. For this, the fluorescence has been measured in the lean flame at 25 mm above the burner surface. At this height, OH concentration gradients are small, so the measurement is not sensitive to the exact height. The calculated concentration at \( x = 25 \text{ mm} \) in the lean flame is \( 2.13 \times 10^{15} \text{ cm}^{-3} \).

Peeters\[3\] varied the equivalence ratio with \( \pm 0.05 \). At \( x = 25 \text{ mm} \), the predicted OH concentration at \( \Phi = 0.83 \) and at \( \Phi = 0.93 \) are 13 \% smaller and 16 \% larger, respectively. We reproduced OH measurements in the flat flame five times, where the flame was set in every measurement session. The OH signal has been averaged for every session. The standard deviation obtained from this set of average values is 10 \%. The turbulent flame measurements of OH, discussed in Chapter 7, have been carried out at the same day under exactly the same circumstances, and one average flat flame OH signal has been used for the calibration in all flames. The error in the flame setting is random in origin, which gives an OH concentration that may differ with about 10 \% from the predicted value. In the calibration procedure this error is systematic and improves comparison of the OH concentrations for different turbulent flame settings.

### 6.3.2 Rich flame

In the rich flame, the relative OH measurements obtained with single emission-line detection, follow the predicted profile very well, even better than in the lean flame. According to the PREMIX calculations, the decrease of OH concentration with increasing \( x \) is faster in the rich flame, and this is also shown by the measurements. In the rich flame the predicted OH concentrations are smaller at all radial locations, which is expected from the smaller amount of oxygen provided in the rich flame.

At \( x = 25 \text{ mm} \), the predicted OH concentration is 5.8 times smaller than in the lean flame at the same location. The ratio of the OH measurements at \( x = 25 \text{ mm} \) in the lean and rich flame is 6.8, which differs 17 \% from the predicted value, a value that is in agreement with the combined errors in both flame settings.

Calculation of the flame with \( \Phi = 1.12 \) and 1.22, yielded OH concentrations which are 26 \% larger and 15 \% smaller, respectively, than for \( \Phi = 1.17 \).

### 6.3.3 Comparison with literature

For different equivalence ratios, Cattolica\[87\] measured OH concentrations in a flat methane/air flame; Kaiser et al.\[53\] measured OH concentrations in a stoichiometric methane/air flame. Both used laser absorption to measure OH. According to Kaiser et al.\[53\], the OH concentrations of Cattolica are systematically too high, because of the definition of the path length in the absorption measurements. Cattolica\[87\] found that OH concentrations in a \( \Phi = 0.9 \) flame were less than 8 \% higher than in the stoichiometric (\( \Phi = 1.0 \)) flame in the range \( x = 3 \)
6.4 Distribution of single emission-line detected OH in the lean laminar flat flame

Figure 6.4: OH concentrations in the rich (Φ = 1.17) laminar premixed flat flame, predicted by adiabatic calculation (---), predicted by calculation with modelled radiation losses (--), measured with single emission-line detection (+). At x = 25 mm the relative measurements were matched with the calculations with modelled radiation.

to 10 mm. Therefore, the OH concentration in a stoichiometric flat flame presented by Kaiser et al., should be close to our lean flame results. At 3 mm, the OH concentration found by Kaiser et al.[53] was $8.4 \times 10^{15}$ cm$^{-3}$, compared to $9.6 \times 10^{15}$ cm$^{-3}$ in our lean flame, a difference of 14%. At 10 mm, Kaiser et al.[53] found a concentration of $4.0 \times 10^{15}$ cm$^{-3}$, compared to a concentration we found of $3.6 \times 10^{15}$ cm$^{-3}$, respectively, a difference of 10%. Taking the difference in equivalence ratio into account, the OH concentrations, calculated with PREMIX with radiative heat losses included, are close to the OH concentrations measured by Kaiser et al.[53] and, in addition, the profiles match very well with our relative OH measurements, both with single emission-line detection and broadband detection.

6.4 Distribution of single emission-line detected OH in the lean laminar flat flame

In the single emission-line detection setup, signal levels are very low, because of the use of many mirrors and a large, double spectroscope. Due to the low signal level, the single emission-line detected signal fluctuates more strongly due to shot noise. The signals obtained with the broadband detection setup are much
Chapter 6. Flat flame measurements

Figure 6.5: Distribution of a set of 500 single shot OH signal measurements in the lean ($\Phi = 0.88$) laminar flame ($\cdots$), obtained with the single emission-line detection setup at 5 axial locations, and Poisson distribution functions determined by the average signal ($\cdots$).

larger, and the standard deviation of the OH flat flame signal is 8 % at $x = 25$ mm. With single emission-line detection the standard deviation ranges from 46 to 100 %, depending on the signal level, and in this section we show, that the single emission-line detected OH signal follows a Poisson distribution, as expected for shot noise. We measured the concentration profile of the lean flat flame at 5 axial locations, storing 500 single shot measurements.

The camera is cooled to about $-20$ °C to suppress dark current noise. We measured the dark current noise by observing the standard deviation of the camera signal without light and we found this noise to contribute to the standard deviation of the laminar flame signal with about 5 %. As a result, the main noise source left is shot noise.

According to Eckbreth[24], shot noise is the result of the probabilistic process of photo cathode emission, and is Poisson distributed. For a photo multiplier tube, the standard deviation in the number of detected electrons is equal to the square root of this number of electrons. Consequently, the signal to noise ratio increases with the square root of the signal. In our case with a phosphor plate and photo diode instead of a anode to detect electrons, the signal level is not equal to the number of electrons that hit the phosphor plate. We therefore multiplied the signal with a factor 3, with which an excellent agreement is obtained between the measured distribution functions and the Poisson distributions at all 5 axial locations, see Figure 6.5. It should be noted that the broadening of
the distribution of the signal due to shot noise does not affect the average.

In another series of measurements where 10 shots were accumulated, the signal was 10 times higher, whereas the standard deviation was only $\sqrt{10}$ times higher, which is in agreement with the Poisson distribution.

6.5 Conclusions

Temperature measurements in lean and rich laminar premixed flat flames have been found to be sensitive to radiation correction, because of the small velocities in this type of flame. Profiles are concave in shape, probably because of catalytic effects. In the post-flame zone, the measurements corrected with the method proposed by Kaskan[83] and with an emissivity of 0.3, are less than 36 K smaller than the predicted values. In both flames, the predicted temperature gradients due to radiative heat losses correspond to the experimental gradients, when not too close to the burner surface. Both in the measurements and in the calculations, the post-flame temperatures in the rich flame did not differ more than 20 K from those in the lean flame.

Relative OH concentrations, in both the lean and rich flame, and obtained with both the single emission-line detection and broadband detection, fitted well to the calculated profiles. The OH concentration in the lean flame at $x = 25$ mm is used as a reference for the turbulent OH signals, and according to the PREMIX calculations this concentration is $2.13 \times 10^{15}$ cm$^{-3}$. The flame setting errors result in a 10 % change in the reproduced OH concentration measurement. This agrees with the difference in measured and calculated ratio of lean and rich flame OH concentration. In the calibration procedure a small but unavoidable deviation from the reference concentration is systematic in character.
Chapter 6. Flat flame measurements
Chapter 7

Temperatures and OH concentration fluctuations in turbulent flames

7.1 Introduction

In this chapter, we discuss the average temperature and the turbulent fluctuations of OH in flames under different conditions, given in Table 4.2. Measurements of OH in turbulent nonpremixed flames with one- and two-dimensional detection reported in literature have been discussed in Section 5.2.

This chapter is organized as follows. We discuss main flame characteristics, like flame length and width. Based on instantaneous, two-dimensional images of OH, we discuss the influence of fuel and air flow rates and exit flow temperature on OH in the flame. The qualitative observations of this section will be quantified with one-dimensional OH measurements, and expressed in radial profiles of average OH concentration and standard deviation, in concentration distributions and in instantaneous gradients. Results of average temperatures, measured with thin thermocouples, are presented, and compared with the average OH concentrations. From the one-dimensional OH profiles, concentration distributions relative to the instantaneous position of the OH zone have been derived, showing the fluctuations of peak OH concentration. The OH zone width in the different flames was measured, and the largest scales transporting OH. More information on the spatial structure of OH concentrations is obtained from joint distribution functions of concentration and gradient and from spatial correlation of the one-dimensional OH profiles. From spatial correlation functions length scales of turbulent OH fluctuations have been derived, both for the large and the small scale structure of turbulence.

7.2 Results of main flame characteristics

In this section, we discuss the global flame characteristics visible flame length and flame width in relation to the different fuel and primary air velocity combinations and exit flow temperature.
7.2.1 Visible flame length

The flame length was based on the average location of the visible flame tip, which was yellow in all flames, though faint in flame IV. According to Glassman[7], the yellow flame emission is from soot burning in the reaction zone. Burn out of all soot takes some time, so the visible flame length based on the yellow emission is somewhat larger than the actual flame length based on the location of the reaction zone[7]. Due to the fluctuating character of location of the flame tip, we expect the accuracy of length measurement to be in the order of 0.05 m. We will relate the observed flame length to the flow rates. The visible flame lengths, designated with $L_f,\text{vis}$, are given in Table 7.1.

Observing flames I, II and III, with equal air excess values and hence with equal ratio $U_j/U_a$, we find that the differences between the lengths of flames I–III are not significant, in agreement with the turbulent flame length predicted by (2.7). According to (2.7), the flame is shorter when the primary air exit velocity, $U_a$, is larger. In flame III and IV, $U_j/U_a = 5.0$ and 2.7, respectively, so the length of flame IV should by about half that of flame III. We find that the visible flame length of flame IV indeed is significantly shorter; the difference between the visible flame lengths $L_f,\text{vis}$ of flames III and IV is smaller than between the flame lengths $L_f$ of these flames predicted by (2.7). This is because in (2.7) the turbulent diffusion of air into the fuel jet was assumed only to depend on the primary air flow velocity fluctuations and not on central jet velocity fluctuations, and this assumption will be less valid in flame III than in flame IV. Still, we find that the higher primary air exit velocity increases turbulent diffusion of air into the fuel jet.

Driscoll et al.[48] presented flame length measurements of flames with a methane jet ($d_j = 3.7$ mm, $Re_j = 5 \times 10^3$), coaxial air issuing from an annulus (outer annulus diameter was 8.7 mm, inner annulus diameter not specified), and low velocity secondary air. They found a reduction of 10 % in flame length when the ratio of primary air exit velocity to fuel exit velocity was raised from 0.20

Table 7.1: Visible flame length $L_f,\text{vis}$ for flames I to VI, and the visible flame length multiplied by $d_a/d_a^*$ to correct for the difference in jet development of the preheated flames V and VI. The accuracy is about 0.05 m.

<table>
<thead>
<tr>
<th>flame</th>
<th>$L_f,\text{vis}$</th>
<th>$L_f,\text{vis}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>0.87</td>
<td>-</td>
</tr>
<tr>
<td>II</td>
<td>0.90</td>
<td>-</td>
</tr>
<tr>
<td>III</td>
<td>0.85</td>
<td>-</td>
</tr>
<tr>
<td>IV</td>
<td>0.70</td>
<td>-</td>
</tr>
<tr>
<td>V</td>
<td>0.67</td>
<td>1.00</td>
</tr>
<tr>
<td>VI</td>
<td>0.52</td>
<td>0.79</td>
</tr>
</tbody>
</table>
(like in flame III) to 0.36 (like in flame IV). This decrease is somewhat smaller than the difference in flame length between flame IV and III: flame IV is 18% shorter than flame III.

The heated flames V and VI have fuel and primary air mass flow rates equal to flames III and IV respectively. Preheating of the flows accelerates both fuel and primary air exit velocity. According to (2.7), this acceleration should not change the flame length. However, we also noticed that the turbulent diffusion of a heated jet is larger, as if the flow were from a smaller diameter (i.e. the effective diameter) jet. In the preheated flames, the effective annulus hydraulic diameter, given by (2.8), $d_a^*/d_a = 0.67$. Hence, the flame length in the case of preheated flows is to be corrected to be comparable with the length of the flames that are not preheated. The corrected flame length is given by $L_{f,vis} = L_{f,vis}d_a/d_a^*$, which is about 1.5 times $L_{f,vis}$. These corrected values are also given in Table 7.1.

The corrected length of flame V is somewhat larger than of the nonpreheated flame with equal mass flow rates, flame III, because the primary air velocity fluctuations not only determine the turbulent diffusion of air into the fuel jet. The corrected length of flame VI, with larger primary air velocity, is close to that of the nonpreheated flame with equal mass flow rates, flame IV, indicating that the flame lengths of the flames with high primary air velocity, flames IV and VI, are mainly determined by the primary air velocity fluctuations. Note that, although preheating of the flows increases flame temperatures and consequently affects chemical reaction rates, the flame length of a turbulent nonpremixed flame is determined by turbulent diffusion and not by chemistry.

The above results can be summarized as follows. Increase of the primary air velocity decreases the flame length. The turbulent diffusion of air into the fuel jet is not only determined by the eddy viscosity of the primary air, so the influence of the primary air velocity on the flame length is not as strong as predicted by (2.7). The preheated flames are shorter than the nonpreheated flames. Correction with the ratio of the effective and real annulus diameter shows that this is due to the larger turbulent diffusion of the preheated primary air jet issuing in a cold secondary air flow.

### 7.2.2 Flame width $w_{OH}$

We define the flame width $w_{OH}$ as the radial distance between the locations of maximum average OH concentration, obtained from one-dimensional measurements of OH. Results are shown in Figure 7.1. Visible flame width is strongly affected by concentration and temperature fluctuations. Unlike the visible flame width, the flame width based on the separation of the maximum OH concentration locations, can be related to the model presented in Section 2.2, which is used to predict the average location of the reaction zone (Figure 2.1).

It will be discussed in Section 7.8.4 that the location of maximum average OH concentration corresponds to the average location of the OH zone, and the average location of the OH zone approximately coincides with the average location
Figure 7.1: Flame width based on the distance between the radial locations of maximum average OH concentration, $w_{OH}$, for flame I ($\circ$), II ($\triangle$), III ($\square$), IV ($\diamond$), V ($\cdot\cdot\cdot\cdot\cdot\cdot$) and VI ($\cdot\cdot\cdot\cdot\cdot\cdot$).

of the reaction zone. Measurements have been obtained for axial distance up to 300 mm, or 50 fuel jet diameters. The accuracy with which $w_{OH}$ was determined is 5%, or about 1.5 mm at $x = 300$ mm.

The exit jet velocity $U_j$ of flame II is larger than that of flame I, and this gives a wider flame, despite the equal ratio $U_j/U_a$. For a fully developed turbulent jet, the jet spread depends on the velocity ratio $U_j/U_a$. The turbulence of flames I is not fully developed. This is discussed more extensively in Chapter 8. For the transitional, smaller Reynolds number flames the entrainment increases with $U_j$, even when $U_j/U_a$ is kept constant. The entrainment increases with increasing $U_j$, which is larger in flame II than in flame I. As a result, flame II is wider than flame I. The increased entrainment moves the outer boundary of the shear layer away from the jet axis, and in Section 2.2 it is shown that this is also the case for the average location of the reaction zone. The increased entrainment giving a wider shear layer, results therefore in a wider flame. The flame III is slightly wider than flame II, indicating that also the jet of flame II is not completely developed.

We find that flame IV is somewhat narrower than flame III. In Section 8.9 we show that increase of the primary air velocity reduces the integral length scales in the fuel jet. The large primary air velocity decreases the size of the large shear layer eddies that are mainly responsible for the entrainment and hence for the flame width. The preheated flame with large primary air velocity, flame VI, is significantly narrower than the flames II–V. The influence of preheating appears not to be very large in the flames with smaller primary air mass flow rate, compare
the width of flames III and V.

7.3 Estimation of Damköhler numbers

A characteristic turbulence time scale in the flame close to the nozzle can be calculated with the large-eddy turnover time given in (2.10) and the average primary exit velocities of the different flames, given in Table 4.2. The primary air velocity has a strong influence on the reaction zone. Close to the nozzle, the jet width in (2.10) is in the order of magnitude of the fuel nozzle diameter. For axial locations close to the nozzle, the turbulence time scales are presented in Table 7.2.

In Section 2.3.4 we presented estimations of the order of magnitude of the two-body chemistry time scale $\tau_{ch2}$ for the flames with initially cold flows and for the preheated flames. With the turbulence time scales and the chemistry time scales the Damköhler number $Da_2$ can be calculated, which is also shown in Table 7.2. In the flames that are not preheated, flames I to IV, $Da_2$ decreases with increasing primary air exit velocity. In flame IV both turbulence time scales are smallest, resulting in a very small value of $Da_2$. According to Correa and Gulati[17], local extinction should be most pronounced in the smallest $Da_2$ flame. We also see that preheating decreases chemistry time scales with a larger amount than the turbulence time scales, so preheating is expected to increase $Da_2$ and hence to suppress local extinction, compare the values of $Da_2$ of flames III and V, and of IV and VI.

Table 7.2: Estimated turbulent time scales derived from (2.10), based on the average primary air exit velocities $U_a - U_c$ and the fuel nozzle diameter $d_j$. The two-body chemistry time scales $\tau_{ch2}$ are based on laminar flamelet extinction calculations. The two-body chemistry Damköhler number $Da_2$ is the ratio of $\tau_1$ and $\tau_{ch2}$.

<table>
<thead>
<tr>
<th>flame</th>
<th>$\tau_1$ (ms)</th>
<th>$\tau_{ch2}$ (ms)</th>
<th>$Da_2$ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>3.16</td>
<td>2.5</td>
<td>1.26</td>
</tr>
<tr>
<td>II</td>
<td>2.14</td>
<td>2.5</td>
<td>0.86</td>
</tr>
<tr>
<td>III</td>
<td>1.46</td>
<td>2.5</td>
<td>0.58</td>
</tr>
<tr>
<td>IV</td>
<td>0.78</td>
<td>2.5</td>
<td>0.31</td>
</tr>
<tr>
<td>V</td>
<td>0.56</td>
<td>0.5</td>
<td>1.10</td>
</tr>
<tr>
<td>VI</td>
<td>0.30</td>
<td>0.5</td>
<td>0.60</td>
</tr>
</tbody>
</table>
7.4 Qualitative observations on instantaneous 2D OH images

We now make qualitative observations on the OH structure in the six different flames from instantaneous two dimensional OH images. We refer to these observations in the next sections, to match the quantitative results from one-dimensional measurements with these images.

In Figures 7.3–7.8 we show images of OH for the regions $x = 15$ to 93 mm, 100 to 178 mm and 185 to 263 mm. Of all six flames we give two sets of images, measured at different times. It should be noted that there is no correlation between the images covering different regions, grouped to represent one flame, and between the images covering the same region. The instantaneous images of OH concentration show a cross section along the jet centerline of the (on average) axisymmetric flame. This results in two separate OH zones which may be wrinkled or curled, and which sometimes are interrupted.

7.4.1 Soot interference

Sharp dark streaks are found in the images covering $x = 100$ to 263 mm of flame I. An example of this is shown in Figure 7.2. From about $x = 150$ mm, flame I is bright-yellow due to the presence of soot. In images of flame emission these streaks were not visible. Hence, the signal of the streaks is induced by the laser light. According to Puri et al. [88] this can be attributed to laser scattering of soot or PAH fluorescence. The laser pulse duration is very short and the images are frozen. This implies that the streaks in the images are the result of scattering from large clusters of particles of about 10 mm long. The scattering of soot was also observed by Dasch and Heffelfinger [89] in an ethylene flame. They found that "the soot tends to occur in oblong patches oriented in the streamwise direction" [89], and that the size of these patches was in the order of magnitude of 25 mm. This corresponds to our observations, see Figure 7.2. In our case, the OH structures can usually be discriminated from the the soot interference, which for $x < 263$ mm is present only in flame I.

7.4.2 Deformation of the OH structure

We relate the model for the interaction between the reaction zone and the shear layers (Sections 2.2 and 2.3) to the instantaneous images of OH (Figures 7.3–7.8) for flames I–VI, respectively. In Section 2.2 we noticed that close to the nozzle, the reaction zone and the OH are found near the outer boundary of the shear layer, so not a strong influence of the shear layer turbulence was expected in this region. (This shear layer is visualized by detecting NO seeded to the fuel flow, and is discussed in Section 8.4.) Flames I, II and III have jet Reynolds numbers ranging from 4.9 to 9.7 $\times$ 10³. Small scales exist in the flows because both fuel and primary air are turbulent at the burner exit. These scales are essential for
7.4. Qualitative observations on instantaneous 2D OH images

Figure 7.2: Instantaneous images of OH in flame 1, covering $x = 100$ to $178$ mm (a) and $x = 185$ to $263$ mm (b). The arrows indicate the streaks due to soot interference.

The small-scale mixing between fuel and air. These scales, however, are too small to be visible in the images. Deformations in the OH zone in these flames that are visible in these images are therefore mainly of large scale.

In Section 7.2 we discussed the effect of preheating the flows on main flame characteristics, like length and width. It was concluded that the preheated flames V and VI are more turbulent than their initially cold counterparts with equal mass flow rates, III and IV, respectively.

Heating accelerates the fuel and primary air flows but decreases the Reynolds numbers based on the exit diameter, exit velocity and viscosity at exit temperature. OH is found in regions with high temperature and in those regions the relative temperature rise due to heating is much smaller than the relative temperature rise of the air at the burner exit, remote from the hot reaction zone. Near the reaction zone, the effect of primary air temperature on the viscosity are therefore not very large: viscosity of air increases with a factor of 5 when temperature is raised from 300 to 700 K, but with about 15 % when flame temperature is raised from, say, 2000 to 2300 K. When a Reynolds number is defined using local properties the effect of the larger viscosity on this local Reynolds number is small. The effect of the increased velocity is much stronger.

The OH zone is deformed by turbulence at the location of the reaction zone. In flames I and II the deformation of the OH zone is mainly large scale, and this only for larger $x$. This is because of the large separation between the reaction zone and the shear layer, see Chapter 2. In flames IV–VI the OH zone is strongly deformed by scales of different sizes. A high level of turbulence is associated with a wide spectrum of scales.

We can arrange the instantaneous images in order of increasing level of turbulence deforming the OH structure, and the order of the flames is I, II, III, IV,
Figure 7.3: Instantaneous OH images of flame I, covering the regions $x = 15$ to 93 mm, 100 to 178 mm and 185 to 263 mm, all taken at different moments. Two images at different moments are shown for every region.
7.4. Qualitative observations on instantaneous 2D OH images

Figure 7.4: Instantaneous OH images of flame II, covering the regions $x = 15$ to $93$ mm, $100$ to $178$ mm and $185$ to $263$ mm, all taken at different moments. Two images at different moments are shown for every region.
Figure 7.5: Instantaneous OH images of flame III, covering the regions $x = 15$ to $93$ mm, $100$ to $178$ mm and $185$ to $263$ mm, all taken at different moments. Two images at different moments are shown for every region.
Figure 7.6: Instantaneous OH images of flame IV, covering the regions $x = 15$ to $93$ mm, $100$ to $178$ mm and $185$ to $263$ mm, all taken at different moments. Two images at different moments are shown for every region.
Figure 7.7: Instantaneous OH images of flame V, covering the regions $x = 15$ to 93 mm, 100 to 178 mm and 185 to 263 mm, all taken at different moments. Two images at different moments are shown for every region.
7.4. Qualitative observations on instantaneous 2D OH images

Figure 7.8: Instantaneous OH images of flame v1, covering the regions \( x = 15 \) to 93 mm, 100 to 178 mm and 185 to 263 mm, all taken at different moments. Two images at different moments are shown for every region.
v, VI, although the order of flames V and IV is disputable. This order corresponds
to the order of increasing primary air exit velocity. The primary air velocities of
flames V and IV are comparable. We notice that the primary air exit velocity has
a large influence on the turbulent deformation of the OH zone, and it is a better
indication for this deformation than the Reynolds number based on the velocity
and viscosity at the annulus exit. The order of increasing turbulence of flames III,
v, IV and VI is in agreement with the order based on the flame length of these
flames, discussed in Section 7.2.1. This is because a stronger wrapping-up of the
flame surface results in a shorter flame.

7.4.3 Holes in the OH zone and local extinction
It is found that in many cases, the OH zones are interrupted, see for example
flame IV for the range 15 < x < 93 mm (Figure 7.6). In three dimensions, the
essentially tubular shaped OH region is deformed by turbulence. This OH region
cannot be deformed in such a way that, when intersected by a plane that coincides
with the centerline axis, the cross section is interrupted. Hence, these interrupts
are the indication of a hole in the OH zone. According to Seitzman et al.[65] and
Chen and Goss[10] a hole in the OH zone implies a hole in the reaction zone and
is the indication of local extinction (Section 2.3.5): locally OH is not produced.
Local flame extinction occurs when the local gradient of the mixture fraction
(or, equivalently, the scalar dissipation) becomes so large, and the reaction zone
becomes so thin, that not enough heat is produced to compensate for the loss of
heat.

Holes in the OH structure, due to local extinction, are observed in flames
III, IV, and to a much lesser extent in the preheated flames V and VI. The smallest
Reynolds number flames I and II show little or no holes in the OH structure.

Williams[15] was not sure whether flamelet re-ignition could occur and if
it could, in what way. Observation of many images of flame III showed that the
number of holes decreases with the axial distance in this flame. In Section 2.3.5
it was concluded that local extinction is connected to the Damköhler number
of two-body reactions, Da2. The turbulent time scales increase with axial distance
and the premixing of the fuel and oxidizer with hot combustion products results
in chemistry time scales decreasing with x. Da2, therefore, increases with axial
distance. This implies that the formation of holes becomes less probable with
axial distance. Holes that are convected are expected to grow with axial distance
when no re-ignition would take place. The decreasing number of holes with axial
distance we have observed, therefore may indicate that extinct flamelets can be
re-ignited, probably by the hot combustion products which are present in larger
amounts at larger axial distance.

In Section 2.3.5, we found that the two-body chemistry time scale in-
creases with a factor of about 5 due to preheating the flows up to 700 K. On the
other hand, the increased velocities due to preheating, gives, according to (2.10),
a turbulence time scale decreased with a factor of 2.4. The overall effect on the
7.5. Results of temperature measurements

Damköhler number \( Da_2 \) is that it increases by preheating when the mass flow rates are kept constant, see also Section 7.3. This makes that \( Da_2 \) of flame V is larger than of flame III, and \( Da_2 \) of flame VI is larger than of flame IV. Indeed, the OH structure of flame VI shows much fewer holes than that of flame IV. This is also the case for flame V, compared to flame III, but this difference is less prominent.

When the initial flow temperature is kept constant, close to the nozzle the chemistry time scale is the same for all nonpreheated flames, where there are few combustion products to mix with the fuel and oxidizer. In that case, a decrease in the turbulence time scale gives a decrease in the Damköhler number. Comparing flames III and IV, \( Da_2 \) of flame III is larger than that of flame IV, which corresponds to the much more frequent local extinction in flame IV. In Section 7.3, the Damköhler number of flame IV was found to be smallest.

When we arrange the six flames in order of increasing \( Da_2 \) based on the local extinction observed in the flames, this order is IV, III, VI, V, II and I. Except for the place of flames I and II in this order, it agrees well with the Damköhler numbers presented in Table 7.2. This difference is probably due to the smaller influence of the primary air velocity on these flames, because the turbulence time scale was based on the primary air exit velocity.

7.4.4 Broad OH regions

In flames I and II (Figures 7.3 and 7.4) in the regions with the largest \( x \) covered, we observe broad regions of OH. This is attributed to a not perpendicular crossing of the OH structure with the laser sheet. In addition, the reaction zone has broadened by diffusion. According to Peters[18], a characteristic time scale of diffusion, determining the width of the reaction zone of a laminar diffusion flame, is the inverse of the dissipation rate, which is proportional to the strain rate, or the local velocity gradient. The local velocity gradients decrease most quickly with \( x \) in the small Reynolds number flames I and II, so the broadening is most clear in these flames.

In flames IV, V and VI, we also find OH in extended regions, but, unlike in flames I and II, the thin, large concentration regions are still present. These extended regions are attributed to pockets with hot combustion products separated from the reaction zone by the strong turbulence in these flames.

7.5 Results of temperature measurements

In this section we discuss average temperature results, measured with thin thermocouples. These temperatures are related to the OH concentration results in a following section.

Average temperatures have been measured with an uncoated 50 \( \mu \text{m} \) Pt/Rh(6\%)-Pt/Rh(30\%) thermocouple. At axial locations of 50, 100, 150, 200
Figure 7.9: Radial average temperature profiles of flame I, at $x = 50$ mm (--), 100 mm (---), 150 mm (----), 200 mm (--), 250 mm (-- ---) and 300 mm (--).

Figure 7.10: Radial average temperature profiles of flame III, at $x = 50$ mm (--), 100 mm (---), 150 mm (----), 200 mm (--), 250 mm (-- ---) and 300 mm (--).
7.5. Results of temperature measurements

Figure 7.11: Radial average temperature profiles of flame IV, at $x = 50$ mm (---), 100 mm (--), 150 mm (-----), 200 mm (--), 250 mm (~---) and 300 mm (---).

Figure 7.12: Radial average temperature profiles of flame V, at $x = 50$ mm (---), 100 mm (--), 150 mm (-----), 200 mm (--), 250 mm (~---) and 300 mm (---).
and 250 mm, radial traverses have been made with a spatial resolution of 1 mm. The thermocouple wire was oriented in radial direction, in order to have a flow at the thermocouple unaffected by the thermocouple holder. Because of this orientation, correction for conductive losses was necessary. The temperatures were corrected for conductive and radiative losses of the thermocouple, using the heat balance of a cylinder, following the method of Bradley and Matthews[80]. The velocity used in this correction was the average axial velocity. The average was taken of 100 measurements by a digital multimeter, connected to the UNIX computer system. The accuracy of the measurements is about 50 K.

The average temperature profiles of flames I and III–VI are shown in Figures 7.10 to 7.13. The absolute value of the radial location $y$ is equal to $r$, where the positive and negative sign correspond to tangential coordinates $\varphi = 0$ and $\pi$, respectively. In the smallest Reynolds number flame, flame I, the maximum average temperature is high for small $x$, 1946 K at $x = 50$ mm. This is due to the shear stresses being smaller than in the other flames, giving a wider reaction zone with higher peak temperature, and due to the absence of local extinction. Hot product gases from the pilot flames, that are transported more slowly in the smaller Reynolds number flame, may also contribute to the high temperatures close to the nozzle. In flame I, the maximum average temperature decreases with $x$. The fluctuations in the location of the reaction zone increase, resulting in lower average temperatures in a wider range of radial locations. Also, in the bright-yellow parts of flame I (for $x \geq 150$ mm) temperatures decrease due to
heat losses by soot radiation.

In flame III, with Reynolds number of fuel and primary air at the burner exit twice as large as in flame I, the maximum average temperature is lower for small $x$, ranging from 246 K at $x = 50$ mm to 41 K at $x = 200$ mm, and is 16 K higher at $x = 250$ mm, a difference that is not significant. This difference for small $x$ is probably due to a thinner reaction zone, and due to local extinction. In flame III the maximum average temperature slightly increases (less than 35 K) with axial distance for $x \geq 100$ mm.

Temperature measurements have also been reported by Sislian et al.[12] for a methane flame issuing in still air ($d_j = 10$ mm, $Re_j = 7.9 \times 10^3$). The burner nozzle was contoured and there were no pilot flames. The jet exit Reynolds number of this flame is comparable with our flame II, so we compare these temperatures with those in our flames I and III. Their measurements are only corrected for radiation and are therefore lower than our measurements, corrected for both radiation and conduction losses. For the range $50 \leq x \leq 250$ mm, the maximum average temperature reported by Sislian et al. is always lower (in the range 40 to 120 K) than in flame I, when our measurements are also only corrected for radiation losses; the maximum average temperature of [12] decreases with $x$, like in flame I. The temperatures of [12] are between those of flame I and III, as expected for a flame with exit Reynolds number close to that of flame II.

Due to the large primary air velocity, flame IV exhibits strong local extinction, most frequent around $x = 100$ mm. At this location, the maximum average temperature in flame IV is 377 K lower than in flame III; at $x = 50$ mm this difference is 310 K. For $150 \leq x \leq 250$ mm, the difference decreases from 257 to 97 K. Temperatures in flame IV remain lower than in flame III for this range of $x$, due to the large excess of air that is only heated but does not take part in the combustion.

The average temperature profiles of the preheated flames V and VI show that both the fuel and air exit temperatures are about 700 K. This result has already been used in Section 4.7. Despite preheating, the maximum average temperature of flame V is not much higher than of flame III, due to the much larger velocities in the preheated flame. The instantaneous images of OH of this flame (Figure 7.7) showed fluctuations of OH that are larger in flame V than in flame III, so we expect that also the temperature fluctuations are larger in this flame V than in flame III. This reduces the maximum average temperatures. For $x \geq 100$ mm, the maximum average temperature in flame V is larger than in the nonpreheated flame with equal mass flow rates, flame III, and the temperature difference becomes almost constant, (158 ± 10) K.

The preheated flame VI, with large primary air velocity, shows that flame VI is narrower than the other flames and that at $x = 250$ mm, the two maxima in the profile have almost merged, suggesting an almost homogeneous turbulence near the jet axis. Due to the larger primary air exit velocity in flame VI, the temperatures are lower than in the other preheated flame, flame V, but this
difference is not very large. The difference in temperatures between flames III and IV is much larger than between the preheated counterparts, flames V and VI. Hence, influence of the primary air velocity on the temperature is smaller when the flows are preheated.

7.6 Average OH concentrations

In the following sections we discuss the results of the one-dimensional detection experiments. We discuss the average concentration profiles of the different flames. The average OH concentration profiles of flames I–VI are shown in Figures 7.14–7.19. The maximum average OH concentrations are compared with the maximum average temperatures at the corresponding axial locations.

7.6.1 General structure of the average profiles

The maximum average concentrations at positive values of $y$ are somewhat larger due to a small effect of laser absorption in combination with not fully saturated fluorescence. In flames with broad average profiles, like flames IV to VI at larger axial distance, the profiles are more skewed due to the larger amounts of OH absorbing the laser light.

Whereas flames II and III do not show a significant increase in OH centerline concentration for $x$ up to 300 mm, an unexpectedly high signal is found for $x \geq 150$ mm in the fuel rich part of flame I. The large average centerline signals are due to soot scattering or PAH fluorescence, also discussed in Section 7.4.1, and should therefore be neglected. We expect that the OH concentrations remain very small for these axial distances corresponding to flames II and III.

Finite centerline concentrations for $x \leq 300$ mm are found in the flames with the strongest deformation of the OH zone, according to the instantaneous images: the flame with the largest primary air velocity, flame IV, the preheated flames with large primary air velocity, flame VI, and to a lesser extent the preheated flame with smaller primary air velocity, flame V. This indicates that the reaction reaches the jet axis in these more turbulent flames. At $x = 300$ mm, the OH concentration in flame VI is almost homogeneously distributed, whereas the temperature measurements showed that in the temperature profile the two maxima have already merged at $x = 250$ mm.

7.6.2 Effect of the primary air velocity on the average concentration

Comparing the average concentration profiles of the nonpreheated flames (flames I to IV) we find that close to the nozzle the maximum average concentration decreases with increasing fuel and primary air velocity for flames I to III, and decreases even further due to an extra increase of primary air velocity (flame IV). In the following sections it is shown that this is due to increasing relative fluctuations and increasing occurrence rate of holes in the OH structure.
7.6. Average OH concentrations

Figure 7.14: Radial average concentration profiles of flame I, at $x = 50$ mm (---), 100 mm (--), 150 mm (---), 200 mm (---), 250 mm (---) and 300 mm (--).

Figure 7.15: Radial average concentration profiles of flame II, at $x = 50$ mm (---), 100 mm (--), 150 mm (---), 200 mm (---), 250 mm (---) and 300 mm (--).
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Figure 7.16: Radial average concentration profiles of flame III, at $x = 50$ mm (—), 100 mm (− −), 150 mm (— —), 200 mm (− − −), 250 mm (-----) and 300 mm (—).

Figure 7.17: Radial average concentration profiles of flame IV, at $x = 50$ mm (—), 100 mm (− −), 150 mm (— —), 200 mm (− − −), 250 mm (-----) and 300 mm (—).
7.6. *Average OH concentrations*

Figure 7.18: Radial average concentration profiles of flame v, at $x = 50$ mm (--), 100 mm (-- -), 150 mm (——), 200 mm (-- --), 250 mm (-----) and 300 mm (---).

Figure 7.19: Radial average concentration profiles of flame v1, at $x = 50$ mm (--), 100 mm (-- -), 150 mm (——), 200 mm (-- --), 250 mm (-----) and 300 mm (---).
Figure 7.20: Comparison of the profiles of average OH concentration and temperature in flame III at $x = 50$ mm (——) and 250 mm (—–). Lower curves represent the average OH concentration, upper curves the average temperature.

At larger axial distance, the maximum average concentrations of flames I–IV differ less than 35%. The difference in average OH concentration between flames I to IV is therefore most dominant close to the nozzle: at $x = 50$ mm the maximum average OH concentration of flame IV is a factor 4 smaller than that of flame I.

### 7.6.3 Effect of preheating on the average concentration

When we compare the preheated flames, V and VI, with the nonpreheated flames with equal fuel and primary air mass flow rates, flames III and IV respectively, we find that mainly close to the nozzle, the average concentrations in the preheated flames are higher: at $x = 50$ mm 28% larger in flame V than in flame III, and 1.4 times larger in flame VI than in flame VI. This difference close to the nozzle is primarily due to the much less frequent local extinction, which was already observed in the instantaneous two-dimensional OH images.

### 7.6.4 Comparison of profiles of average OH concentration and temperature

OH is found in the regions with highest temperatures, and therefore the profiles of average OH concentration and temperature are very similar. We compare the profiles of OH and temperature in flame III at $x = 50$ mm and 250 mm, shown in Figure 7.20.
Figure 7.20 shows that the maxima of OH and temperature are at the same radial location, but that for larger $x$ the radial gradient of the average temperature is smaller than that of the average OH concentration. We find that the centerline average OH concentration is very close to zero at $x = 250$ mm, whereas the centerline average temperature has reached 1166 K.

7.6.5 Maximum average OH concentration versus temperature

The maximum average temperature and the maximum average OH concentration at different axial locations are shown in Figures 7.21 and 7.22 for the six different flames. Because high temperature and high OH concentration are both expected in the (narrow) reaction zone, and because the average OH concentrations and temperatures are strongly affected by turbulent fluctuations, correspondences between the maximum average temperature and maximum average OH concentration are expected, but the differences are even more interesting.

In flame I, both the maximum average OH concentration and the maximum average temperature decrease with axial distance. In the discussion of the concentration distribution functions (Section 7.8) we show that this decrease of OH concentration is due to increasing fluctuations of the location of the reaction zone, which are very small for small $x$. This is probably also the case for the temperature, although there are no temperature fluctuation measurements available.

We find that at larger $x$ the maximum average OH concentration of all flames do not change much with $x$ and do not differ more than about 35 % at $x = 300$ mm, regardless the heating of the fuel and air flows or the value of the exit Reynolds numbers. The maximum average temperature, however, range from 1465 K to 1727 K and decreases with $x$ in the smallest Reynolds number flame, flame I and probable flame II, or increases with $x$ in the other flames. The differences in maximum average temperature are relatively large compared to the differences in maximum average OH concentration.

7.7 Standard deviation

In Figures 7.23–7.28, the profiles of standard deviation of OH fluctuations are shown, related to the average profiles of Figures 7.14–7.19.

For $x \geq 200$ mm in flame I, the large centerline values of the standard deviation are due to soot scattering signals, see also Section 7.4.1. The real OH standard deviations at centerline in flame I are expected to be close to zero, like in flames II and III.

The profiles of standard deviation are wider than the average concentration profiles because the largest fluctuations are on the slopes of the average concentration profiles. The standard deviation maxima are found at a radial distance that is 5 to 10 % larger than the radial location of the maximum average
Figure 7.21: Maximum average temperature at different axial locations of flames I (○), II (△), III (□), IV (○), V (●) and VI (●).

Figure 7.22: Maximum average OH concentration at different axial locations of flames I (○), II (△), III (□), IV (○), V (●) and VI (●).
7.7. *Standard deviation*

![Graph](image1)

Figure 7.23: Radial profiles of OH standard deviation of flame I, at $x = 50$ mm (---), 100 mm (--), 150 mm (— —), 200 mm (— —), 250 mm (— — —) and 300 mm (—).

![Graph](image2)

Figure 7.24: Radial profiles of OH standard deviation of flame II, at $x = 50$ mm (---), 100 mm (--), 150 mm (— —), 200 mm (— —), 250 mm (— — —) and 300 mm (—).
Figure 7.25: Radial profiles of OH standard deviation of flame III, at $x = 50 \text{ mm} (- -)$, 100 mm (---), 150 mm (— —), 200 mm (— —), 250 mm (— —) and 300 mm (—).

Figure 7.26: Radial profiles of OH standard deviation of flame IV, at $x = 50 \text{ mm} (- -)$, 100 mm (---), 150 mm (— —), 200 mm (— —), 250 mm (— —) and 300 mm (—).
Figure 7.27: Radial profiles of OH standard deviation of flame \( v \), at \( x = 50 \text{ mm} \), \( 100 \text{ mm} \), \( 150 \text{ mm} \), \( 200 \text{ mm} \), \( 250 \text{ mm} \) and \( 300 \text{ mm} \).

Figure 7.28: Radial profiles of OH standard deviation of flame \( v \), at \( x = 50 \text{ mm} \), \( 100 \text{ mm} \), \( 150 \text{ mm} \), \( 200 \text{ mm} \), \( 250 \text{ mm} \) and \( 300 \text{ mm} \).
Figure 7.29: Standard deviation relative to average OH concentration of flame III, at $x = 50$ mm (--), $150$ mm (— —) and $250$ mm (---).

Figure 7.30: Standard deviation relative to average OH concentration at the radial location of maximum average OH concentration, of flames I (-o-), II (-Δ-), III (-□-), IV (-○-), V (- -○- -) and VI (- -○- -).
concentration. In a nonpremixed flame, OH is present in narrow regions deformed by turbulence. Because these regions are so narrow and local gradients are large, standard deviations in all turbulent flames investigated are very large; the instantaneous images of OH of the smallest Reynolds number flames (see e.g. Figure 7.3) could give the impression of small fluctuations in OH concentration for small x, but the standard deviations show that this is not true. In Chapter 8, we show that the shear layer in flames I and II is not yet developed for x \leq 175 mm. Hence, the standard deviation of OH in these flames is merely due to instabilities than due to turbulence.

The radial profiles of relative standard deviation are shown for flame III in Figure 7.29; for sake of clarity we only showed the values at x = 50, 150 and 250 mm. The profiles of the other flames are qualitatively equal. The relative standard deviation is minimum where the average concentration is maximum, and in the central region where the OH concentration is close to zero. In all flames, the maximum relative standard deviation increases with x, from about 1.0 at x = 50 mm to 1.5 at x = 250 mm. The effect of flame conditions is more apparent in the standard deviation at the location of maximum average concentration, which is also the location of the local minimum relative standard deviation. The values obtained in all flames these radial locations are shown in Figure 7.30. The smallest exit velocity flames have the smallest relative standard deviation at the location of maximum standard deviation. In flames I to III, the relative standard deviation shown in this figure increase with x. In flame IV there is a maximum at x = 100, which corresponds to the location of most frequent local extinction, as are discussed in Section 7.8. For x \geq 200 mm, the relative standard deviation at the location of maximum average concentration is in the range 0.7 to 0.8 in all flames. Note that we also found that the maximum average concentrations in all flames were also very close at the locations most downstream the nozzle. In the preheated flames (flames V and VI) the minimum relative standard deviation does not change much with x; close to the nozzle the relative standard deviation is already quite large.

The maximum relative standard deviation of the mixture fraction in flame III is about 0.45 at x = 50 mm and decreases with x, as calculated by Peeters[3]. The OH relative standard deviations, however, increase with x and the maximum values are about 3 times larger. As shown by the instantaneous images of OH (Figures 7.3–7.8), the OH remains in narrow regions with increasing x, whereas the mixture fraction spreads over a range increasing with x, and its gradients decrease. Hence, the manifestation of OH in nonpremixed flames in narrow regions makes that near the reaction zone, the standard deviation of OH concentration is much larger than that of the mixture fraction, and increases with x.
Chapter 7. Temperatures and OH concentrations in turbulent flames

7.8 Concentration distributions of OH fluctuations

We now discuss the distributions of concentration fluctuations, which are realizations of probability density functions. (We also use the term concentration distributions or concentration distribution functions for the realizations of the probability density functions.) These distributions are measured at fixed locations in the flames, but also conditional on the maximum in instantaneous OH concentration profiles. The main objective for doing this was to identify the effect of local extinction on the distribution of concentration fluctuations, separately from the effect of intermittency. By identifying the instantaneous maxima in the profiles, we are also able to determine the fluctuations of the OH zone location, of the average OH zone width and of largest eddies wrapping up the OH structure.

7.8.1 Concentration distributions at fixed locations

At every axial location in all six flames, 5000 measurements have been collected. Due to the low repetition rate of the laser (10 Hz), the time between the measurements was long compared to the largest turbulent time scales and the subsequent measurements were uncorrelated. Distribution functions of the concentration fluctuations are therefore constructed by putting the instantaneous values in appropriate bins (i.e. concentration intervals). We chose 70 bins, so the distribution functions contain 70 points.

Background level estimation

A significant part of the collected signal was a background signal, added to the fluorescence signal. The background signal was induced by the laser. The level was in the order of 100 counts, which may seem insignificant compared to a maximum fluorescence signal of about 6000–8000, but is important when investigating the small concentration structure in the distribution functions, or more specifically, the presence of intermittency in the concentration distribution functions.

Due to the horizontal polarization of the laser light, we have not detected any Rayleigh scattering. The UG11 filter in front of the camera made that only UV light was detected. The background signal appeared to be laser induced, mainly from UV laser light scattered by objects in the room and was kept as small as possible by shielding with black cardboard. The background signal is not constant for all array elements, but to a good approximation it is linear along the diode array. The background signal is estimated in every measurement from a linear interpolation of the signal at both edges of the diode array, where there is no fluorescence signal. The distributions of concentration fluctuations are calculated after subtraction of the background signal.
7.8. Concentration distributions of OH fluctuations

*Shifted intermittency peaks*

Intermittency between zero OH concentration outside the OH zone and finite OH concentration in the OH zone results in well defined maxima in the distribution functions at small concentration in all flames and at all locations, except for the radial location of maximum average concentration at \( x = 50 \) mm in the smallest exit velocity flame, flame 1. After the subtraction of the background signal, however, in the vicinity of the radial location of maximum average concentration, these maxima are at concentrations larger than zero. The distribution functions from the one- and two-dimensional measurements are equal in shape. In the two-dimensional measurements, however, the intermittency peaks are much closer to zero. This shift of the intermittency peaks is caused by a smooth transition between background signal and OH fluorescence that was observed in the instantaneous one-dimensional OH profiles but not in the instantaneous two-dimensional images (where the fluctuations in the background signal are also one order of magnitude larger). This smoothed transition is not very prominent in the instantaneous one-dimensional concentration profiles and only manifests in the distribution functions. Because of the difference in the concentration distributions of the one- and two-dimensional measurements, the shifted intermittency peaks in the distributions are thought to be artificial and are attributed to the larger measuring volume in the one-dimensional measurements. (The same high-quality imaging lens was used in both measurements.) Because of the smaller dynamic range of the two-dimensional measurements and the smaller number of measurements, the concentration distribution functions from the one-dimensional measurements are presented, relying on the similarity in shape of the distribution functions in both measurements. The distribution functions are corrected by shifting the intermittency peak to zero. In the procedure to correct the distribution functions, the maximum is kept constant and the function is stretched so that the intermittency peak is at zero concentration. In all flames this shift was largest at the radial location of maximum average concentration and was, for all values of \( x \), between 10 and 18% of the maximum instantaneous concentration.

*Shape of the concentration distribution functions*

In a nonpremixed flame, OH is usually found in narrow structures affected by turbulence. Therefore, at most locations in the different flames, the most probable concentration is (nearly) equal to zero. Turbulent fluctuations in the location of the narrow OH zone make that at some instances at a certain location, OH concentrations are large. When the gradients of OH are very large in the narrow OH zone, the concentration distribution function can be bimodal: the concentration at this location is either small or large. When concentration gradients are not so large, the concentration is either close to zero or in a larger concentration range, and the distribution function will extend to large concentrations, with usually a maximum at zero concentration.
Chapter 7. Temperatures and OH concentrations in turbulent flames

Figure 7.31: Distribution functions of concentration fluctuations in flame 1, at $x = 50$ mm, at $r = 5.8$ (a), 6.7 (b), 7.6 (c), 8.6 (d) and 9.5 mm (e), where (c) corresponds to the location of maximum average concentration and (d) to the location of maximum standard deviation.

Figure 7.32: Distribution functions of concentration fluctuations in flame 1, at $x = 150$ mm, at $r = 8.0$ (a), 9.2 (b), 10.5 (c), 11.7 (d) and 12.9 mm (e), where (b) corresponds to the location of maximum standard deviation and (c) to the location of maximum average concentration.
7.8. Concentration distributions of OH fluctuations

Figure 7.33: Distribution functions of concentration fluctuations in flame I, at $x = 250$ mm, at $r = 9.7$ (a), 12.0 (b), 14.2 (c), 16.5 (d) and 18.8 mm (e), where (c) is the location of maximum average concentration and the location of maximum standard deviation.

Figure 7.34: Distribution functions of concentration fluctuations in flame II, at $x = 50$ mm, at $r = 5.4$ (a), 6.3 (b), 7.2 (c), 8.1 (d) and 9.0 mm (e), where (c) corresponds to the location of maximum average concentration. The location of maximum standard deviation and is between (c) and (d).
Figure 7.35: Distribution functions of concentration fluctuations in flame III, at \( x = 50 \) mm, at \( r = 5.0 \) (a), 5.9 (b), 6.8 (c), 7.7 (d) and 8.6 mm (e), where (c) corresponds to the location of maximum average concentration. The location of maximum standard deviation and is between (c) and (d).

Figure 7.36: Distribution functions of concentration fluctuations in flame IV, at \( x = 50 \) mm, at \( r = 4.5 \) (a), 5.4 (b), 6.3 (c), 7.2 (d) and 8.1 mm (e), where (c) corresponds to the location of maximum average concentration. The location of maximum standard deviation and is between (c) and (d).
7.8. Concentration distributions of OH fluctuations

Figure 7.37: Distribution functions of concentration fluctuations in flame V, at $x = 50$ mm, at $r = 4.0$ (a), 4.8 (b), 5.7 (c), 6.7 (d) and 7.6 mm (e), where (c) corresponds to the location of maximum average concentration. The location of maximum standard deviation and is between (d) and (e).

Figure 7.38: Distribution functions of concentration fluctuations in flame VI, at $x = 50$ mm, at $r = 3.4$ (a), 4.3 (b), 5.2 (c), 6.1 (d) and 7.0 mm (e), where (c) corresponds to the location of maximum average concentration. The location of maximum standard deviation and is between (c) and (d).
In most flames, the shape of the distribution functions is comparable for larger axial distance. Close to the nozzle, distributions with different shapes are found for different radial locations. In flame I these differences are most pronounced, as can be seen in Figure 7.31. For small \( x \), the fluctuations in the location of the OH zone are not much larger than the width of the OH zone, see also Section 7.8.4, so at the radial location of maximum average concentration, the concentration is most often in the range \( 10 \) to \( 20 \times 10^{15} \) cm\(^{-3} \), and concentrations close to zero are not probable. At radial locations corresponding to the slopes of the average OH profile, low concentrations become more and more probable. At certain radial locations, the distribution function becomes bimodal, with both a maximum at zero and at a large concentration. Where the distribution is most symmetric, the standard deviation is maximum and the skewness zero. For radial locations more remote from the location of maximum average concentration, the distribution functions have most-probable small concentrations and a large positive skewness.

For increasing axial distance, the probability of concentrations equal to zero increases due to the increasing turbulent fluctuations of the location of the OH zone in combination with the fact that this zone remains rather narrow. This is shown in Figures 7.32 and 7.33, with the distributions of flame I at \( x = 150 \) and 250 mm, respectively. With increasing axial distance in flame I, the most-probable large concentration at the radial location of maximum average concentration present at \( x = 50 \) mm (Figure 7.31(c)) is substituted by a most probable zero-concentration for larger \( x \) (Figures 7.32(c) and 7.33(c)).

For larger axial distance the shape of the distribution functions becomes comparable for all flames, with only a difference in maximum concentrations. Note that we already have found that for larger \( x \) the maximum average OH concentrations and the relative standard deviations in all flames are very close. We only show the axial dependence of the distributions of flame I (Figures 7.31, 7.32 and 7.33 for \( x = 50, 150 \) and 250 mm, respectively), which is representative for all flames. The differences in the distributions of the various flames is most pronounced at \( x = 50 \) mm. In Figures 7.34 to 7.38 the distributions of concentration fluctuations at \( x = 50 \) mm are shown for flames II to VI. In all figures the locations are chosen so that the maximum average concentration is in the third image (c), and the steps in radial distance are in all cases 0.9 mm.

In flame II, with fuel and primary air velocities 40% larger than in flame I, there is a distribution function with negative skewness (Figure 7.34(c)), like in flame I (Figure 7.31(c)). Away from the location of maximum average concentration, the skewness is always positive (Figures 7.31–7.33).

For flames with velocities larger than in flame II, like in flames III to VI, distribution functions at all radial locations have a positive skewness (Figure 7.35) where the skewness is minimum at the location of maximum average concentration.

In flame IV the primary air velocity is large and the flame is not pre-
heated. The instantaneous two-dimensional images of OH (Figure 7.6) showed many holes in the OH structure. Both the temperature (Figure 7.11) and the average OH concentration (Figure 7.17) is smaller than in flame III, with smaller primary air velocity. Comparing the distribution functions at $x = 50$ mm of flame IV (Figure 7.36) with that of flame III (Figure 7.35), we find that the probability of concentrations close to zero is much larger, which is most prominent at the radial location of maximum average concentration. This zero-concentration probability is therefore not only due to intermittency but mainly due to local extinction, giving holes in the OH structure. More evidence of this is given by the (conditional) distribution functions of the peak concentrations.

In the preheated flames V and VI the maximum concentrations are larger than in the flames that are not preheated. At $x = 50$ mm, the zero-concentration probability is also much smaller in flame VI than in flame IV, due to the decreased local extinction. The distribution functions of flames V and VI are comparable.

Except for some locations in flames I and II, the distribution functions all consist of a rather flat part and a maximum at zero concentration, with a finite width. Increased average concentration results in a smaller zero-concentration probability and a higher probability of the high concentrations, where the probability distribution remains flat in the high concentration range. Top-hat functions with a Dirac function at zero concentration, therefore best describes the experimentally determined OH fluctuations distribution functions, although the broader maximum is not represented. β-functions can better represent this broad maxima, but are not able to represent the flat probability distribution at high concentrations well.

### 7.8.2 Distribution of peak OH concentration fluctuations

The probability of zero concentration in the distribution functions shown in Figures 7.31–7.38, represent the cases that the OH zone has been moved by turbulence far enough from the location of observation, but also the cases of local extinction, where the OH zone has temporarily disappeared. To study the influence of local extinction on the concentration distribution functions, we identified in every instantaneous one-dimensional OH concentration profile the peak concentration $\hat{c}$.

In Figure 7.39 we show an instantaneous radial OH concentration profile, with the identified peak concentration $\hat{c}$, the instantaneous radial location of the peak concentration $r_\hat{c}$ and the instantaneous full width at half maximum of the radial OH zone cross section, $\delta_{FWHM}$. From the data set of 5000 profiles, distribution functions of the peak concentration, of the peak location and of the width have been constructed. These distribution functions are conditional on the location of the OH zone, and hence on a value of the mixture fraction close to stoichiometry.

Barlow et al.[47] measured distribution functions of OH conditional on the mixture fraction $0.75 < \xi / \xi_{st} < 0.95$, in an air diluted methane flame (25 %
Figure 7.39: Instantaneous radial OH concentration profile, with peak concentration $\dot{c}$, radial location of the peak concentration $r_\delta$ and the full width at half maximum of the radial OH zone cross section, $\delta_{\text{FWHM}}$.

methane and 75 % air, by volume, $d_j = 7.2$ mm, $Re_j$ in the range $1.77 \times 10^3$ to $2.1 \times 10^4$, $x/d_j = 20$). The shape of the conditional distribution functions is very similar to those presented in Figures 7.40–7.42.

For not too large axial distances, the OH zone is not evolved by eddies very large compared to the OH zone width, but only affected by small turbulent eddies, of a size equal to or smaller than the width of the OH zone. As a consequence, at these axial locations the instantaneous radial profiles of OH do not show more than one maximum at each side of the jet axis. In that case a hole in the OH zone corresponds to an instantaneous profile in which the maximum has disappeared. Hence, the distribution function of the peak concentrations gives the probability of holes in the OH structure, although it should be noted that this probability is weighed with the size of the holes. At downstream locations, larger turbulent eddies evolving the OH zone may obscure the presence of holes in this convoluted OH zone. We show, however, the distribution functions for axial distance up to $x = 250$ mm, because these distributions also allow for conclusions on the change of maximum instantaneous concentration with axial distance.

In the flame with the smallest velocities, flame 1, the concentration distribution function taken at the fixed radial location of maximum average concentration (Figure 7.31(c)), shows that concentrations are in the range $0 < c < 18 \times 10^{15} \text{ cm}^{-3}$, with a preference for the larger concentrations. The peak OH concentration found in every instantaneous radial profile, however, fluctuate within a narrower range $10 < \dot{c} < 18 \times 10^{15} \text{ cm}^{-3}$ (Figure 7.40(a)). The concentrations
7.8. *Concentration distributions of OH fluctuations*

Figure 7.40: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame I, at \( x = 50 \) (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).

Figure 7.41: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame II, at \( x = 50 \) (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).
Figure 7.42: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame III, at $x = 50$ (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).

Figure 7.43: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame IV, at $x = 50$ (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).
7.8. Concentration distributions of OH fluctuations

Figure 7.44: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame v, at $x = 50$ (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).

Figure 7.45: Distribution functions of the peak OH concentration of every instantaneous radial profile in flame v1, at $x = 50$ (a), 100 (b), 150 (c), 200 (d) and 250 mm (e).
c < 10 \times 10^{15} \text{ cm}^{-3} \text{ at fixed locations (Figure 7.31), are therefore due to the fluctuations in location of the OH zone because they do not occur near the center of the OH zone. In Chapter 8 we show that the shear layer in flame I is still developing for } x < 175 \text{ mm, with large-scale instabilities. These instabilities apparently displace the OH zone but do not cause local extinction.}

There is not a strong dependence on axial distance in the conditional distribution functions. The Gaussian shaped functions slightly shift to smaller concentrations for axial distance up to 150 mm and slightly increases again for larger } x.

With increased velocities (flames II, III and IV, respectively), very small peak concentrations start to occur and increase with exit velocity. The small peak concentrations are due to holes in the OH zone. We find that the probability for 
1 \leq \hat{c} \leq 5 \times 10^{15} \text{ cm}^{-3} \text{ is very small. This shows that the peak concentration can decrease to a value of about } 5 \times 10^{15} \text{ cm}^{-3}, \text{ but jumps to a value close to zero instead of decreasing further, when the flame temperature becomes too low.}

The high occurrence rate of holes in flame IV observed in the instantaneous OH images (Figure 7.6), is also very clear in the distribution of the peak concentration, shown in Figure 7.43. We find that at } x = 100 \text{ mm the holes are most frequent, like in flame III (Figure 7.42). At about this axial distance, the shear layer between the primary and secondary air starts to affect the reaction zone (Section 2.2). Also, for smaller } x \text{ the pilot flames limit the local extinction by keeping the fuel and air mixture sufficiently hot. In the conditional concentration distribution functions of flames III and IV, the most-probable peak concentrations are equal. The only difference is the probability of zero peak concentration, } i.e. \text{ in the occurrence rate of holes. Although the flame structure of flame IV is different from flame III, affecting both average temperatures and average OH concentrations, the OH in the burning mixture appears not to be strongly affected by the larger air velocity. Even though at larger axial distance, the convection of the OH zone obscures many holes, there is still a finite probability to find no OH concentration at all in the one-dimensional cross section at large axial distances.}

The conditional distribution functions of OH mass fraction presented by Barlow et al.[47] for the flames with } Re_j = 1.77 \times 10^3 \text{ to } 2.1 \times 10^4 \text{ are similar to those of our flames I–III. Local extinction only occurred in the flame with the largest exit Reynolds number. This is because their fuel was premixed with air, and because in our flames local extinction is sensitive to the primary air velocity.}

The preheated flames exhibit much fewer local extinction, as can be seen in Figures 7.44 and 7.45, but some holes are obscured by the stronger convection of the OH zone in the preheated flames. The decreased number of holes has also been observed in the instantaneous two-dimensional images (Figures 7.7 and 7.8) and followed from the Damköhler number analysis of Sections 2.3.4 and 7.3. The probability of zero peak concentrations in the preheated flames has decreased with respect to flames III and IV. Flame VI shows a slightly larger number of holes than flame V because of the larger primary air velocity.


7.8. Concentration distributions of OH fluctuations

7.8.3 Maximum concentration of all measurements

The maximum OH concentration is the largest concentration of all measurements. This maximum concentration can be derived from either the conditional (peak) concentration distributions, Figures 7.40–7.45, or from the concentration distribution functions at the location of maximum average OH concentration, Figures 7.31–7.38. The maximum OH concentration varies with flame and with axial location.

The maximum OH concentrations in flames I and II are almost equal at the same axial distance, but are about 20% larger than in flames III and IV for small $x$. In flames I and II the maximum concentrations decrease with $x$ for $x < 150$ mm and slightly increase for larger $x$, whereas they increase with $x$ in flames III and IV.

Laminar flamelet calculations by Peeters[3] showed that by increasing the strain rate the peak temperature decreases from 2134 K for very small strain rate to 1871 K for high strain rate and near extinction, whereas the maximum OH concentration in this laminar flamelet decreased with only 10%, from 20.1 to $18.1 \times 10^{15}$ cm$^{-3}$. The maximum OH concentration in a laminar flamelet therefore appears to be rather insensitive to the strain rate.

The maximum OH concentration in the turbulent flames, decreases with a larger amount when the exit velocities, or strain rates, are increased. This is not predicted by laminar flamelet calculations and is attributed to the influence of temperature on the broadband detected OH fluorescence signal. The distribution function shown in Figure 7.42(a) shows that the flame is often close to extinction, so shear stresses are generally rather large. In Chapter 3 we found that a decrease in fluorescence signal of about 30% is to be expected from a maximum temperature decreasing from 2208 K to 1766 K, or about 18% from the 263 K temperature decrease mentioned above. Hence, we expect that the decrease in maximum OH concentration does not occur in the flame. The decrease corresponds to change expected from broadband detection in combination with detailed chemistry laminar flamelet calculations.

The average concentrations are primarily determined by the turbulent fluctuations and not by the maximum values. The maximum average OH concentrations in the different flames differ much more than the maximum concentrations. If the fact that the maximum OH concentration decreases due to increased exit velocities indeed stems from the influence of temperature on the broadband detected fluorescence signal, correction for this artifact would make the differences between the different flames less pronounced but would not change the trends.

In flame III, the maximum OH concentration increases with $x$, whereas the maximum average concentration slightly decreases with $x$. Therefore, if the change in maximum instantaneous concentration is artificial and is corrected for, the maximum average concentration in flame III still decreases with $x$. In com-
bustion models based on a single conserved scalar, the mixture fraction standard deviation decreases with $x$, and consequently the average OH concentration, see Peeters[3]. Hence, the maximum average OH concentration decreasing with $x$ is not predicted by these models.

The maximum concentration in flame I at $x = 50$ mm is in very good agreement with the maximum OH concentration in a laminar diffusion flame of $20 \times 10^{15}$ cm$^{-3}$, predicted calculations of laminar flamelet with small strain rate, carried out by Peeters[3]. In flame I for larger axial distance and in the other flames, the maximum concentrations are smaller, most probably due to the influence of temperature on the broadband detected signal. Calculations of a laminar flamelet with preheated flows predict a maximum OH concentration between 29 and $20 \times 10^{15}$ cm$^{-3}$, for strain rates ranging from small ($a = 100$ s$^{-1}$) to close to extinction ($a = 1900$ s$^{-1}$). The maximum concentrations found in the preheated flames V and VI are between these two extremes. Taking the broadband detection and calibration biases into account, the agreement with the concentrations predicted by laminar flamelet calculation is very good.

### 7.8.4 Fluctuations in OH zone location

The one-dimensional detection allowed for identification of the peak concentration in the instantaneous radial OH profile. In addition to the peak concentration, we have also determined the location of this peak concentration. Fluctuations of this location are expressed in distribution functions. The peak location distributions have been found to be symmetric about the average peak location. The average peak location was in all cases equal to the location of maximum average concentration. So, whenever we have mentioned "the location of the maximum average concentration", this can also been read as "the average location of the OH zone".

In Table 7.3 we give the standard deviations of peak location fluctuations $\sqrt{\langle r_x^2 \rangle}$ of flame I. Comparing the peak location fluctuations in this flame with the average OH zone width, discussed in Section 7.8.5, we find that at $x$

<table>
<thead>
<tr>
<th>$x$ (mm)</th>
<th>$\sqrt{\langle r_x^2 \rangle}$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>0.59</td>
</tr>
<tr>
<td>100</td>
<td>1.07</td>
</tr>
<tr>
<td>150</td>
<td>1.57</td>
</tr>
<tr>
<td>200</td>
<td>2.50</td>
</tr>
<tr>
<td>250</td>
<td>4.48</td>
</tr>
<tr>
<td>300</td>
<td>4.68</td>
</tr>
</tbody>
</table>
= 50 and 100 mm the fluctuations are smaller than the average OH zone width: the displacement of the OH zone is therefore generally smaller than its width at these locations in flame I. For \( x = 200 \) mm and larger, the peak location standard deviation is several times larger than the average OH zone width. Standard deviations of the peak location of flame II are comparable, differing less than 30 % for \( x \leq 300 \) mm. At all \( x \) covered in flame III the peak location standard deviations are larger than 3 mm, and hence several times larger than the average OH zone width at all locations. This also the case for flames IV to VI.

### 7.8.5 Fluctuations of OH profile width

The width of the reaction zone, and hence of the OH zone, is determined by the mixing of fuel and air. Not too far downstream, this mixing is by small eddies originating in the burner, because the fuel and primary air flows become turbulent in the burner. The size of these eddies is about 0.5 mm, or 3 % of the hydraulic diameter of the annulus inside the burner.

We have determined the width of the instantaneous OH profile at one side of the centerline. The instantaneous width is defined as the full-width-at-half-maximum (FWHM), and designated with \( \delta_{\text{FWHM}} \), see also Figure 7.39. Fluctuations of this width were expressed by distribution functions. These distribution functions provide information on two phenomena. First, when the OH zone is perpendicularly crossed, the width of the OH profile is the width of the OH zone itself, and it is interesting to know whether this width is affected by flame conditions. We designate the average OH zone width by \( l_{\text{OH}} \), which is a measure of small scale turbulence. Secondly, the largest width of the OH profile, when the OH zone is not perpendicularly crossed, is determined by the largest eddies deforming and wrapping up the OH zone. The largest OH profile width, serving as a measure of large scale turbulence, is designated by \( L_{\text{OH}} \).

The average OH zone width is assumed to be equal to the most probable value of \( \delta_{\text{FWHM}} \), which is not much larger than the minimum value of \( \delta_{\text{FWHM}} \) in the distribution functions. In this, we follow Stepowski et al.[59] who presented OH zone width values for a hydrogen/air nonpremixed flame \( (Re_j = 1.5 \times 10^4, \, d_j = 10 \text{ mm}) \) and who found a OH zone width of 1.5 mm, independent of \( x \), for \( 1 < x/d_j < 13 \). Chen and Goss[10] observed in a turbulent methane flame \( (Re_j \approx 10^4, \, d_j = 5 \text{ mm}, \, x/d_j < 60) \) "thin flamelets" as being single peaks in the one-dimensional OH profiles and they found a width of these flamelets equal to 1 mm. According to Chen and Goss[10], the largest widths are due to "wrapping of the OH structure around vortices" (in our words: convolution of the OH zone) and therefore indicate the size of the largest eddies deforming the OH structure.

As an example of the distribution functions of \( \delta_{\text{FWHM}} \), in Figure 7.46 we show the distribution functions for flame V at \( x = 50, 150 \) and 250 mm. In all flames, the shape of these distribution functions is generally the same. In the flames smallest exit velocities (flames I, II and III), the distribution functions extend to smaller values of \( \delta_{\text{FWHM}} \), in flame V1 larger values are found than shown...
Figure 7.46: Distribution functions of the instantaneous OH zone width, $\delta_{FWHM}$, in flame V for $x = 50$ (---), 150 (--- ---) and 250 mm (---).

In Figure 7.46. Values are discussed in the following.

**Average OH zone width**

Values of the average OH zone width $l_{OH}$ are given in Table 7.4, for $x = 50$ and 300 mm. We find that with increasing exit velocities of fuel and primary air (flames I, II and III) and additionally increasing the primary air velocity (flame IV), the OH zone width decreases, due to larger shear stresses. The values are comparable to those found by Chen and Goss\cite{10} for a methane flame. In the preheated flames the average OH zone width is slightly larger than in the flames that are not preheated. In all flames, the average OH zone width increases with

<table>
<thead>
<tr>
<th>flame</th>
<th>$l_{OH}$ (mm)</th>
<th>$x = 50$ mm</th>
<th>$x = 300$ mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>1.45 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>II</td>
<td>1.25 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>III</td>
<td>1.13 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>IV</td>
<td>0.98 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>V</td>
<td>1.13 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>VI</td>
<td>1.13 ± 0.15</td>
<td>1.7 ± 0.5</td>
<td></td>
</tr>
</tbody>
</table>
7.9. Comparison of single-line and broad-band detected OH

$x$, but this increase is small: less than a factor 2 between $x = 50$ and 300 mm. Within the accuracy of the measurements, at $x = 300$ mm the average OH zone width is equal in all flames. In flame 1 at $x = 50$ mm, the average OH zone width is 1.45 mm, whereas the standard deviation of OH zone location fluctuations is 0.59 mm. Hence the location fluctuations are smaller than the average OH zone width, but the values are still in the same order of magnitude. The values of $l_{OH}$ are compared with the Taylor micro scales of OH fluctuations in Section 7.11.

Estimation of the size of the largest eddies

As discussed before, the distribution functions of the full-width-at-half-maximum also give an indication of the size of large eddies wrapping up the OH structure. The distribution functions of $\delta_{\text{FWHM}}$ are very asymmetric, and the large eddy scale $L_{OH}$ is difficult to determine. A good estimate appeared to be the average value of $\delta_{\text{FWHM}}$ plus three times the standard deviation of $\delta_{\text{FWHM}}$. Scaled with $x$, $L_{OH}$ is almost constant for $150 \geq x \geq 300$ mm. The results from $x = 150$ to 300 mm are averaged to give $L_{OH}/x$ and its accuracy, and the values are given in Table 7.5.

Table 7.5: Macroscopic length scale $L_{OH}$, scaled with $x$. $L_{OH}$ is derived from average plus 3 times the standard deviation of $\delta_{\text{FWHM}}$. The average value of the values at $x = 150$, 200, 250 and 300 mm are given, the uncertainty is estimated from the standard deviation of the values at 4 axial locations.

<table>
<thead>
<tr>
<th>flame</th>
<th>$L_{OH}/x$ $(10^{-2})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>3.01 ± 0.28</td>
</tr>
<tr>
<td>II</td>
<td>3.44 ± 0.77</td>
</tr>
<tr>
<td>III</td>
<td>3.96 ± 0.48</td>
</tr>
<tr>
<td>IV</td>
<td>4.37 ± 0.39</td>
</tr>
<tr>
<td>V</td>
<td>6.06 ± 0.87</td>
</tr>
<tr>
<td>VI</td>
<td>6.86 ± 0.87</td>
</tr>
</tbody>
</table>

We find that the macroscopic length scale $L_{OH}$ is larger in the flames with stronger deformation of the OH zone, and which have been identified as more turbulent: $L_{OH}$ increases with primary air exit velocity.

7.9 Comparison of single-line and broad-band detected OH

In Chapter 3 it has been found that the broadband detected OH is affected by composition and temperature more severely than single emission-line detected
OH. In this section we compare turbulent flame measurements of OH, obtained with single emission-line detection and broadband detection.

In Section 6.3, a similar comparison has been discussed for OH measurements in the laminar premixed flat flame. In that section, it has been found that the average concentrations obtained with both methods matched very well. However, the concentration measurement fluctuations are much larger in the case of single emission-line detection: due to the very small signals, shot noise is very significant.

Like in the flat flame, the single emission-line detected OH measurements in the turbulent flame is affected by shot noise. Shot noise broadens the OH fluctuation distribution functions. Hence, the maximum concentration derived from the single emission-line detected OH distribution functions is larger than in the case of insignificant shot noise. The latter is the case for the broadband detection, where signals are almost two orders of magnitude larger.

The influence of shot noise on the distribution function can be calculated, because according to Eckbreth[24] shot noise is Poisson distributed. This Poisson distribution manifested in the flat flame OH measurements (Section 6.3). We designate the probability density function of the detected signal fluctuations (including shot noise) by \( f_{\text{total}}(S_F) \), and that of the turbulent fluctuations of the OH signal by \( f_t(S_F) \). The Poisson distribution of shot noise is given by \( f_{\text{Poisson}}(S_F, \langle S_F \rangle) \) given by:

\[
f_{\text{Poisson}}(s, \langle s \rangle) = \frac{(s)^{s}e^{-s}}{s!}
\]  

(7.1)

Note that the fluorescence signal \( S_F \) is discrete. The probability density of a signal \( S_F \) is the integration of the probability densities of all signals related to the concentration \( S_F \) multiplied by the probability that the signal in the absence of shot noise is \( S_F \) and the signal with shot noise is \( S_F \). In other words:

\[
f_{\text{total}}(S_F) = \int_{0}^{\infty} f_{\text{Poisson}}(S_F, \langle S_F \rangle = S_F') f_t(S_F')dS_F'.
\]

(7.2)

We compare the OH measurements with both methods, making use of the argument that shot noise is negligible in the case of broadband detection. This is done for the locations of maximum average OH concentration in flame III at \( z = 100, 150, 200 \) and 250 mm. We assume for the moment that the probability density function \( f_t \) is represented by the broadband detected distribution function. The signal in this distribution function is brought to the same level as in the single emission-line detected distribution function by multiplication with the ratio of the average signals of the flat flame OH. We did not subtract shot noise from the single emission-line detected distribution functions, but instead added noise to the broadband detected distribution functions. Note that no parameters have been fitted in this procedure.

In Figure 7.47 we present distribution functions obtained with single emission-line detection, with broadband detection and the distribution functions
where shot noise was added to the broadband detected distribution functions. We find that the maximum concentrations as estimated from the single emission-line detected measurements are about 20 \% larger than from the broadband detected measurements. Also, the probability of large concentrations is larger in the case of broadband detection.

When shot noise is added, the distribution functions of the broadband detected signal become very similar to those of the single emission-line detected signal. Consequently, the difference in distribution functions between the two detection methods is very well explained by the influence of shot noise on signals in the case of single emission-line detection. Then, the influence of temperature and composition on the maximum OH concentration measured with broadband detection has to be much smaller than the influence of shot noise on the maximum signal in the case of single emission-line detection. Hence, the maximum
concentrations in flame III are biased due to broadband detection by less than about 10%. This confirms the confidence in the broadband detection method.

7.10 Instantaneous gradients

For detailed modelling of nonpremixed combustion, the relation of physical quantities at different locations is essential. For example, the transport equation of the concentration variance contains a dissipation term with the squared concentration fluctuation gradient, which is usually modelled with a Taylor microscale. Also, when molecular diffusion is included in combustion models using PDF methods, the relation between e.g. the mass fractions and their gradients is important. In 1990, Pope[20] discussed the "Progress and Challenges of turbulent combustion modelling". Pope outlined a future model of turbulent combustion, combining both the advantages of the laminar flamelet model and models using PDF methods.

In the laminar flamelet modelling, the local, instantaneous composition is related to the mixture fraction and its dissipation rate. The local reaction zone is assumed to be a steady, plane laminar flamelet, which is strained by the turbulent field. The chemistry of a laminar, nonpremixed flame is rather well developed. This method is applicable when turbulence is weak and/or chemistry is fast, so in the large Damköhler number limit. When turbulence time scales are too small (or chemistry time scales too large), the flamelet may be extinguished or the fuel and air may premix, destroying the nonpremixed flamelet. Furthermore, the smallest turbulent eddies can be smaller than a characteristic length scale of combustion, like the reaction zone thickness. In that case, partial premixing of fuel and air can occur. In addition, at larger axial distance the fuel and air mix with combustion products, changing the composition and the temperature of the reacting flows. The flamelet model "is the method of choice, when it is applicable"[20], for it resolves the small-scale spatial structure, which is dominated by molecular diffusion and chemical reactions: at large Damköhler number a characteristic length scale of the reactions is much smaller than a characteristic turbulence length scale.

According to Pope[20], it is a challenge to represent the small-scale structure also with PDF methods, that can cope with turbulent structure inside regions of combustion. In PDF modelling a transport equation is solved for a joint PDF, in which the most sophisticated PDF models velocities, temperature, dissipation rate of turbulent kinetic energy and local composition are included. (Local composition is represented by a vector \( \phi \) containing all species mass fractions and enthalpy.) Molecular diffusion still has to be modelled. Since diffusion is governed by the steepest gradients, it is associated with the smallest scales[20]. When local gradients are included in the joint PDF, molecular diffusion can be calculated. In the case of isotropic turbulence, inclusion of the gradient \( \nabla \phi \) is equivalent to inclusion of the scalar dissipation, \( D(\nabla \phi)^2 \). This scalar dissipation is to a good
Figure 7.48: Joint distribution of OH concentration and radial gradient, measured in flame I at $x = 50$ mm, at $y = 7.34$ (a) and 7.95 mm (b). The location of maximum average concentration is at $y = 7.65$ mm. Contours are at 10 (outer contour), 20, 30, ... out of 5000 measurements.

With our one-dimensional detection of OH we can measure the joint distribution of OH concentration and radial gradient, or of concentration and the square of the gradient. It should be kept in mind that this single scalar joint distribution is not the joint distribution of the mixture fraction and its dissipation. The joint OH distributions are largely influenced by the sharply peaked OH profiles. Only Schäfer et al. [90] have presented experimental evidence of the relation between instantaneous OH concentration and OH gradient for a premixed turbulent natural gas-air flame, presented as a scatter plot; when the OH concentration increased from zero, the most probable gradient increased, starting at zero; a maximum value of the most probable gradient is reached at a certain concentration and for larger concentrations the most probable gradient decreases to zero again.

7.10.1 Joint distribution of concentration and gradient

We now show the relation between concentration and gradient under various conditions. This relation is shown by the joint distribution of concentration and gradient, or of concentration and the squared gradient (on a logarithmic scale). This is done for cases with very different structure of the OH zone: first we observe the case of small $x$ ($x = 50$ mm) in flame I. Instantaneous images of OH in this flame (see Figure 7.3) show that the OH structure is only slightly wrinkled. Secondly, we observe the case of $x = 50$ mm in flame III, where there
Figure 7.49: Joint distribution of OH concentration and the squared radial gradient, measured in flame I at $x = 50$ mm, at $y = 6.59$ (a) and 7.65 mm (b). The location of maximum average concentration is at $y = 7.65$ mm. Contours are at 20 (outer contour), 60, 100, 140 ... out of 5000 measurements.

Figure 7.50: Joint distribution of OH concentration and the squared radial gradient, measured in flame III at $x = 50$ mm, at $y = 5.75$ (a) and 6.95 mm (b). The location of maximum average concentration is at $y = 6.81$ mm. Contours are at 20 (outer contour), 60, 100, 140 ... out of 5000 measurements.
Figure 7.51: Joint distribution of OH concentration and the squared radial gradient, measured in flame III at \( x = 250 \) mm, at \( y = 12.72 \) (a) and 17.26 mm (b). The location of maximum average concentration is at \( y = 17.56 \) mm. Contours are at 20 (outer contour), 60, 100, 140 ... out of 5000 measurements.

are some holes in the OH zone in addition (see Figure 7.5). A third case is the joint distribution in the same flame III at larger \( x, (x = 250 \) mm) where the OH zone is more strongly deformed (see Figure 7.5). Finally, the relation between the concentration and its gradient is shown for flame IV, also at \( x = 250 \) mm. This flame is more turbulent than flame III, see also Figure 7.6.

For \( y \) smaller than the average radial location of the OH zone, the gradient is mainly positive and is positively correlated with the concentration: a larger concentration gives usually larger gradient, see Figure 7.48(a). (We have seen that the location of maximum average concentration is also the average location of the OH zone.) At the location of observation, the gradients are on average positive, but the fluctuations in the radial location of the OH zone are so large that the negative slope of the OH profile can be found at this location. For \( y \) larger than the average location of the OH zone, the gradient is on average negative, but is due to fluctuations in the location of the OH zone sometimes negative for this value of \( y \), see Figure 7.48(b). The gradient is largest when the concentration is about half the maximum value. The combinations of concentration and gradient are confined to a narrow band, indicating that the fluctuations are mainly due to fluctuations in the location of the OH zone, a conclusion that was already drawn from the distribution functions of maximum instantaneous concentration.

The joint distribution of the concentration and the squared gradient does not show a difference between the positive and negative gradients. Figures 7.49(a) and (b) show that for different radial locations the the concentration-gradient
Figure 7.52: Joint distribution of OH concentration and the squared radial gradient, measured in flame IV at \( x = 250 \text{ mm} \), at \( y = 10.75 \) (a) and 15.29 mm (b). The location of maximum average concentration is at \( y = 15.90 \text{ mm} \). Contours are at 20 (outer contour), 60, 100, 140 ... out of 5000 measurements.

pairs are confined to the same band and that the most probable pair shifts within this band.

The main difference between the joint distribution functions in flame I and flame III at \( x = 50 \text{ mm} \) (Figure 7.50) is the location of the most probable concentration-gradient pair, which is closer to the combination of zero concentration and small gradient, due to the holes in the OH zone: in a hole, both the concentrations and the gradients are small. The shape of the joint distribution functions of flames I and III is, however, similar.

For larger axial distance in flame III (Figure 7.51) large concentrations are less probable, so the most probable concentration-gradient pair has moved further towards the zero-concentration axis in combination with smaller gradients. The measurements, however, are confined to the same band as in the previous cases (Figures 7.49 and 7.50).

The joint distributions of flames III and IV at \( x = 250 \text{ mm} \) are almost equal, in spite of the different turbulent structure of the OH, visible in the instantaneous images of OH (Figures 7.3 and 7.7, respectively).

In the preheated flames, the maximum concentrations are larger than in the flames with initially cold flows, see Section 7.8.1. The shape of the joint distribution functions of the preheated flames is equal to those of the other flames, except for a stretch towards larger concentrations. The gradients in the preheated flames do not differ significantly from the gradients in the other flames.

Concluding, we can say that in all flames, the OH concentration and its
gradient are strongly correlated, and that the shape of the joint distribution function of concentration and the logarithm of the squared gradient is independent of the flame conditions or location in the flame. The flame conditions or location determine the most probable combination of concentration and gradient. Preheating only changes the joint distribution with respect to the concentrations and not with respect to the gradients.

7.11 Macro and micro scales from spatial correlation

7.11.1 Introduction

In many combustion models, length- (or time-) scales are important parameters to characterize the size of turbulent eddies or the reaction zone thickness. Borghi[19] uses one single length scale to characterize combustion chemistry and one to characterize turbulence. These phenomenological quantities are often not defined accurately and are therefore difficult to measure. Since turbulent combustion models with detailed chemistry are currently being developed, experimental evidence of length scales of turbulent fluctuations of many different species is required. For example, in the transport equation of the mean square concentration fluctuations of a species \( s \), \( \langle e_s'^2 \rangle \), the term \( \langle (\nabla e_s')^2 \rangle \) is modelled with a length scale \( \lambda_{d,s} \), as \( \langle (\nabla e_s')^2 \rangle = 2\langle e_s'^2(x, y, t) \rangle / \lambda_{d,s}^2 \).

Spatial or temporal correlation principles are applied to measurements of turbulent fluctuations of velocities to analyze the energy density at a specific frequency or, analogously, at a specific eddy size, to describe the energy cascade. This gives rise to different scales: integral scales for large macroscopic eddies, smaller Taylor micro scales to scale instantaneous gradients, as discussed above, and the Kolmogorov scales characterizing the smallest eddies. Integral length scales and Taylor micro scales can be derived from correlation functions, see for example Hinze[91].

The cascade process concepts have been developed for equilibrium turbulence, whereas for \( x/d_j < 50 \) in our flames turbulence is still developing. Also, when we apply these concepts to OH radical concentration measurements, we have to realize that radicals are not only transported by turbulent eddies of a wide range of sizes, but also are subject to chemical reactions. Williams[15] dissuades application of the cascade process concepts to scalars that are not conserved, for the production and destruction of these scalars interfere with the cascade process. However, spatial correlations can be studied without relating the results to the cascade process of turbulent eddies: spatial correlation functions simply describe the relation of turbulent fluctuations of a physical quantity at different locations, and represent the range of this relation. When this physical quantity is a reacting species, the relation of fluctuations at different locations is not only determined by turbulent eddies, but also by chemical reactions. Length scales derived from spatial correlation of reacting species, therefore, are affected by the chemical re-
actions, and generally do not represent the spatial structure as such. Comparison of the length scales of reactive species with those of nonreactive species, shows to what extent transport of reacting species is affected by chemical reactions, showing good agreement, except close to the average location of the reaction zone. In Chapter 8, this comparison is made.

The peaked structure of the OH profiles in combination with not completely developed turbulence very often gives very negative correlations and, consequently, very small or even negative integral length scales. This negative correlation is because for a large range of the radial distance, the concentrations that are correlated are on both flanks of the peaked radial OH profile: a radial displacement of the OH zone gives a positive fluctuation at one location and a negative fluctuation at the other side and vice versa. Straightforward calculation of the integral length scale from the spatial correlation therefore may give unrealistic results.

The Taylor micro scale derived from the correlation of the fluctuations at neighbouring points is a good approximation of the dissipation scale, relating the instantaneous gradients to the fluctuations. As mentioned above, this dissipation scale can be used in the transport equation of the mean square species fluctuations. Although spatial correlations and the associated length scales are originally based on the cascade process concepts, they are very useful in the study of the spatial structure of nonconserved scalars.

It may be because of considerations like that of Williams[15] that little results of spatial correlation of nonconserved scalars have yet been published. Seitzman et al.[65] calculates the correlation of a single two-dimensional OH image with its shifted version to derive autocorrelation length scales. This gives a characteristic size of all large scale structures in one complete image. From large sets of images one can compose distribution functions of these length scales. This strategy is comparable to our distribution functions of OH zone widths, presented in Section 7.8.5. Dasch and Heffelfinger[89] calculated two-dimensional spatial correlation functions and integral length scales from soot scattering images in a similar way as presented here.

Autocorrelation spectra and covariance spectra of reacting species have been studied by Kosály[92]. Kosály's study was an analysis of data from Bilger et al.[93]. It involved the simple case of a turbulent shear flow, mixing two reactants of a single chemical reaction without heat release. The frozen chemistry limit (zero Damköhler number) and the equilibrium chemistry limit (infinite Damköhler number) were studied. The effect of the chemistry rate appeared to be quite small in this simple case, but this result cannot be extrapolated to the much more complicated combustion chemistry case with significant amounts of heat release.

Spatial correlation of velocities and turbulence length scales in isothermal jets and heated jets can be found in Wygnanski and Fiedler[94], and Corrsin and Ubooi[95].
7.11. Macro and micro scales from spatial correlation

7.11.2 Definitions of spatial correlation and scales

The dissipation length scale is a characteristic length scale associated with instantaneous gradients. We can model spatial gradients of the concentration of species \( s \) with the ratio of the fluctuations and the dissipation length scale \( \lambda_{d,s} \):

\[
\left( \frac{\partial c_s'(x, y, t)}{\partial y} \right)^2 = \frac{2\langle c_s^2(x, y, t) \rangle}{\lambda_{d,s}^2},
\]

(7.3)

analogous to the definitions by Hinze[91] for velocity fluctuations and gradients. In the case of homogeneous turbulence this dissipation scale is equal to the Taylor micro scale, derived from the spatial correlation. Turbulent nonpremixed flames are not homogeneous in the observed regions. The difference between the Taylor micro scale and the dissipation scale, however, is small, as will be shown in a following section. The spatial correlation of concentration fluctuations of species \( s \) is defined as:

\[
\rho_s(x, y, \delta) = \frac{\langle c_s'(x, y - \delta/2, t) \times c_s'(x, y + \delta/2, t) \rangle}{\sqrt{\langle c_s^2(x, y - \delta/2, t) \rangle} \times \langle c_s^2(x, y + \delta/2, t) \rangle}.
\]

(7.4)

\( \rho_s(x, y, \delta) \) obeys:

\[
|\rho_s(x, y, \delta)| < \rho(x, y, 0) \equiv 1 \text{ for } \delta > 0,
\]

(7.5)

and from (7.4) it is apparent that

\[
\rho_s(x, y, \delta) = \rho(x, y, -\delta).
\]

(7.6)

When the turbulent fluctuations are not periodic functions of \( y \),

\[
\lim_{\delta \to \infty} \rho_s(x, y, \delta) = 0.
\]

(7.7)

Scaling of the autocovariance, \( \langle c_s'(x, y - \delta/2, t) \times c_s'(x, y + \delta/2, t) \rangle \), with the variance \( \langle c_s^2(x, y, t) \rangle \), instead of scaling with the product of standard deviations at \( y - \delta/2 \) and \( y + \delta/2 \) like in (7.4), is not allowed because this scaled autocovariance is not bounded by unity and cannot serve as a coefficient. The scaling used in (7.4) also levels out inhomogeneous OH concentration fluctuations.

For small \( \delta \), (7.4) can be approximated by a parabola,

\[
\rho_s(x, y, \delta) = 1 - \frac{\delta^2}{\lambda_s^2} + \cdots,
\]

(7.8)

defining the Taylor micro scale \( \lambda_s \), or equivalently,

\[
\frac{d^2 \rho_s}{d\delta^2} \bigg|_{\delta=0} = -\frac{2}{\lambda_s^2}.
\]

(7.9)

This definition is analogous to the definition of the Taylor scale of turbulence by e.g. Hinze[91]. In the case of homogeneous turbulence, where \( \langle c_s^2(x, y - \delta/2, t) \rangle \)
is equal to $(c_s^2(x, y + \delta/2, t))$, it is easily shown that in a first approximation the dissipation length scale (7.3) is equal to the Taylor micro scale (7.9), by writing $c_s'(x, y \pm \delta, t) \approx c_s'(x, y, t) + \partial c_s'(x, y, t)/\partial y$.

The integral length scale of concentration fluctuations is defined analogous to the definition of the integral length scale of turbulence by for example Hinze[91], Wygnanski and Fiedler[94] or Corrsin and Uberei[95]:

$$\mathcal{L}_s = \int_0^\infty \rho_s(x, y, \delta)d\delta. \quad (7.10)$$

Some authors, e.g. Kwon et al.[96], use instead of (7.4) a definition that is not symmetric, correlating radial locations $y$ with $y + \delta$. When the correlation function depends on location, so does the integral length scale, obtained by integration of the correlation function. An asymmetric definition of the correlation then shifts the radial structure of the integral length scale towards one side, making it difficult to compare the integral length scales locally with the average OH structure.

According to Hinze[91], lateral velocity correlation functions from isotropic turbulence are negative for a large range of $\delta$. Corrsin and Uberei[95], show that an even smaller negative value for the lateral correlation coefficient is found in isothermal and heated jets and that the range of $\delta$ where $\rho_s < 0$ is much larger. They hypothesize that this is due to "a slight irregular waving of the jet as a whole"[95]. A negative part in the correlation function decreases the integral length scale. Significant correlation can therefore be found between points that are separated by a distance (much) longer than the integral length scale. The integral length scale need not even be positive. Other definitions of the integral length scale are possible, in which both significant positive and negative correlation contribute to the integral length scale, for example by integrating the absolute value or the square of the correlation function. We, however, follow the convention and use definition (7.10), to be able to compare our results with those found in literature.

### 7.11.3 Simulation of spatial correlation

OH concentration profiles in flames differ from for example profiles of instantaneous velocities or temperatures in heated jets. Therefore, it is necessary to first investigate the influence of the peaked OH profiles on the spatial correlation functions, and hence on the integral lengths scales and on the Taylor micro scales. We do this by analyzing simulations of fluctuating instantaneous OH concentration profiles and the spatial correlation functions of these simulated profiles.

We represent the one-dimensional cross section of the instantaneous OH zone with a Gaussian function, with fluctuating location and peak value. The location of the simulated zone is determined by the location of the peak value, and
Figure 7.53: Simulated correlation functions of a Gaussian peak with fluctuating peak location and peak value, for different values of peak location standard deviation. The standard deviation of peak location fluctuations relative to the width of the simulated OH zone, \( \sigma_{\text{loc}}/\sigma_w \), is 0.44 (---), 0.78 (--), 1.44 (---) and 3.42 (---), the peak value fluctuation \( \sigma_p \) is 0.34 of its average value. The Taylor micro scales relative to the peak width, \( \tilde{\lambda}_s/\sigma_w \), are 0.84, 0.79, 1.01 and 1.30 respectively.

described with a nearly normal (Gaussian) peak location distribution. This distribution function is derived from the sum of three uniformly distributed pseudo-random numbers. The width of this location distribution is determined with a standard deviation \( \sigma_{\text{loc}} \), which was varied in the simulations. The fluctuations of the peak value are also modelled with a nearly normal distribution, with the standard deviation of the peak value \( \sigma_p \) as parameter, where the average peak value is unity. We observe the correlation at the average location of the simulated OH zone. The simulated correlation functions are designated with \( \tilde{\rho}(\delta) \). The width of the simulated OH zone is given by \( \sigma_w \), which was kept constant in the simulations and used to normalize the spatial parameters, \( \delta \) (as used in (7.4), the standard deviation of the zone location, integral length scale and Taylor micro scale. We define \( \sigma_w \) as the half width of the Gaussian function describing the OH zone, at \( 1/e \) of the maximum value.

The simulated concentration fluctuations at a fixed location are either due to location fluctuations of the zone or due to peak value fluctuations. In reality, both are due to influence of large- and small-scale turbulent structures on the OH zone. The very negative correlation is typical for the case where the fluctuations at a fixed location are mainly due to fluctuations in the location
of the zone. At one side of the zone the concentration increases while on the other side the concentration decreases with a movement of the zone, see also Figure 7.48. When the fluctuations in the location of the zone become very large, the two concentrations that are correlated are more often both on the same side of the zone or, equivalently, they have radial gradients with equal sign. Hence, the correlation will be less negative.

The simulated correlation functions are shown in Figure 7.53 for different values of the simulated zone location fluctuations, $\sigma_{loc}$. The values of $\sigma_{loc}/\sigma_w$ are 0.44, 0.78, 1.44 and 3.42. The value of peak value fluctuations $\sigma_p$ is 0.34, which is in the same order of magnitude as the measured peak concentration fluctuations of flame I (Figure 7.42(a)). We find that when $\sigma_{loc}$ is small, the minimum correlation and the Taylor micro scale decrease for increasing $\sigma_{loc}$, and increase when $\sigma_{loc}$ becomes relatively large. Minimum values of the correlation coefficient are in the range $-0.68$ to $-0.29$. Taylor micro scales scaled with $\sigma_w$ are $0.84, 0.79, 1.01$ and $1.30$, for the values of $\sigma_{loc}/\sigma_w = 0.44, 0.78, 1.44$ and $3.42$, respectively.

Simulated OH zone location fluctuations in combination with very small peak value fluctuations, gives minimum correlations as small as $-0.98$. Peak value fluctuations in the absence of location fluctuations give positive correlations. For constant location standard deviation $\sigma_{loc}$, increasing peak value fluctuations $\sigma_p$ give larger minimum correlations and larger Taylor micro scales. The Taylor micro scale strongly depends on the peak value fluctuations when $\sigma_{loc}/\sigma_w$ is much smaller than unity, but for larger $\sigma_{loc}/\sigma_w$, fluctuations at a fixed location are dominated by peak location fluctuations and the Taylor scale is almost independent of $\sigma_p$: a factor of 8.7 in $\sigma_p$ gives a factor of 4.3 in the Taylor micro scale (ranging from 0.4 to $1.7 \times \sigma_w$) when $\sigma_{loc}/\sigma_w$ is 0.23, but only 3% difference in the Taylor micro scale (about $1.3 \times \sigma_w$) when $\sigma_{loc}/\sigma_w$ is 1.8.

From calculations with different parameters, it appeared that a more negative minimum correlation is not always associated with a smaller Taylor micro scale. With $\sigma_p = 0.14$ (which is smaller than in the previous example), the Taylor micro scale was $40\%$ larger in the case of $\sigma_{loc}/\sigma_w = 0.78$ than in the case of $\sigma_{loc}/\sigma_w = 0.44$, whereas the minimum correlation was 7% more negative.

A minimum Taylor micro scale was found to be in the order of magnitude of $0.2 \times \sigma_w$ for a value of $\sigma_{loc}/\sigma_w$ that is much smaller than unity (very small fluctuations in the location of the zone) but still dependent on $\sigma_p$. In the actual OH measurements, as discussed in the previous sections, fluctuations in the radial location of OH zone are always larger than about half the average OH zone thickness, see Tables 7.3 and 7.4. For realistic values of $\sigma_p \approx 0.34$ and $\sigma_{loc}/\sigma_w > 0.5$, the Taylor micro scale is in the range 0.8 to 1.3 times $\sigma_w$, or 0.6 to 0.9 times the full-width-at-half-maximum of the simulated OH zone.

In most cases where the fluctuation of simulated zone location is dominant in the simulated concentration fluctuations, the correlation functions are negative over a large range of $\delta$ and the value of the integral length scales is negative. In the correlation functions shown in Figure 7.53, the integral length
scale $\hat{L}$ scaled with $\sigma_w$ are $-6.2$, $4.1$, $3.4$ and $-11.0$, for the values of $\sigma_{loc}/\sigma_w = 0.44$, $0.78$, $1.44$ and $3.42$, respectively. The integral length scales then do not have any significance with respect to turbulent length scales.

For increasing axial distance, the instantaneous OH profiles more and more deviate from the structure with two narrow zones as found upstream. The simulations have shown that the fluctuations in the location of the narrow OH zones is primarily responsible for strong negative correlations. When turbulence gradually develops giving a wider range of eddies, OH zones are wrapped around eddies larger than the OH zone width. This gives broader profiles with multiple peaks in the one-dimensional instantaneous concentration profiles. Concentration fluctuations of this origin give correlations which are usually less negative than in the case of OH profiles with zones that are not wrapped around large-scale eddies and of which the location fluctuates over a distance in the order of magnitude of the OH zone width. Hence, the integral length scales need not be negative in the more developed turbulent fluctuations of OH, but any negative part in the correlation function gives a decrease of the integral length scale. Even when the OH zone is strongly convoluted and the OH in the one-dimensional profiles is not in narrow regions, the Taylor micro scales can still be quite small, however, for they depend on gradient fluctuations which can remain large at downstream locations.

We summarize the results obtained in the previously discussed simulations. When the OH profile consists of two narrow zones, the fluctuations in the radial location of the simulated OH zone, result in correlations almost reaching $-1$. When location fluctuations are larger than the width of the zone, the minimum correlation is less negative. Peak value fluctuations give positive correlations. Together with location fluctuations the peak value fluctuations contribute to the fluctuations at a fixed location. Larger peak value fluctuations give negative correlations closer to zero. For a realistic case of peak location fluctuations which are not much smaller than the peak width and of peak value fluctuations of about 35% of the average peak value, the Taylor micro scales are in the range of 0.6 to 0.9 times the full-width-at-half-maximum of the simulated OH zone. The OH profile can only be represented with two narrow zones when the OH zone is not wrapped around eddies much larger than the OH zone width, because this causes broadening of the instantaneous radial OH profiles. We see in the following that in flames I, II and III at many axial locations the integral length scales have negative values.

### 7.11.4 Measured correlation functions

We now discuss the shape of the measured correlation functions. This is necessary when analyzing the integral length scale results. At a number of locations in the different flames, the integral length scales appeared to be negative due to
negative parts in the spatial correlation functions, as was also predicted by the simulations. This was mainly in the flames where large turbulent eddies did not strongly deform the OH structure.

In the previous section the simulations of correlation functions have shown that when location fluctuations are the main source of fluctuations at a fixed location in space, the correlation functions have very small, negative parts and consequently the integral length scales have very small or negative values. In flame I, the minimum value of the spatial correlation coefficient was most negative for small $x$ and for the radial locations of the maximum average concentration. This is shown in Figure 7.54 for $x = 100$, 200 and 300 mm. These correlation functions are comparable to the simulated correlation functions.

In flame III the distribution functions of the peak concentration fluctuations showed that for small $x$ in this flame, the peak concentration fluctuations are much larger than in flame I. Fluctuations in the location of the OH zone are in this flame less dominant than in flame I. We therefore find correlation functions in flame III (not shown in a figure), similar in shape to those of flame I, but with minimum values of the correlation function closer to zero: at $x = 50$ mm $-0.3$ in flame III compared to $-0.8$ in flame I, both for the radial location of maximum average concentration. The minimum correlation remains fairly constant with $x$ in flame III, unlike that of flame I (Figure 7.54).

In flame IV local extinction is the dominant source of OH concentration fluctuations for $x = 50$ and 100 mm. The peak concentration fluctuations result in positive correlations and the minimum correlation is $-0.014$ and $-0.003$ at $x =
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Figure 7.55: Correlation functions of flame IV, for $x = 100$ (---), 200 (– –) and 300 mm (---), for $y$ equal to the location of maximum average concentration.

Figure 7.56: Correlation functions of flame IV, for $x = 300$ mm and $y = 8.2$ (---), 16.4 (– –) and 24.6 mm (---). $y = 16.7$ mm corresponds to the radial location of maximum average concentration.
Figure 7.57: Results of the integral length scales $L_{OH}$ for flames IV (---), V (--), and VI (----) for $x = 300$ mm.

50 and 100 mm, respectively. For larger axial distance, the minimum correlation becomes more and more negative. This is shown in Figure 7.55 for $x = 100$, 200 and 300 mm for the radial locations of maximum average concentration. The correlation functions in the preheated flames V and VI are comparable to that of flame III.

The spatial correlation functions in flame IV at $x = 300$ mm are shown in Figure 7.56, for $y = 8.2$, 16.4 and 24.6 mm. For the sake of clarity, we did not show the correlation function at $y = 0$ mm in Figure 7.56, because it almost coincides with that for the radial location of maximum average concentration ($y = 16.4$ mm). The negative correlations are most dominant at the radial location of maximum average concentration and at the centerline.

The integral length scales have positive values for all radial positions in the flames IV, V and VI for $x \geq 200$ mm. The radial profiles of the integral length scale in these flames are discussed in the next section.

7.11.5 Integral length scales

In Figure 7.57 we show the integral length scale values for the different radial locations in flames IV, V and VI at $x = 300$ mm.

The integral length scales appear to be largest in the outer parts of the flame. This was also observed by Wygnanski and Fiedler[94] for the integral length scales in an isothermal jet, see also Section 8.9.

The integral length scales are minimum where the average concentration is maximum. In flame VI the two maxima of the average profile have merged, and
a rather flat profile of $L_{OH}$ is expected around $y = 0$ mm. The profiles of $L_{OH}$ of flames IV and V are almost equal, and are comparable to the integral length scales of flame VI. This indicates that the interaction between turbulence and the OH is comparable in these flames. This is expected, because the spatial structure of turbulence is primarily determined by the scale of the experiment (e.g. the jet diameter $d_j = 6$ mm).

We also derived integral length scales from the two-dimensional OH images, yielding longitudinal and lateral length scales. Values have been obtained in the far field, $500 < x < 800$ mm, or $83 < x/d_j < 133$, of a flame with fuel exit velocity equal to that of flames III and IV, and with primary air exit velocity of $5.8$ m/s, between the primary air velocity of flames III and IV. Recall that the exit velocities do not have a large influence on the length scales. The length scales have been obtained at the radial locations of maximum average OH concentration, where the integral length scales are relatively small, see Figure 7.57. We found values for the longitudinal and lateral integral length scales $L_{x,OH}/x = (1.1 \pm 0.3) \times 10^{-2}$, and $L_{y,OH}/x = (0.7 \pm 0.3) \times 10^{-2}$, respectively. Hence, the longitudinal integral length scales are about $60\%$ larger than the lateral scales. Wygnanski and Fiedler[94] obtained integral length scales of turbulent velocity fluctuations in an isothermal jet ($Re_j = 10^5$, $d_i = 26.4$ mm, $x/d_j = 90$), both in the longitudinal and radial direction. At centerline, they found $L_{x,u}/x = 3.83 \times 10^{-2}$ and $L_{y,u}/x = 1.57 \times 10^{-2}$, yielding a ratio $L_{x,u}/L_{y,u} = 2.4$. This ratio is larger than the ratio of the OH integral length scales, but the results are not in contradiction. The integral length scales of OH in the central part of the flame are about a factor $2$ smaller than those found by Wygnanski and Fiedler[94]. This is because OH is not a conserved scalar: chemical reactions decrease the correlation and thus the integral length scales. See for a more extensive discussion Section 8.9.

### 7.11.6 Taylor micro scales

Unlike the integral length scales, the Taylor micro scales are not limited to the regions where the interaction of turbulence with OH is very strong, for the Taylor micro scales are based on the local gradients. We therefore investigate the Taylor micro scales in all flames. Only in the central part of flames I to III, there are no significant amounts of OH, so the Taylor micro scale is not defined there.

The separation $\delta$ is quantified by the finite size of the elements in the diode array: $\delta = \delta_n = n\Delta r$, where $\Delta r$ is the diode separation projected at the measuring volume. The Taylor micro scales are derived from the correlation coefficient at the first non-zero value of $\delta$, so from $\rho_s(x,r,\Delta r)$. The parabola from which the Taylor micro scale is derived therefore consists only of one data point. More data points in the parabola would give a larger value of the Taylor micro scale and we like to have the highest resolution in $\lambda_{OH}$. Consequently, we have to accept a larger random error in $\lambda_{OH}$ because of the use of less data points.

In the different flames, the values of the Taylor micro scales are very
Figure 7.58: Results of the Taylor micro scales $\lambda_{OH}$ for flame III at $x = 100$ (a), 200 (b) and 300 mm (c).

close. Radial profiles of the Taylor micro scales in flame III and of flame VI are shown in Figure 7.58 and 7.59, respectively. Similar profiles are found for flames I, II and III. In flame VI, OH is found at the centerline even at $x = 50$ mm, so the Taylor micro scale are defined around $y = 0$, see Figure 7.59. For larger $x$, the profiles of $\lambda_{OH}$ are comparable for all flames, compare Figures 7.58(c) and 7.59(c). Like the integral length scales, the Taylor micro scales are minimum at the radial location of maximum average concentration. From the scattering of the measurements, as shown in Figure 7.58 and 7.59, we estimated the accuracy of the measurements to be 7%.

In the discussion of the simulated spatial correlation results we have shown that for realistic values of the fluctuations in peak value and OH zone location, the Taylor micro scale was in the same order of magnitude as the width of the OH zone. In Section 7.2.2 we found that the OH zone widths at $x = 50$ mm ranged from 1.45 mm in flame I to 0.98 mm in flame IV and was 1.7 mm in all flames at $x = 300$ mm (Table 7.4). Indeed, the values of the Taylor micro scale at the radial locations of the maximum average concentration are in the same order of magnitude as the OH zone widths. At $x = 50$ mm, the values of the minimum Taylor micro scales of flames I, II, III and IV are 0.63, 0.60, 0.71 and 0.94 mm, respectively. The opposed trend for flames II-IV is because of the increasing influence of holes in the OH zone with increasing exit velocities.

The minimum values of the Taylor micro scales for the different flames are given in Table 7.6, for $x = 50, 100, 250$ and 300 mm. We also show the centerline values of the Taylor micro scales for $x = 300$ mm for the flames where
7.11. Macro and micro scales from spatial correlation

Figure 7.59: Results of the Taylor micro scales $\lambda_{OH}$ for flame VI at $x = 100$ (a), 200 (b) and 300 mm (c).

Table 7.6: Taylor micro scales of OH, $\lambda_{OH}$, obtained at the radial location of maximum average concentration at $x = 50$, 100, 250 and 300 mm, for flames I to VI, and of the scaled Taylor micro scale obtained at the centerline, $\lambda_{OH|cl}$ at $x = 300$ mm, for flames IV, V and VI. The accuracy of $\lambda_{OH}$ is estimated to be 7%.

| flame | $\lambda_{OH}$ (mm) | $\lambda_{OH|cl}$ (mm) |
|-------|---------------------|-----------------------|
|       | $x = 50$ mm | $x = 100$ mm | $x = 250$ mm | $x = 300$ mm | $x = 300$ mm |
| I     | 0.63 ± 0.04 | 0.83 ± 0.06 | - | - | - |
| II    | 0.60 ± 0.04 | 0.81 ± 0.06 | 1.6 ± 0.1 | 1.9 ± 0.1 | - |
| III   | 0.71 ± 0.05 | 1.05 ± 0.07 | 1.7 ± 0.1 | 1.9 ± 0.1 | - |
| IV    | 0.94 ± 0.07 | 1.35 ± 0.10 | 2.0 ± 0.1 | 2.1 ± 0.1 | 2.5 ± 0.2 |
| V     | 0.87 ± 0.06 | 1.22 ± 0.09 | 2.0 ± 0.1 | 2.2 ± 0.2 | 2.7 ± 0.2 |
| VI    | 1.00 ± 0.07 | 1.24 ± 0.09 | 2.0 ± 0.1 | 2.3 ± 0.2 | 2.4 ± 0.2 |
the OH concentration fluctuations are significant. The soot scattering or PAH fluorescence observed for $x > 150$ mm in flame I, see Section 7.4.1, resulted in very large gradients at these locations, making calculation of the Taylor micro scale impossible. When scaled with the axial distance, the Taylor micro scales decrease with $x$. The minimum values decrease with a factor of about 2 between the values at $x = 50$ mm and 300 mm in all flames, indicating that the OH scales near the average location of the reaction zone are not proportional to $x$, as could be the case for Taylor micro scales of velocity in developed turbulence.

At $x = 50$ mm, the minimum Taylor micro scale of flame I is larger than that of flame II, probably due to the wider OH zone. Apart from this, we find that the Taylor micro scales slightly increase with exit velocities, and that the differences at $x = 300$ mm are relatively smaller than at $x = 50$ mm.

We compared the dissipation length scale, $\lambda_{4,\text{OH}}$ (from (7.3)) with the Taylor micro scale, $\lambda_{\text{OH}}$ (from (7.9)). Analysis of both (7.3) and (7.9) shows that when the standard deviation is independent of $y$, $\lambda_{4,\text{OH}} \approx \lambda_{\text{OH}}$. At locations in the flames where the radial gradients of $\langle c_s^2 \rangle$ are large, there is a difference between both scales. This is mainly the case for small $x$. In Figure 7.60, the profiles of both the dissipation length scale and the Taylor micro scale in flame III at $x = 50$ mm are shown. The differences are negligible near the radial location of maximum average concentration and towards the centerline, but are significant where the fluctuations change most strongly with $y$. The radial gradients in $\langle c_s^2 \rangle$ decrease with $x$ and the difference between the dissipation length scale and the Taylor

![Figure 7.60: Comparison of the Taylor micro scale $\lambda_{\text{OH}}$ (---) and the dissipation length scale $\lambda_{4,\text{OH}}$ (---) for flame III at $x = 50$ mm.](image-url)
micro scale decreases. Within the accuracy of the measurements, both scales are equal at $x = 250$ mm.

### 7.11.7 Summary of various length scales of turbulent OH fluctuations

In Table 7.7 we summarize the values of the various length scales of OH at $x = 300$ mm, discussed separately before: the average OH zone width $l_{OH}$, the minimum and maximum Taylor micro scale $\lambda_{OH}$ at $x = 300$ mm, the minimum and maximum integral length scale $L_{OH}$ at $x = 300$ mm, and the size of the largest eddies broadening the one-dimensional OH profiles, $L_{OH}$.

The minimum values of the Taylor micro scales of OH (at the radial location of maximum average concentration) and the average OH zone widths are of the same order of magnitude. Hence, in the case of a species which is confined to narrow regions, the length scale used to scale the spatial gradients is of the same order of magnitude as the average width of these regions. The minimum values of the Taylor micro scales increase with exit primary air velocity. Recall that the flame numbers are in the order of increasing primary air velocity, and that with increasing primary air velocity the flame becomes more turbulent and the OH zone becomes more strongly convoluted (Section 7.4.2). The maximum Taylor micro scales are less than a factor two larger than the minimum Taylor micro scales in all flames at $x = 300$ mm. In addition, the minimum and maximum values of the different flames differ less than 20 and 36%, respectively. Hence, for large $x$, the Taylor micro scales of the different flames are comparable and are not sensitive to the radial location.

At $x = 300$ mm ($x/d_1 = 50$) the maximum values of the integral length scales are in the range 6 to 10 mm, the Taylor micro scales are in the range

<table>
<thead>
<tr>
<th>flame</th>
<th>$l_{OH}$ (mm)</th>
<th>$\lambda_{OH}$ (mm)</th>
<th>$L_{OH}$ (mm)</th>
<th>$L_{OH}$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>min.</td>
<td>max.</td>
<td>min.</td>
<td>max.</td>
</tr>
<tr>
<td>I</td>
<td>1.7 ± 0.5</td>
<td>–</td>
<td>–</td>
<td>9.0 ± 0.8</td>
</tr>
<tr>
<td>II</td>
<td>1.7 ± 0.5</td>
<td>1.86 ± 0.12</td>
<td>2.78 ± 0.12</td>
<td>6.6 ± 0.6</td>
</tr>
<tr>
<td>III</td>
<td>1.7 ± 0.5</td>
<td>1.92 ± 0.12</td>
<td>2.47 ± 0.12</td>
<td>6.0 ± 0.5</td>
</tr>
<tr>
<td>IV</td>
<td>1.7 ± 0.5</td>
<td>2.10 ± 0.12</td>
<td>3.37 ± 0.12</td>
<td>0.92 ± 0.72</td>
</tr>
<tr>
<td>V</td>
<td>1.7 ± 0.5</td>
<td>2.20 ± 0.12</td>
<td>3.31 ± 0.12</td>
<td>1.03 ± 0.29</td>
</tr>
<tr>
<td>VI</td>
<td>1.7 ± 0.5</td>
<td>2.25 ± 0.12</td>
<td>3.37 ± 0.12</td>
<td>1.72 ± 0.20</td>
</tr>
</tbody>
</table>
2.5 to 3.4 mm. Hence, in the outer region of the flame, the ratio of the integral length scale and the Taylor micro scale is (2.8 ± 0.8), in all flames. In Section 8.9 we show that for the same \( x \), the ratio of the scales of a nonreactive scalar is about 4. Hence, the ratio \( \mathcal{L}_{\text{OH}}/\lambda_{\text{OH}} \) is somewhat smaller than the corresponding ratio of scales of a nonreactive scalar. In isothermal jets and flames, the reported ratios \( \mathcal{L}_u/\lambda_u \) or \( \mathcal{L}_T/\lambda_T \) are all in the same order of magnitude, for a wide range of \( x/d_j \): for \( x/d_j = 50 \), Pitz and Drake[97] found \( \mathcal{L}_u/\lambda_u \) in the range 2.5 to 3.4 in a hydrogen/air turbulent nonpremixed flame \((Re_j = 8.5 \times 10^3, d_j = 3.2 \text{ mm})\); for \( x/d_j = 20 \), Corrsin and Uberoi[95] found \( \mathcal{L}_u/\lambda_u = 2.4 \) and \( \mathcal{L}_T/\lambda_T = 1.8 \) in a heated jet; for \( x/d_j = 90 \), Wygnanski and Fiedler[94] found \( \mathcal{L}_u/\lambda_u = 4 \) at centerline and 16 at \( r/x = 0.15 \), in an isothermal jet.

At the radial location of maximum average concentration, the integral length scale is smaller than the Taylor micro scale, because the integral length scale is significantly affected by the chemical reactions producing and destroying OH. The largest integral length scales, found in the periphery of the OH profiles, are comparable for the different flames. This is in contrast with \( L_{\text{OH}} \), which is a measure of the distance over which OH is displaced by turbulence. The values of \( L_{\text{OH}} \) are larger due to the different definitions of both length scales used to represent the large scale eddies transporting OH. (In the outer parts of the OH concentration, the correlation functions are not strongly negative, so this can not be cause of the difference between \( \mathcal{L}_{\text{OH}} \) and \( L_{\text{OH}} \).) Observing the correlation functions shown in Figures 7.55 and 7.56, we see that \( L_{\text{OH}} \) and the largest distance with finite correlation are in the same order of magnitude.

The length scale, used to derive the large-eddy turnover time in Section 7.3, was the jet diameter (6 mm). This value appears to be in the same order of magnitude as the maximum value of the integral length scale in all flames.

7.12 Conclusions

Measurements of OH radical concentration fluctuations have been presented and analysed for flames under different conditions, where laser induced fluorescence was used with two-dimensional detection for visualization purposes and with one-dimensional detection for more quantitative analysis. Radial profiles of average OH concentration and temperature have been compared.

The lengths of the different flames showed that a flame with a large primary air velocity was shorter and thus more turbulent than the one with the smaller primary air velocity. This indicates a rather strong influence of this primary air flow on the flame structure. Exit velocities in the preheated flames are 2.4 times larger than in the cold-flow cases and this results in shorter flames, again indicating stronger turbulent diffusion. In the flames with large primary air velocity, the decrease of flame length agreed reasonably well with the theory of faster development of the heated primary air annulus jet: with a factor of 1.3
$\approx \sqrt{2.4}$. The flame with the smallest exit velocities and exit Reynolds number of about 5000, is significantly narrower than the flames with larger velocities. Hence, the shear layer turbulent is still developing in the range of $x$ covered. The flame with large primary air velocity and preheated flows is narrower. Hence, the large primary air velocity decreases the size of the largest eddies which are responsible for the flame width.

In the nonpreheated flames with primary air exit velocity about 5 times smaller than the fuel exit velocity, (flames I, II and III, with exit Reynolds numbers ranging from 5 to $10 \times 10^3$) the OH structure is only deformed by eddies of the largest scale, about 10 mm, indicating that the reaction zone is deformed by not well-developed turbulence. In flames with the Reynolds numbers of about $10^4$, some holes in the OH zone have been observed, indicating local extinction. The occurrence rate of holes is drastically increased by increasing the primary air flow rate. This also results in a more turbulent flame, as evidenced by a shorter flame and by stronger deformation of the OH structure. The holes in the OH structure are less frequent in the preheated flames. Preheating also gives a strong deformation of the OH zone, with a wider range eddy sizes. Due to the larger velocities in the preheated flames, the time scale of turbulence decreases with a factor of about 2.4, but laminar flamelet calculations showed that the chemistry time scale of two-body reactions decreases faster. Hence, the Damköhler number for two-body chemical reactions increases and local extinction is less frequent in the preheated flames compared to the nonpreheated flames with equal mass flow rate.

Temperature measurements showed that in the smallest velocity flames, the maximum average temperature decreased with axial distance. Increase of the exit velocities with a factor of two (changing from flame I to III), gives a decrease of the maximum average temperature at $x = 50$ mm of 246 K, but does not significantly change the maximum average temperature at $x = 250$ mm (the accuracy is about 50 K). The large primary air velocity of flame IV gives an extra decrease of maximum average temperature of 377 K at $x = 100$ mm compared to flame III. Comparing the preheated flame with relatively small primary air exit velocity, flame V, with the nonpreheated flame with equal mass flow rates, flame III, we find that for $x = 50$ mm, the maximum average temperatures differ less than 10 K, but for $x = 100$ mm and larger, the maximum average temperature in flame V is $(158 \pm 10)$ K larger. For $x = 50$ mm, preheating apparently does not influence the maximum average temperatures, whereas for larger $x$, preheating results in an increase of maximum average temperature that is constant with $x$. The maximum average temperatures of both preheated flames do not differ much, so the influence of a large primary air velocity is not as strong as in the nonpreheated flames.

Average profiles of OH concentration show that the maximum average concentration decreases with axial distance in all flames, except in the nonpre-
heated flame with large primary air velocity; in this flame there is strong local extinction close to the nozzle. Maximum average concentrations are largest in the smallest velocity flame, flame I. At $x = 50$ mm, this maximum average OH concentration is about $11 \times 10^{15}$ cm$^{-3}$ in this flame I, and only $2.8 \times 10^{15}$ cm$^{-3}$ in the nonpreheated flame with large primary air velocity, flame IV (accuracy about 10%). At $x = 300$ mm, the maximum average OH concentrations in the six flames with different mixing conditions and initial flow temperatures, differ less than 35%, with an average value of $4.0 \times 10^{15}$ cm$^{-3}$. The radial locations of maximum average OH concentration correspond to those of maximum average temperature.

Due to the manifestation of OH in narrow structures, the standard deviations of OH concentration are very large, also in the lowest velocity flames I and II, of which we show in the next chapter that shear layer turbulence is not completely developed for $x < 175$ mm. Hence, in these flames, the large standard deviations are expected to arise from instabilities like in a Von Kármán vortex street. At the radial location of maximum average OH concentration, the relative standard deviation at $x = 50$ mm increases with a factor 1.6 when the fuel and primary air exit velocity increased with a factor 2 and with another 51% by an extra increase of the primary air velocity: at $x = 50$ mm the relative standard deviation ranges from 0.32 in flame I to 0.77 in flame IV. The differences decrease with $x$. At $x = 300$ mm, the relative standard deviations of all flames differ less than 16%. The relative standard deviation in the preheated flames varies only 17% with $x$.

Intermittency and local extinction results in peaks in the distribution functions at concentration equal to zero. Only in the smallest velocity flames close to the nozzle, radial locations have been identified where the most probable concentration was at a large concentration of about $13 \times 10^{15}$ cm$^{-3}$, instead of zero. Maximum instantaneous concentrations are about $20 \times 10^{15}$ cm$^{-3}$. In the preheated flames this maximum concentration is slightly larger. The distribution functions generally consists of a peak at zero of finite width, together with a rather flat distribution at larger concentrations. This flat part suggests modelling of the distribution function with a combination of a Dirac delta function and a top-hat distribution. In some cases, however, the width of the peak at zero concentration is rather wide, and a $\beta$-function is more appropriate.

The one-dimensional detection provides measurement of the peak instantaneous concentration in every measurement. Distribution functions have been constructed for the fluctuations in these peak concentrations. In the smallest velocity flames the fluctuations of the concentration at a fixed location appear to be dominated by instabilities in the location of the OH zone, for the fluctuations of the peak value are significantly smaller than fluctuations of the value at the (fixed) location of maximum average concentration. When holes occur, the peak distribution function shows a peak at zero concentration in addition to a Gaussian shaped distribution for concentrations between about 5 and $15 \times 10^{15}$ cm$^{-3}$. The
probability of concentrations between these two cases is small, indicating that local extinction is associated with a minimum concentration. Local extinction are reduced by preheating.

Comparison of the distribution functions obtained with single emission-line detection and with broadband detection revealed differences which were quantitatively shown to be due to shot noise which was very significant in the single emission-line detection measurements. Hence, the systematic influence of temperature and composition on the broadband detected OH was found to be relatively small (about 10 %).

Joint distribution functions of concentration and instantaneous gradient showed that in the smaller velocity flames and close to the nozzle, large concentrations coincide with small gradients and large gradients with intermediate concentrations. For more developed turbulent fluctuations, large concentrations are associated with larger gradients and are thus not independent. In all flames with initially cold flows, the pairs of concentration and gradient are all in the same band, with a most probable concentration-gradient pair determining the different flames and locations.

Spatial correlations have been obtained from the one-dimensional OH profiles, resulting in values of both integral and Taylor micro length scales of OH concentration fluctuations. Also, we have determined the average width of the OH zone in perpendicular cross section and the maximum broadening of one side of the instantaneous OH profiles, as an indication of largest eddies wrapping up the OH structure. Simulations show that when the OH profiles consist of narrow zones with fluctuating location and peak level, the correlation functions obtain strongly negative values, giving often negative values of the integral length scale. This was observed in the experiments, where relevant integral length scale results were only to be obtained at the largest axial distances in the flames with the largest velocities and the strongest deformation of the OH structure. The integral length scales are smallest at the radial location where the average concentrations are maximum. This is because OH is not only transported by turbulent eddies, but also takes part in chemical reactions which decrease the correlation length. Minimum values of the integral length scales are about 1 mm. Much larger values are found in the outer regions of the flame, which are a better indication of the large-scale eddies transporting OH. In the different flames, these large scales range from 6 to 10 mm at $x = 300$ mm. The scales are largest in the flames identified as more turbulent, the preheated flames and the nonpreheated flame with large primary air velocity. This trend was also found with the large scale derived from the OH profile broadening, although values are nearly a factor of two larger due to the difference in definition of the length scales. Taylor micro scales have been determined for all flames. Like the integral length scales, the Taylor micro scales are smallest where the average concentrations are maximum and where gradients are largest. The difference for the different flames was largest for the smallest axial distance, yielding Taylor micro scales in the range of 0.6 to 1.0 mm at $x =$
50 mm, and in the range of 1.9 to 2.3 mm at $x = 300$ mm. The Taylor micro scales increase with exit velocities: they are smallest in the flame with the smallest exit velocities, and largest in the preheated flame with large primary air velocity.

The Taylor micro scales are in the same order of magnitude as the average OH zone width. At $x = 50$ mm, this width is largest in flame I, 1.45 mm, and decreases when the exit velocities and the additionally the primary air velocity are increased, giving a value of 0.98 mm in flame IV. Preheating increases the average OH zone width to 1.13 mm. At $x = 300$ mm, the average OH zone width is $(1.7 \pm 0.5)$ mm in all flames.

Although visual observation of the different flames and the visualization of OH showed a strong dependence of flame conditions, at the largest axial distances covered ($x/d_j \approx 50$) the average OH concentrations, the standard deviations, the distribution functions, the average OH zone width, and the Taylor micro scales of the different flames are close, when observed at the radial location of maximum average OH concentration.
Chapter 8

Visualization and fluctuations of NO seed concentrations in turbulent isothermal jets and turbulent flames

8.1 Introduction

In Section 5.3 we outlined several strategies for conserved scalar measurements in flames, one of which was seeding the fuel with NO. In this chapter, we discuss results of one- and two-dimensional measurements of NO concentrations in the case of NO seeded to nitrogen in isothermal jets and in the case of NO seeded to natural gas in the six different flames also discussed in Chapter 7.

We studied an isothermal jet of nitrogen issuing in low velocity secondary air and also one with a primary air flow, using the same configuration as in the flame experiments. Nitrogen was chosen instead of air, because oxygen in air might serve as an efficient quencher of excited NO. Our results are compared with literature on isothermal jets. Concentration fluctuation measurements in jets of air have been reported by Becker et al.[99] and Pitts[8,100], in jets of methane by Birch et al.[101], Pitts and Kashiwagi[102], and Schefer et al.[103], of scalar dissipation in isothermal jets of methane by Namazian et al.[104], of velocity and temperature fluctuations and length scales of turbulence in a heated jet by Corrsin and Uberoi[95], and of velocity fluctuations and length scales in an isothermal jet by Wygnanski and Fiedler[94]. By comparing the results of both jets, we analyze the influence of the annulus jet flow.

In addition to the study of turbulent mixing of "fuel" in isothermal jets, we investigated the turbulent mixing of fuel in the flames I–VI. Measurements of fluctuating concentrations have not been transformed into results of fluctuating mass or mole fractions, because no information on the local temperatures or densities was available. Also, excited NO is sensitive to quenching, especially by water vapour present in flames, as is shown in Section 8.2. We show that quenching reduces the total flux of excited NO molecules in flames significantly, but not in the isothermal jets. Conclusions on the mixture fraction distributions in the flames can therefore not be drawn from the NO measurements. The one- and two-dimensional NO measurements, however, still provide information on the
large scale mixing of the fuel flow with combustion products and fresh air, and on the influence of combustion on this mixing. Quenching is strongest near the reaction zone. Close to the nozzle the reaction zone does not coincide with the region of largest shear stresses. Therefore, in this region, large-scale turbulent structures in the shear layer are visible without strong interference of the NO fluorescence with quenching processes.

In the initial part of the flame, pure fuel is found in the central jet, and the NO fluorescence is not strongly influenced by quenching, except near the reaction zone. Qualitative comparisons therefore can be made with CH₄ measurements presented in literature. Namazian et al. [105] have compared CH₄ imaging results of a lifted turbulent non-premixed methane flame results of an isothermal jet of methane.

8.2 Estimation of NO quenching rates

In Section 8.3, it is shown that the total flux of excited NO molecules is not conserved in the flames, whereas it almost is in isothermal jets of nitrogen in issuing in air. Although the fluorescence is nearly saturated (Figure 5.12), strong quenching can still decrease the number of excited molecules, because of the broadband detection of NO fluorescence, see also Section 3.6.2.

McMillin et al. [71] give quenching cross sections for quenching of NO by various molecules, at a temperature of 2000 K. The quenching cross section of water vapour decreases with temperature, see Raiche and Crosley [106], but water is primarily found in the high temperature regions (Figure 8.1), so the value of the quenching cross section at 2000 K is used. The quenching cross section of nitrogen increases sharply with temperature, see Raiche and Crosley [106] and Thoman et al. [107]. It is negligible at room temperature and at 2000 K still rather small.

Hence, we did not implement the temperature dependence of the quenching cross section in our model. The calculated equilibrium mole fractions of these species are shown in Figure 3.3, and the calculated temperatures in Figure 8.1, as function of the mixture fraction $\xi$. The quenching rate is given by (3.12).

Both the quenching rates $Q_s$ for quenching of NO by species $s$ and the total quenching rate $Q$ are shown in Figure 8.2. For small values of the mixture fraction, the quenching rate is dominated by $O_2$. Near the reaction zone, quenching is dominated by $H_2O$ and $CO_2$, where the quenching rate of $CO_2$ remains dominant in the fuel rich part (large $\xi$). CO also plays an important role for mixture fraction values around 0.2. Due to the high nitrogen content of the natural gas used, $N_2$ has an almost constant contribution to the quenching rate. The total quenching rate decreases with $\xi$. The effect of this quenching is shown in Section 8.3.
8.2. Estimation of NO quenching rates

Figure 8.1: Laminar diffusion flame in equilibrium: temperature (a) and mole fractions of O₂ (---), N₂ (--), H₂O (---), CO₂ (----) and CO (-----) (b), as function of the mixture fraction ξ.

Figure 8.2: Estimated quenching rates $Q_s$ of NO by species O₂ (---), N₂ (--), H₂O (---), CO₂ (----) and CO (-----), and the total quenching rate $Q = \sum_s Q_s$ (---, upper curve), calculated for a laminar nonpremixed flame in equilibrium, with temperature and composition given in Figure 8.1.
8.3 Is NO seeded to the fuel a conserved scalar in flames?

In this section, we investigate to what extent the NO fluorescence signal can be used to represent a conserved scalar, both in isothermal jets and in flames. The fluorescence signal is proportional to the concentration, which in its turn is proportional to the density and the mass fraction.

To measure the axial distance dependence of the total flux of fluorescing NO molecules, three situations have been investigated: first a free NO-seeded nitrogen jet issuing in low velocity (0.3 m/s) secondary air flow. This jet is called jet I, with an average exit jet velocity equal to that of flame III. The exit Reynolds number of this jet, based on the average exit velocity and the nozzle diameter, is $7.7 \times 10^3$. Secondly a free NO-seeded nitrogen jet with air flow from the annulus issuing in a low velocity coflow, called jet II. The average exit velocities of the central jet, of the annulus flow and of the coflow are equal to those of flame III. We compared the results of jet I and jet II with the results obtained in flame III, defined in Section 4.7, also discussed extensively in Chapter 7.

In the isothermal jets I and II, and in the flames I–VI, we mixed NO with fuel with a volume fraction of 0.74 %, inserted into the fuel flow at about 2.5 m (or about 300 tube diameters) upstream the burner exit. The reproducibility of this initial volume fraction is about 15 %, whereas the volume fraction did not vary more than about 5 % during the measurements of every jet or flame.

The chemical reactions removing NO are slow compared to turbulent mixing processes present in our flames[20,70]. Conservation of the NO concentration, however, does not imply conservation of the number of fluorescing NO molecules or, equivalently, of the NO fluorescence signal: quenching of the fluorescing NO molecules results in loss of fluorescence signal, see Stärner et al.[67]. Water vapour, oxygen, carbon monoxide, carbon dioxide and nitrogen are species with large quenching cross sections that are also present in significant amounts.

8.3.1 Conservation equations

The fluorescence signal is proportional to the number of fluorescing molecules and constants such as optical parameters and Einstein coefficients of spontaneous emission. (In this we neglect the difference in Einstein coefficients of the various rotational levels that are populated by rotational relaxation, which is a reasonable assumption in the case of NO.) The relation between the total flux of fluorescing molecules and the total flux of molecules, involves the Boltzmann factor, saturation, quenching and relaxation rates, which are to some extent sensitive to composition and/or temperature, see Chapter 3.6.2. In the absence of quenching, the life time of NO fluorescence (220 ns[106,108]) is longer than the gating time (40 ns). The quenching decreases the fluorescence life time, but due to the short gating time initially this does not strongly affect the fluorescence signal. The combined effect of NO disappearing by chemical reactions and the
influence of temperature or composition fluctuations on the relation between the NO concentration and the fluorescence signal (or the number of fluorescing NO molecules) follows from the check of fluorescing molecules conservation.

We like to know whether the fluorescence signal of NO seeded to the fuel of a jet, is conserved. Then, \( \langle \rho Y_{NO} u \rangle \) should be conserved, where \( Y_{NO} \) is the mass fraction of NO. We assume that:

\[
\langle \rho Y_{NO} u \rangle \approx \langle \rho Y_{NO} \rangle \cdot \langle u \rangle. \tag{8.1}
\]

According to (2.18):

\[
\langle \rho Y_{NO} \rangle \cdot \langle u \rangle \propto \langle c_{NO} \rangle \cdot \langle u \rangle. \tag{8.2}
\]

In an axisymmetric configuration, the conservation of the number of NO molecules gives a constant flux of the number of NO seeding particles at all \( x \). This flux is then approximately given by:

\[
\int_0^\infty 2\pi \langle c_{NO}(x, r, t) \rangle \cdot \langle u(x, r, t) \rangle r dr = \text{constant}. \tag{8.3}
\]

To check whether the flux of fluorescing NO molecules is conserved, we use the fact that the number of fluorescing molecules is directly proportional to the fluorescence signal and we replace \( c_{NO} \) in (8.3) by the fluorescence signal \( S_F \). The resulting integral is allowed to be a function of \( x \):

\[
\int_0^\infty 2\pi \langle S_F(x, r, t) \rangle \cdot \langle u(x, r, t) \rangle r dr = I(x). \tag{8.4}
\]

As discussed in Section 8.2, the quenching rates are larger for smaller values of the mixture fraction and thus for larger radial distances. On the other hand, the maximum mixture fraction decreases with \( x \), resulting in quenching rates increasing with \( x \). This implies that if quenching affects the number of fluorescing NO molecules, this number will decrease with \( x \). Oxygen is an effective quencher, so in the isothermal jets the total flux of fluorescing NO molecules might also decrease. (Recall that the quenching rate is proportional to \( T^{-1/2} \).) The levels of oxygen in a flame are lower due to the consumption of oxygen, but the additional quenching by water vapour, carbon monoxide and carbon dioxide will give a stronger quenching in the flame than in the isothermal jets.

8.3.2 Decrease of the number of fluorescing NO molecules with axial distance

We evaluated the integration of (8.4) for jet I, jet II and flame III, using velocity measurements carried out by Stroomer[2]. Measurements of the average axial velocity in the isothermal jet I are available for \( x = 50 \) to 250 mm, in isothermal jet II for \( x = 50 \) to 200 mm, and in flame III for \( x = 50 \) to 250 mm, all at 50 mm intervals. NO fluorescence profiles have been measured for \( x = 50 \) to 250 mm
in both isothermal jets and for $x = 50$ to $300$ mm in flame III, again at $50$ mm intervals.

In Figure 8.3 we show the values of the integral (8.4) for the different values of $x$, where the integral is normalized on its largest value at $x = 50$ mm. The accuracy is about 10%. $I(150$ mm) of jet I is 5% larger than unity, a deviation that is within the accuracy of the measurement. In both jets and in the flame, the values of $I(x)$ at $x = 50$ mm and $100$ mm are equal, given the accuracy of measurement. Hence, we can assume these values are also equal to the value at $x = 0$: $I_0 \equiv I(0) = I(50$ mm) = $I(100$ mm). Due to small differences in the initial conditions of the seeding concentration, of the exit velocity and due to accuracy of the evaluation of (8.4), the value of $I(50$ mm) for jet I is somewhat smaller than for jet II, whereas the values for jet II and flame III are equal.

Figure 8.3 shows that in both isothermal jets the decrease of total flux of NO molecules is less than 10%, which is not significant. Hence, quenching by O$_2$, which is the only candidate for quenching in the isothermal jets, appears not to influence the NO fluorescence signal significantly, maybe because of the short gating time. $I(x)/I_0$, however, significantly decreases with $x$ in flame III. About 50% of the fluorescing NO has disappeared at $x = 250$ mm (which is about 30% of the flame length) and this decrease depends almost linearly on $x$ for $x > 100$ mm. In flames, the reduction of NO fluorescence signal is mainly due to quenching by H$_2$O and CO$_2$ and to a lesser extent by CO. In addition to the quenching, the decrease in the total flux of fluorescing NO molecules in the flame is also caused by the temperature dependence of the Boltzmann factor, which could not be prevented completely despite the careful selection of the absorption line (Section 5.7.3).
In the next section, we relate the decrease in the amount of fluorescing NO molecules to a change in the average fluorescence profiles.

### 8.3.3 Change of the fluorescence profiles due to quenching

We want to know in what way the decrease in number of fluorescing NO molecules changes the average radial fluorescence profiles. Direct comparison of the average radial profiles of NO is not possible, because the higher temperatures at the sides of the NO profiles decrease the density, giving narrower concentration profiles. Analogously, the velocity profiles of flame III are wider than those of the jet with annulus flow, jet II, because of the thermal expansion in the flame. The profiles of \( \langle S_F \rangle \cdot \langle u \rangle \), however, are not very sensitive to the changing density in the flame.

The profiles of \( \langle S_F \rangle \cdot \langle u \rangle \) are approximately Gaussian in shape. The full-width-at-half-maximum \( w_{SU} \) of these profiles of jet II and of flame III can be compared to see the effect of the flame on the radial distribution of the fluorescing NO concentration. Values of \( w_{SU} \) are given in Table 8.1.

#### Table 8.1: Width (FWHM) \( w_{SU} \), scaled with \( x \), of the profiles of \( \langle S_F \rangle \cdot \langle u \rangle \) of jet I, jet II and flame III. The estimated accuracy is 5%.

<table>
<thead>
<tr>
<th></th>
<th>( x/d_j )</th>
<th>( w_{SU}/x )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(mm)</td>
<td>(mm) jet I</td>
</tr>
<tr>
<td>50</td>
<td>8.3</td>
<td>0.122</td>
</tr>
<tr>
<td>100</td>
<td>16.7</td>
<td>0.119</td>
</tr>
<tr>
<td>150</td>
<td>25.0</td>
<td>0.126</td>
</tr>
<tr>
<td>200</td>
<td>33.3</td>
<td>0.133</td>
</tr>
</tbody>
</table>

In jet I, the scaled width \( w_{SU}/x \) slightly increases with \( x \) for \( x \geq 100 \) mm; the average value is 0.13. In jet II, the primary air flow results in a narrower width, the scaled width reaching a value of 0.10 for larger \( x \).

#### Table 8.2: Centerline values of \( \langle S_F \rangle \cdot \langle u \rangle \), multiplied by \( x^2 \) for scaling, of jet I, jet II and flame III. The estimated accuracy is 10%.

<table>
<thead>
<tr>
<th></th>
<th>( x/d_j )</th>
<th>( \langle S_F(x,0,t) \rangle \cdot \langle u(x,0,t) \rangle \times x^2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(mm)</td>
<td>(10^2 counts·m⁻³/s) jet I</td>
</tr>
<tr>
<td>50</td>
<td>8.3</td>
<td>1.78</td>
</tr>
<tr>
<td>100</td>
<td>16.7</td>
<td>1.96</td>
</tr>
<tr>
<td>150</td>
<td>25.0</td>
<td>1.94</td>
</tr>
<tr>
<td>200</td>
<td>33.3</td>
<td>1.64</td>
</tr>
</tbody>
</table>
For all $x$ covered, the scaled width of flame III is about 20% narrower than that of jet II. The quenching is strongest for lower values of the mixture fractions, and thus for larger radial distances. This quenching gives a narrower profile of $\langle S_F \rangle \cdot \langle u \rangle$. It should be noted that the total flux of fluorescing NO molecules is proportional to the square of the width.

Let's also observe the axial dependence of the centerline value $\langle S_F(x, 0, t) \rangle \cdot \langle u(x, 0, t) \rangle$. The values of the centerline value are given in Table 8.2. The profiles of $\langle S_F \rangle \cdot \langle u \rangle$ of jet II being narrower than those of jet I, corresponds to the fact that the centerline values are larger, resulting in a constant value of the integral value of (8.4). For $x = 50$ to 150 mm, the centerline values in flame III are about twice as large as in jet II, but the centerline value of flame III decreases faster than that of jet II. In combination with the smaller width $w_{SO}$, this results in a total flux of fluorescing NO molecules being almost constant for $x \leq 100$ mm, and decreasing linearly for larger $x$.

### 8.3.4 Conclusions about conservation

In the isothermal jets, both with and without annulus flow, the NO fluorescence signal does not change more than about 10%, which is an insignificant change. Hence, we consider the NO concentration in the isothermal jets to be conserved. Quenching with the most efficient quenching present in these flows, $O_2$, apparently did not strongly influence the total flux of fluorescing NO molecules for a given $x$. In flame III, the total flux decreased with about 50% for $x = 50$ to 250 mm. The product of the average fluorescence concentration and the average axial velocity is not influenced by the density in the flame. Due to the faster quenching in the case of the flame, mainly for smaller values of the mixture fraction, the width of the profiles of $\langle S_F \rangle \cdot \langle u \rangle$ increases slower with $x$ in the flame than in the isothermal jet and the center value decreases faster with $x$, resulting in a decrease with $x$ of the total flux of fluorescing NO.

### 8.4 Visualization of NO seed in flames

In this section we discuss the results of two-dimensional visualization of NO used as fuel marker. The visualization results of the isothermal jets were of poor quality due to nonhomogeneous laser intensity distribution in the laser sheet and due to the fact that the images were recorded in the VHS instead of the S-VHS mode. Results of isothermal jet visualization are therefore not presented here. For the isothermal jet results, see the discussion of the one-dimensional measurements in Section 8.5 and following. The visualization results obtained in flames are best suited for investigating the large scale mixing of fuel with combustion products and air. This mixing originates in the shear layer between the fuel and air flows.

The conclusions from Section 8.3 should be kept in mind in this latter comparison: the NO fluorescence signal does not represent a conserved scalar,
because quenching removes excited NO molecules, especially for mixture fraction values smaller than about 0.2. Fortunately, for small $x$, the region with most intense shear layer mixing is closer to the jet axis (Section 2.2) so that the shear layer mixing is still visible in the images.

The images show the existence of large-scale structures at axial locations where the OH structure was only little deformed. As pointed out in Section 2.2 this due to the fact that for not too large $x$ OH is found at the outer boundaries of the shear layer between fuel and air. The eddies in the shear layer and near the reaction zone, are shown by contour plots of NO-marker images with contours in the shear layer and where OH is expected.

### 8.4.1 Instantaneous images

In Figures 8.4–8.9, images are shown of NO in flames I–VI. Recapitulating Table 4.2, in flame I, II and III, the fuel and primary air exit velocities have been increased subsequently, keeping the ratio of fuel exit velocity and primary air velocity constant. In flame IV the primary air velocity is nearly twice as large as that of flame III. Flames V and VI are preheated, where the fuel and primary air mass flow rates of flames V and VI are equal to those of flames III and IV, respectively.

In every figure, we show two different images for every region of $x$. The regions cover $x = 25$ to 75 mm, 75 to 125 mm, and 125 to 175 mm. It should be noted that the camera gain was adjusted at every location, to match the signal of the maximum concentration to the maximum signal allowed by the camera. The axial decrease of concentration is therefore not apparent.

In Figure 8.4 we see that for $x = 25$ to 75 mm in the flame with small exit velocities, flame I, the interface of NO-containing fuel mainly consists of large-scale structures that start to develop from about 35 to 40 mm from the nozzle. In the region $x = 75$ to 175 mm, in many images the NO-containing fluid is shows periodicities, with a wavelength of 10 to 13 mm. With a velocity in the order of magnitude of 5 m/s, the frequency of these periodicities is about 450 Hz. The structure resembles a Von Kármán vortex street, see Tritton[109]. The periodic structures are not equally clear in every instantaneous image, as can be seen in Figure 8.4, and the periodicities are not stable. The images of Figure 8.4 show that entrainment of combustion products and air into the fuel jet, is only by large eddies. Because these large-scale eddies are not accompanied by smaller eddies, we find that shear layer turbulence is still developing. In this flame, the maximum concentration did not decrease significantly in the range of $x$ between 25 and 175 mm, and the camera gain was equal at the three stations.

In Figure 8.5 of flame II, with exit velocities 40 % larger than in flame I, it can be seen that large eddies pass almost periodically in the region $x = 25$ to 75 mm, alternating at both sides in the two-dimensional cross section. The separation between these eddies is about 7 mm, with a velocity of about 7.5 m/s
Figure 8.4: Instantaneous images of NO as fuel-marker in flame 1, covering $x = 25$ to $75$ mm, $75$ to $125$ mm and $125$ to $175$ mm. For every region, two different images are shown.
Figure 8.5: Instantaneous images of NO as fuel-marker in flame II, covering $x = 25$ to $75$ mm, $75$ to $125$ mm and $125$ to $175$ mm. For every region, two different images are shown.
Figure 8.6: Instantaneous images of NO as fuel-marker in flame III, covering $x = 25$ to $75$ mm, $75$ to $125$ mm and $125$ to $175$ mm. For every region, two different images are shown.
Figure 8.7: Instantaneous images of NO as fuel-marker in flame IV, covering $x = 25$ to $75$ mm, $75$ to $125$ mm and $125$ to $175$ mm. For every region, two different images are shown.
Figure 8.8: Instantaneous images of NO as fuel-marker in flame v, covering $x = 25$ to $75$ mm, 75 to $125$ mm and 125 to $175$ mm. For every region, two different images are shown.
8.4. Visualization of NO seed in flames

Figure 8.9: Instantaneous images of NO as fuel-marker in flame VI, covering $x = 25$ to $75$ mm, $75$ to $125$ mm and $125$ to $175$ mm. For every region, two different images are shown.
in the shear layer this gives a frequency of 1 kHz (the jet velocity is about 15 m/s). The large-scale eddies start to develop from \( x \approx 28 \text{ mm} \). In the region \( x = 75 \text{ to } 125 \text{ mm} \), the interface is less structured, and smaller eddies have been produced. Even in the region \( x = 125 \text{ to } 175 \text{ mm} \), however, structured large scale motion of the NO is visible, indicating that the shear layer turbulence is still developing.

From the smallest axial distances covered in flame III, \( x \approx 25 \text{ mm} \), small-scale structures are present in addition to the larger structures and no periodic alternation of large-scale structures is observed, see Figure 8.6. This is in contrast with the smaller Reynolds number flames II and I, where large scale structures are very dominant close to the nozzle. The instantaneous NO distribution in the two-dimensional cross section is significantly wider than in flames I and II. At some locations, the instantaneous NO distribution is totally broken, indicating either locally completely disappeared NO (due to quenching), NO being displaced by combustion products or air, or NO having moved outside the cross section plane. These effects are all due to turbulence, influencing the flow at centerline.

The NO structure of flame IV is very similar to that of flame III for not too large axial distances. The larger primary air velocity apparently does not strongly influence the central fuel jet flow in the region \( x = 25 \text{ to } 125 \text{ mm} \), except for a slightly more frequently broken two-dimensional NO structure. This is in contrast with the OH, which is strongly influenced by the larger primary air velocity. In the region \( x = 125 \text{ to } 175 \text{ mm} \), the larger primary air exit velocity gives a more homogeneous spatial NO distribution, with smaller maximum concentrations.

The structure of NO in the preheated flames V and VI, does not differ much from the nonpreheated counterparts, flames III and IV, compare Figures 8.6 and 8.8, and Figures 8.7 and 8.9. The gain in the preheated flames could not be raised to have a signal in the region \( x = 125 \text{ to } 175 \text{ mm} \) as large as in the nonpreheated flames, because the seeding concentration is smaller in the preheated flames due to thermal expansion.

### 8.4.2 Contours near shear layer and reaction zone

It was stated before, that for not too large axial distance OH is not found near the shear layer. We illustrate this with contours in the images of the NO fuel marker, where the shear stresses are largest and where OH is expected. Although the real mixture fraction cannot be derived from the NO fluorescence signal, we can qualitatively locate the reaction zone. We assumed that the reaction zone was located where the NO fluorescence signal is approximately 5% of the maximum fluorescence signal. The shear layer is indicated with the contour where the NO fluorescence signal is 50% of the maximum value.

In Figure 8.10, the two contours of the NO signal are shown of the regions \( x = 25 \text{ to } 75 \text{ mm} \), and \( 75 \text{ to } 125 \text{ mm} \) of flame II, and these images correspond to the instantaneous NO-marker images of this flame shown on the left hand side.
8.4. Average NO seed profiles

Figure 8.10: Contours indicating NO fluorescence signals equal to 0.5 (inner contours) and 0.05 times the maximum fluorescence signal, of flame II for $x = 25$ to 75 mm (a), and 75 to 125 mm (b). The corresponding instantaneous images of NO can be found in Figure 8.5.

of Figure 8.5. In the region $x = 25$ to 75 mm, the small-scale frays in the outer contour at the right hand side, are attributed to the smaller signal-to-noise ratio at this side of the flame, where the laser energy is smaller due to absorption, in combination with the small signal at the location of the outer contours. Apart from this, the outer contour does not deviate far from a straight line. No wrapping up of the outer contour occurs, in correspondence with the OH structure in this flame (Figure 7.4). The contour closest to the axis indicates the shear layer and clearly shows the existence of large-scale structures present in the shear layer. Only little influence of these structures is noticed on the outer contour, and thus on the reaction zone. This was also found by Chen and Goss[10], in their visualization of OH together with particles seeding the fuel flow. In the region $x = 75$ to 125 mm, the outer contour shows the start of a convolution.

In Figure 8.11 we show the contours of an instantaneous NO image in the region $x = 25$ to 75 mm of flame III, corresponding to the lower left instantaneous image of this flame shown in Figure 8.6. Like in flame II, the inner contour indicating the shear layer, displays large-scale eddies that have only little influence on the outer contour indicating the reaction zone, which shows only some large-scale deviations from a straight line.

The above results confirm the idea that the OH is too far from the shear layer to be curled by shear layer structures in most flames for not too large axial distances.
8.5 Average NO seed profiles in isothermal jets and turbulent flames

In this section, we discuss the average profiles of the NO fluorescence signal, both in the two isothermal jets and in the turbulent flames. In Section 8.3 it has been discussed that the NO fluorescence is influenced by quenching processes in the presence of combustion, and that this affects both the widths and the centerline values of the profiles. We investigate the influence of the primary air flow on the isothermal jet. We discuss the shear layer mixing characteristics of the different flames, giving different average concentrations, widths and standard deviations. We compare the profiles with fitted Gaussian shaped profiles. The average profiles are presented in a way that the radial coordinate is divided by the axial distance, and the fluorescence signal is multiplied by the axial distance. It is not suggested that this is the best scaling to map all profiles, but it is only to have a radial extent and centerline value in the same order of magnitude for all axial distances. Better scaling would require determination of extra parameters that depend on the location and the flow conditions.

8.5.1 Average radial profiles in isothermal jets

In Figure 8.12, the scaled profiles are shown of the nitrogen jet without primary air flow, jet 1. At \( x = 50 \text{ mm} \) \( (x/d_j = 8.3) \), we find that the scaled average concentration profile is slightly wider than at larger \( x \) and also that the scaled centerline value is larger. This is due to a not completely developed turbulent jet flow at this axial distance. Comparing the shape of the concentration profile
at $x = 50$ mm with a Gaussian shaped profile, we find that the actual profile is slightly too narrow near the centerline. The scaled profiles at $x = 100$ mm ($x/d_j = 16.7$) and $150$ mm ($x/d_j = 25$) map very well; in Figure 8.13 it is shown that at $x = 150$ mm (and therefore also at $x = 100$ mm), to a very good approximation the NO concentration profile is Gaussian in shape.

For $x = 200$ mm ($x/d_j = 33.3$) and $250$ mm ($x/d_j = 41.7$), the scaled centerline concentration is somewhat smaller than for $x \leq 150$ mm, and the concentration profiles have a flat part around the jet axis, giving a deviation from the Gaussian shaped profile that matched the profiles at $x = 100$ and $150$ mm. This only occurs in the widest profiles of all: for the largest values of $x$ in jet I, which is wider than jet II and than all flames. This flat part is caused by off-axis signals being slightly too large because of the laser beam divergence: fluorescence is saturated (Section 5.12), and the decrease of laser intensity, associated with the diverging laser beam, does not affect the fluorescence signal, whereas the divergence gives a measuring volume, increasing with distance from the focus at centerline.

The centerline values of jet II decrease with $x$ with the same amount as in jet I. The profiles of jet II are narrower than of jet I. The influence of increasing measuring volume is not noticed in jet II. At $x = 50$ mm, the profiles of jet I and jet II have the same width but a slightly different shape: for jet II the slopes of the profile are steeper.
Figure 8.13: Comparison of the average NO-marker concentration in jet I at $x = 150$ mm (—), with a Gaussian shaped profile ( - - ).

Stroomer[2] found that in jet II the centerline average axial velocity decreases slower with $x$ than in jet I. At $x = 250$ mm, the average centerline velocity of jet I and jet II are 0.25 and 0.45 times the centerline velocity at $x = 50$ mm, respectively. Hence, the annulus jet flow suppresses development of the central jet: the slower decrease of the axial velocity in jet II is associated with narrowing of the scaled NO-seed profiles, with conservation of the NO concentration, see Section 8.3.

### 8.5.2 Average radial profiles in flames

The average NO fluorescence profiles of flame III are shown in Figure 8.16. The density is of course not constant in the flame. Near the reaction zone, in the outer region of the jet, this gives a decrease in the NO concentration, and consequently a narrower profile. Also, the centerline temperature increases with $x$, giving a centerline concentration decreasing faster with $x$ than in the isothermal situation of jet II.

For $x \leq 150$ mm, the average centerline signals are larger in the flame than in the jet. The shape of the profiles is again Gaussian to a very good approximation, see for $x = 150$ mm Figure 8.17.

The profiles of the other flames are qualitatively equal in shape to those of flame III. We discuss the axial dependence of the centerline fluorescence signal and of the width in the following sections.
Figure 8.14: Average NO-marker concentration in jet II, for scaling multiplied by $x$, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (—), $100$ (— —), $150$ (— — —), $200$ (— — —) and $250$ mm (— — —); this corresponds to $x/d_j = 8.3$, 16.7, 25.0, 33.3 and 41.7, respectively.

Figure 8.15: Comparison of the average NO-marker concentration in jet II at $x = 150$ mm (—), with a Gaussian shaped profile (— —).
Figure 8.16: Average NO-marker concentration in flame III, for scaling multiplied by $x$, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (---), 100 (____), 150 (----), 200 (-----) and 250 mm (-----); this corresponds to $x/d_j = 8.3, 16.7, 25.0, 33.3$ and 41.7, respectively.

Figure 8.17: Comparison of the average NO-marker concentration in flame III at $x = 150$ mm (---), with a Gaussian shaped profile (---).
8.5. Average NO seed profiles in isothermal jets and turbulent flames

Figure 8.18: Average NO fluorescence signal at centerline, multiplied by $x$, as function of axial distance $x$, jet I (---), jet II (--x--), flame I (--o--), II (--$\Delta$--), III (--$\square$--), IV (--$\circ$--), V (--$\square$--) and VI (--$\circ$--).

8.5.3 Axial dependence of average centerline signal

The centerline NO fluorescence signals of both isothermal jets and of flame III have already been discussed in the previous section and can be found in Figures 8.12, 8.14 and 8.16. We now compare the centerline NO signals of both jets and of all flames. The results are shown in Figure 8.18. For sake of clarity, we multiplied the centerline signals with $x$, because the simple value of the signal gives little discrimination for large $x$ where the signals are small, and the inverse of the signal gives little discrimination for small $x$ for the same reason. Recall that the fluorescence signal $S_F$ is proportional to the concentration of the seed molecules, so $\langle S_F \rangle \propto \langle \rho Y_{NO} \rangle$, see Section 2.4.

At $x = 50$ mm, the centerline value of flame I to IV are about 40% larger than the centerline value of jet II at this value of $x$. In Section 8.3 we found that the flux of NO particles in jet II and in flame I are very close. Hence, the centerline concentrations of NO are larger in the flames because of a smaller entrainment of air into the fuel jet: this will be discussed more extensively in Section 8.8, when the distribution functions of NO fluctuations are considered. For larger $x$, the centerline values approach the values of the isothermal jets and become lower for $x \geq 150$ mm, due to changing density and the quenching of excited NO.

In the flame with lowest exit velocities, flame I, the centerline value decreases very slowly with $x$ for $x \leq 150$ mm, giving a scaled centerline value which is larger at $x = 100$ mm than at $x = 50$ mm. In all other flames and in both
isothermal jets, however, the decrease with \( x \) is proportional to \( x^{-1} \) or faster. This slower decrease of the centerline concentration in flame I is reproduced by other measurements and indicates a slower jet development, which can also been seen in the visualization results, see Figure 8.4. In flame II, with 40 % larger exit velocities than flame I, this decrease is not significantly slower than in flames III and IV.

The centerline value of the flame with large primary air velocity, flame IV, is at \( x = 50 \), 200 and 250 mm 16 % larger, and at \( x = 100 \) and 150 mm 10 % larger. The maximum instantaneous signal at \( x = 50 \) in flame IV is about 15 % larger than in flame III, indicating that the seeding concentration in flame IV has been larger by this amount. Correcting for this effect by matching the maximum signals, we find that for the centerline signals in flame IV are almost equal to those in flame III for \( x \leq 250 \) mm, and about 15 % smaller for \( x = 300 \) mm. Thus, the influence of the larger primary air velocity on the centerline is not very strong with respect to the NO as fuel maker for \( x \leq 250 \) mm, but is expected to become stronger for larger \( x \).

The preheated flames have a centerline value which is about 2.5 times smaller for all \( x \). The initial seeding volume fraction in the preheated flames V and VI is equal to those in the other flames. The fluorescence signal, however, represents NO concentrations. Preheating to a temperature of nearly 700 K, gives a decrease of the density, with a factor of about 2.4, which agrees with the reduction of the average centerline signal at \( x = 50 \) mm. For larger \( x \) the entrainment of cold secondary air decreases the difference in centerline temperature with that of the nonpreheated flames. This would decrease the difference in NO concentration between the preheated and nonpreheated flames. The signals, however, are 2.5 times smaller for all \( x \). Hence, the entrainment in the preheated flames is larger than in the other flames. The difference in the centerline signal between the flames V and VI is smaller than 10 % for all \( x \) covered, so like in the flames III and IV, again the influence of the large annulus velocity is not very strong for the range of \( x \) covered.

8.6 Width of average NO seed profiles in isothermal jets and flames

The average and instantaneous NO concentration profiles provide information on the spread of the central jet flow in the isothermal jets and the different flames. We compare the widths of the NO profiles with those of the average axial velocity profiles, for jet I. We investigate the influence of the primary air flow, and of the influence of flame conditions on the width of the NO profiles. The comparison of the widths of the NO profiles of the jets with those of the flames are obscured by the density varying with radial location and the effect of quenching, most prominent in the outer regions of the jet. In Section 8.3 we have shown that the
8.6. Width of average NO seed profiles in isothermal jets and flames

![Graph](image)

Figure 8.19: Width of NO profiles, $w_{\text{NO}}$ (---) and of axial velocity profiles, $w_u$ (-----), of jet I.

radial profiles of $\langle S_F \rangle \cdot \langle u \rangle$ are about 20% narrower in flame III than in jet II. The width used here is the full-width-at-half maximum (FWHM) of the NO profile, indicated with $w_{\text{NO}}$.

8.6.1 Comparison of average NO and axial velocity profiles in an isothermal jet

For jet I, we compare the average profiles of the NO concentration and of the axial velocity, measured by Stroomer[2]. The values of the width of the average axial velocity profiles, $w_u$, and of the average NO concentration profiles, $w_{\text{NO}}$ are given in Figure 8.19. We find that, except for $x = 50$ mm, the NO profile of jet I is wider than the axial velocity profile. For $x = 250$ mm ($x/d_j = 41.7$) the NO concentration profile is 1.3 times wider than the axial velocity profile. This larger spread of concentration profiles compared to the axial momentum profiles is related to the turbulent Schmidt number.

The turbulent Schmidt number is discussed by Forstall and Shapiro[110], cited by Hinze[91]. They found a similar value of the ratio of concentration and velocity width. They obtained results in a helium seeded jet of air issuing in coflowing air ($d_j = 6.4$ mm, $x/d_j \leq 136$, jet velocities $U_j$ ranged from 27 to 67.5 m/s, coflow velocities $U_c$ ranged from 13.5 to 27 m/s, $Re_j$ ranged from $1.3 \times 10^4$ to $3.2 \times 10^4$). At $x/d_j = 50$, Forstall and Shapiro[110] found a concentration profile being $(1.26 \pm 0.04)$ times larger than the velocity profiles, independent of the initial velocity ratio and a turbulent Schmidt number of 0.70, also independent
Figure 8.20: Full width at half maximum of the average NO-seed profiles, $w_{NO}$ of jet I (−−−), jet II (−×−), flame I (−○−), II (−Δ−), III (−□−), IV (−○−), V (−○−) and VI (−○−).

of the initial velocity ratio.

For jet I, linearization of the NO profile width yielded $w_{NO} = (0.23 \pm 0.02)(x - (2.5 \pm 1.5)d_j)$, see also Table 8.3. For $x/d_j = 30$ this gives a value for $w_{NO}/x$ of 0.211. This is very close to the width scaled with $x$ found by Birch et al.[101] for a natural gas (95% methane) jet issuing into air ($d_j = 12.65$ mm, $Re_j = 1.6 \times 10^4$). They found a width equal to 0.194$x$, for a range of $x/d_j$ between 10 and 40 (we measured from $x/d_j = 8.3$ to 41.7). Becker et al.[99] found the same value in a jet of air, ($d_j = 6.35$ mm, $Re_j = 5.4 \times 10^4$), despite the difference in density and in Reynolds number. The widths found by Birch et al. and by Becker et al. agree very well with our results; the difference ranges from −4% at $x/d_j = 10$ to 6% at $x/d_j = 30$.

Concluding, the NO concentration profiles are 1.3 times wider than the average axial velocity profiles because the turbulent Schmidt number is about 0.70, and the width of the NO profiles agree very well with the values for the concentration width found by Birch et al.[101] and by Becker et al.[99].

8.6.2 Influence of primary air flow

In Figure 8.20 we show widths, derived from the average NO fluorescence profiles of jet I and jet II, together with those of the flames, which are discussed later. The axial velocity profile of jet II contains the contribution of the annulus flow, making the results very sensitive to the definition of the width, so we will not include the width of the axial velocity profile of this jet in the discussion. The
jet with annulus flow, jet II, has a much smaller spread of the NO-seed profile, due to the annulus flow. Figure 8.20 shows that the NO profile width of jet II is (1.6 ± 0.1) times smaller than the width of jet I, for \( x = 150 \) to 250 mm. See Table 8.3 for the linearized relation between \( w_{\text{NO}} \) and \( x \).

### 8.6.3 Width of average profiles in flames

The widths of the average NO-seed profiles of flames I to VI are also shown in Figure 8.20.

The width \( w_{\text{NO}} \) of flame I is narrower than that of flame II, and that of flame II is narrower than that of flame III. An interesting fact is that in flame I, \( w_{\text{NO}} \) at \( x = 100 \) mm is only 10% larger than at \( x = 50 \) mm (with completely developed turbulence, increase of the axial distance with a factor of two would give a width twice as large). Apparently, there is only little entrainment for small \( x \) in flame I, which agrees with the fact that only few large scale eddies have been observed in the instantaneous images shown in Figure 8.4. To a lesser extent, this effect is also noticed in flame II, see Figure 8.20.

The less developed turbulence of flame I results in narrower NO profiles for the complete range of \( x \) covered; at \( x = 250 \) mm, the width of flame I is about 25% narrower than that of the other flames. For small \( x \), the width of flame II is slightly smaller than that of flames III–VI. The widths of flames III–VI are very close for the range of \( x \) covered, so the effect of larger primary air velocity or preheating is rather small.

Table 8.3: Linear approximation of the jet width \( w = \alpha(x + \beta d_j) \) of the axial velocity profiles of jet I, and of the NO profiles of jet I, jet II, and flames I–VI. The relation is determined for the range 50 < \( x < 250 \) mm, or 8.3 < \( x/d_j < 41.7 \) for flames III–VI and both jet I and II, and for more downstream locations of flames I and II.

<table>
<thead>
<tr>
<th>jet</th>
<th>range of ( x/d_j )</th>
<th>( w = \alpha(x + \beta d_j) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>min. 8.3, max. 41.7</td>
<td>( w_u )</td>
</tr>
<tr>
<td>I</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>II</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>flame</td>
<td></td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>I</td>
<td>33.3, 50.0</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>II</td>
<td>25.0, 50.0</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>III</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>IV</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
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<tr>
<td>V</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
</tr>
<tr>
<td>VI</td>
<td>8.3, 41.7</td>
<td>( w_{\text{NO}} )</td>
</tr>
</tbody>
</table>
The width of both jets and of flames III–VI depends almost linearly on the axial distance. The coefficients of the linear approximation of $u_{\text{NO}}$ vs. $x$ are given in Table 8.3. The axial location where the linear approximation predicts a width equal to zero is called the virtual origin of the jets for the range of $x$ covered. We find that in jet I, the virtual origin is at an axial location of about 2.5 jet diameters downstream the nozzle, in jet II this is at about 4 jet diameters upstream the nozzle, and in flames II–VI this is at 4 to 8 jet diameters upstream the nozzle. In flame I, the less developed jet width as function of $x$ becomes approximately linear for $x \geq 150$ mm ($x/d_j \geq 25$).

### 8.7 Standard deviation of NO seed in isothermal jets and flames

We now discuss the standard deviation profiles of the isothermal jets and flames, by comparing their shapes at different axial distances. Finally we discuss the standard deviation relative to the average signal.

#### 8.7.1 Standard deviation profiles in isothermal jets

For developed turbulence in a free isothermal jet, the standard deviation profiles of fuel concentration have off-axis maxima, see Becker et al.\[99\], Birch et al.\[101\] and Pitts and Kashiwagi\[102\]. (Pitts and Kashiwagi\[102\] measured concentration fluctuations in a methane jet issuing in air, $Re_j = 4130$, $d_j = 6.35$ mm.) The maximum value is reported to be 15 \%\[99\], 22 \%\[101\] and 31 \%\[102\] larger than the centerline value. Fluctuations in the fuel concentration are due to shear layer mixing, and the fuel concentration standard deviation is maximum near the center of the shear layer. Velocity measurements by Stroomer\[2\] show off-axis maxima differing only about 4 \% or less from the centerline value at $x = 250$ mm, in agreement with the results reported by Hinze\[91\], Wygnanski and Fiedler\[94\], and Sislian et al.\[12\].

For jet I, the standard deviation is shown in Figure 8.21. At $x = 50$ mm in both jets, the standard deviation is maximum at centerline, indicative of a small exit velocity fluctuation in the case of the isothermal jets, which were fuelled with bottled nitrogen. For larger $x$, the maximum is off-axis as expected for well developed jet turbulence. At $x = 200$ and 250 mm in jet I, the maximum value is 18 and 35 \% larger than the centerline value, respectively, and these values are comparable to those found by Birch et al.\[101\], Pitts and Kashiwagi\[102\] and by Becker et al.\[99\].

Like in jet I, the standard deviation in jet II is maximum off-axis for $x = 100$ mm and larger. At $x = 50$ mm, the standard deviation profile of jet II has steeper slopes, see Figure 8.22, like the profile of average NO at $x = 50$ mm. The centerline value at $x = 50$ is equal to that of jet I. For $x = 100$ mm and larger,
8.7. Standard deviation of NO seed

Figure 8.21: Standard deviation of NO fluorescence in jet 1, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (---), 100 (--), 150 (---), 200 (---) and 250 mm (-----); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.

Figure 8.22: Standard deviation of NO fluorescence in jet II, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (---), 100 (--), 150 (---), 200 (---) and 250 mm (-----); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.
Figure 8.23: Standard deviation of NO fluorescence in flame I, for scaling multiplied by x, as function of radial coordinate, for scaling divided by x, at x = 50 (−), 100 (− −), 150 (− − −), 200 (− − −) and 250 mm (− − −); this corresponds to x/dj = 8.3, 16.7, 25.0, 33.3 and 41.7, respectively.

the centerline standard deviations of jet II are about 30 % larger than those of jet I.

8.7.2 Standard deviation profiles in flames

Standard deviation profiles of the different flames are qualitatively different from the profiles of the isothermal jets, see Figures 8.23–8.28. The nonpreheated flames have off-axis maximum standard deviation at x = 50 mm, with small fluctuations at centerline.

At x = 100 and 150 mm in flame I and at x = 100 mm in flame II, the scaled standard deviation is much larger than at other axial distances. This is attributed to large-scale instabilities causing radial displacements of the fuel flow, observed in the instantaneous images (Figures 8.4 and 8.5). In flame II, the scaled standard deviation at x = 100 mm is significantly larger than at other axial locations.

In flame III (Figure 8.25), the scaled standard deviation at x = 100 is larger than at x = 50 mm and than for x ≥ 150 mm, but the effect is less pronounced than in flame I (Figure 8.23) and II (Figure 8.24), because in flame III the shear layer turbulence is more developed than in flames I and II. The difference between the maximum and the centerline value is only 11 % at x = 250 mm. We find that, except for x = 50 mm, the off-axis maximum standard deviation relative to the centerline value is smaller in flame III than in the isothermal jets.
8.7. Standard deviation of NO seed

Figure 8.24: Standard deviation of NO fluorescence in flame II, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (—), 100 (— —), 150 (— — —), 200 (— — —) and 250 mm (— — ——); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.

Figure 8.25: Standard deviation of NO fluorescence in flame III, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (—), 100 (— —), 150 (— — —), 200 (— — —) and 250 mm (— — ——); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.
Figure 8.26: Average NO signal (a) and standard deviation (b) at \( x = 250 \text{ mm} \) in jet II (---) and flame III (--). 

To compare the standard deviation of jet II with that of flame III, we show in Figure 8.26 the profiles of both the average NO signal and the standard deviation at \( x = 250 \text{ mm} \). We normalized the average profiles on the centerline value; for the scaled standard deviation this was not necessary, because the centerline values are almost equal. We find that for \( r/x \leq 0.05 \), the standard deviation is comparable, but that for larger \( r \), the standard deviation more quickly decreases in the flame. At \( x = 250 \text{ mm} \) the OH concentration is maximum at \( r = 13.6 \text{ mm} \), or \( r/x = 0.070 \); the reaction zone is most probably located around this value of \( r/x \). In this high temperature region of the flame, both the average and the standard deviation of the NO concentration becomes very small. However, the standard deviation relative to the average signal is larger in the flame in this region.

We have seen that the influence of the large primary air velocity on the average NO fluorescence was rather weak. We also concluded that the seed concentration must have been about 15 % larger in flame IV than in flame III. The standard deviations, however, differ by a larger amount, compare the standard deviation of flame III (Figure 8.25) with that of flame IV (Figure 8.27). In flame IV, the centerline standard deviations at \( x = 50, 150 \) and \( 250 \text{ mm} \), are 38 %, 21 % and 60 % larger than of flame III, respectively. So the influence of the primary air flow is larger on the fluctuations than on the average of the NO seed.

The scaled standard deviation of the preheated flame V is shown in Figure 8.28. Standard deviation profiles of both preheated flames are very similar in shape. The larger primary air velocity of flame VI results in centerline standard deviations which are about 25 %, 36 % and 25 % larger than in flame V, for \( x = 150, 200 \) and \( 250 \text{ mm} \), respectively. For \( x = 50 \) and \( 100 \text{ mm} \), the difference is smaller than 7 %, so the influence of the larger primary air velocity on the standard deviation is in the preheated flames only noticed downstream \( x = 150 \)
8.7. Standard deviation of NO seed

![Figure 8.27: Standard deviation of NO fluorescence in flame IV, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (---), 100 (--), 150 (— —), 200 (---) and 250 mm (----); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.](image)

![Figure 8.28: Standard deviation of NO fluorescence in flame V, for scaling multiplied by \( x \), as function of radial coordinate, for scaling divided by \( x \), at \( x = 50 \) (---), 100 (--), 150 (— —), 200 (---) and 250 mm (----); this corresponds to \( x/d_j = 8.3, 16.7, 25.0, 33.3 \) and 41.7, respectively.](image)
mm. Note that the standard deviations in the preheated flames are about equal to those in flames III and IV, whereas the average values are about 2.3 times smaller.

8.7.3 Relative centerline standard deviation

In the region with developed turbulence of an isothermal jet, the standard deviation of axial velocity is proportional to $x$, like the average velocity. Hence, the centerline axial velocity standard deviation, relative to the average axial velocity is constant and, according to Hinze[91] reaches a value of 0.289. Stroomer[2] found a value of 0.25 at $x = 250$ mm ($x/d_j = 41.7$).

Mass fraction fluctuations in a natural gas jet have been studied by Birch et al.[101]. For the relative centerline standard deviation of the methane mass fraction, they found a limiting value of 0.285. This limiting value for methane mass fraction standard deviation was only reached at $x/d_j = 70$, because the average value develops to similarity much more slowly than the standard deviation. The limiting value of the velocity standard deviation was reached at $x/d_j = 30$. Therefore, for all axial distances, the relative mass fraction standard deviation was smaller than the relative axial velocity standard deviation. Our measurements have been carried out for $x/d_j \leq 41.7$ in the case of the isothermal jets, and we found for jet I a relative centerline NO concentration standard deviation of $(0.222 \pm 0.017)$, almost constant in the range $x = 150$ to 250 mm. This value

![Figure 8.29: Relative centerline standard deviation of NO fluorescence, $\sqrt{\frac{\langle S_F^2 \rangle}{\langle S_F \rangle}}|_{cl}$, for jet I (---), jet II (---), flame I (---), II (---), III (---), IV (---), V (---) and VI (---).](image-url)
is in good agreement with the values reported by Birch et al.[101], Pitts and Kashiwagi[102], Dowling and Dimotakis[111] and Becker et al.[99]; at $x/d_j \approx 42$
Birch et al.[101] found a relative mass fraction standard deviation at centerline of 0.23, 81 % of the limiting value of 0.285; a similar value for the relative standard deviations of mass fraction was found by Pitts and Kashiwagi[102], 0.235 (0.216 for the mole fraction), also for a free isothermal methane jet, but the limiting value was not reached for $x/d_j \leq 30$; Dowling and Dimotakis[111] found a limiting value of 0.23, already reached at $x/d_j = 25$, for a ethylene jet issuing in nitrogen ($Re_j = 5 \times 10^3$, jet and ambient fluid densities equal, $d_j = 19$ mm); Becker et al.[99] found a relative standard deviation of 0.206 at $x/d_j = 32$, but still increasing with $x$.

In jet II, the value increased from 0.213 at $x = 100$ to 0.252 at $x = 250$
mm, so a limiting value is not reached, see Figure 8.29. The values are close
to those in jet I, see Figure 8.29. Absolute values of the axial velocity standard
devation of jet II are in the same order of magnitude as those in jet I, differing less
than about 35 %; the axial velocity, however, decreases much slower in jet II than
in jet I. As a result, the relative centerline standard deviation of axial velocity
in this jet is significantly smaller than in jet I: according to Stroomer[2] in the
range 0.12 to 0.19 for $x = 50$ mm to 250 mm.

At $x = 50$ mm, the relative standard deviation in flame III is slightly
smaller than in jet II (Figure 8.29). For larger $x$, the relative standard deviation
is larger (34 % at $x = 250$ mm), not only due to fluctuations, increased by
combustion, but also due to changing density and the effects of varying Boltzmann
factor and of quenching.

At $x = 50$ mm, the relative centerline standard deviation increases when
the exit velocities of the flames are increased, and when, in addition, the primary
air velocity is increased. The extremes are 0.10 in flame I and 0.25 in flame IV,
see Figure 8.29. For larger $x$ in flame I, the relative centerline standard deviation
increases, and reaches a maximum of 0.50 at $x = 150$ mm, and decreases slowly
for larger $x$. Qualitatively the same applies for flame II. At $x = 250$ mm, the
relative standard deviations of flames II and III are very close, and smaller than
that of flame I. The larger primary air velocity of flame IV gives a relative standard
devation which is larger than that of flame III, and the difference increases with
$x \geq 100$ mm. In the preheated flames, the value decreases from $x = 50$ mm until
a minimum value is reached at 100 mm (flame VI) or 150 mm (flame V). The low
average signals in the preheated flames, in combination with standard deviations
which are about equal to those in the nonpreheated flames, give large relative
standard deviations at $x = 250$ mm: 0.69 in flame VI compared to 0.50 in flame
V.

8.7.4 Radial dependence of relative standard deviation
We present the radial profiles of the relative standard deviation of NO for both
isothermal jets and flame III. The profiles of the other flames are similar in shape
Figure 8.30: Relative standard deviation of NO fluorescence in jet 1, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (---), 100 (--), 150 (---), 200 (---) and 250 mm (---); this corresponds to $x/d_j = 8.3$, 16.7, 25.0, 33.3 and 41.7, respectively.

Figure 8.31: Relative standard deviation of axial velocity in jet 1, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (---), 100 (--), 150 (---), 200 (---) and 250 mm (---); this corresponds to $x/d_j = 8.3$, 16.7, 25.0, 33.3 and 41.7, respectively.
8.7. Standard deviation of NO seed

Figure 8.32: Relative standard deviation of NO fluorescence in jet II, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (—), 100 (— —), 150 (— — —), 200 (— —) and 250 mm (— — —); this corresponds to $x/d_j = 8.3, 16.7, 25.0, 33.3$ and 41.7, respectively.

Figure 8.33: Relative standard deviation of NO fluorescence in flame III, as function of radial coordinate, for scaling divided by $x$, at $x = 50$ (—), 100 (— —), 150 (— — —), 200 (— —) and 250 mm (— — —); this corresponds to $x/d_j = 8.3, 16.7, 25.0, 33.3$ and 41.7, respectively.
and the main differences between the six flames are discussed in the previous section.

For $x \geq 100$ mm in jet I, the profiles with the radial distance scaled with $x$, map very well for not to large values of $r/x$. At centerline, the relative standard deviation is minimum, $(0.222 \pm 0.017)$, and it reaches a value of about 2 at $r/x = 0.2$ and $x = 250$ mm. This maximum value increases with $x$, see Figure 8.30. The radial profiles of relative standard deviation of NO-seed concentration are very close to those of relative standard deviation of axial velocity, obtained by Stroomer[2] and shown in Figure 8.31. Hence, relative fluctuations of NO-seed concentration and axial velocity respond in a similar way to the shear layer turbulence.

In jet II, the maximum values are smaller, about 1.0, and do not systematically increase with $x$, see Figure 8.32. For $x \geq 150$ mm, the profiles map rather well.

Close to the centerline the relative standard deviations are larger in flame III than in jet II. However, the maximum value is reached for smaller $r/x$, and also the maximum value is smaller. Consequently, for $r/x \geq 0.1$ the relative standard deviations in flame III become smaller than in jet II. At these radial locations the average signal already is very small, see also Figure 8.16.

8.8 Distribution functions of NO seed fluctuations

In this section, we discuss the distribution functions of the NO fluorescence signal fluctuations. The largest fluorescence signal can be derived from the distribution functions, and with this value we relate the average fluorescence signal in the single isothermal jet to the mass fraction. The influence of the primary air flow in the isothermal jets is discussed by comparing the results of jet I and jet II, the influence of combustion by comparing the results of jet II and flame III, and the influence of the exit Reynolds number by comparing the results of flame I and flame III, and of the influence of the primary air flow by comparing the flames III and IV.

8.8.1 Distribution functions for isothermal jets

In the previous sections, we found that in the isothermal jets, the average concentrations are larger at centerline in the jet with primary air flow, that this jet is narrower and that the standard deviations at centerline are almost equal.

The distribution function are shown both of jet I and jet II, at $x = 50$ mm (Figure 8.34), $x = 150$ mm (Figure 8.35) and at $x = 250$ mm (Figure 8.36). Three different radial locations are shown, at centerline, where the average signal is $2/3$ of the centerline value and where the average signal is $1/3$ of the centerline signal.
8.8. Distribution functions of NO seed fluctuations

Figure 8.34: Distribution functions of NO at $x = 50$ mm, in jet I (a), for $y = 0.0$ (upper), 3.6 (center) and 6.4 mm (lower), and jet II (b), for $y = 0.0$ (upper), 3.2 (center) and 5.9 mm (lower). The range of the abscissa is equal in all figures.

Figure 8.35: Distribution functions of NO at $x = 150$ mm, in jet I (a), for $y = -0.7$ (upper), 11.8 (center) and 20.1 mm (lower), and jet II (b), for $y = -1.3$ (upper), 7.9 (center) and 12.6 mm (lower). The range of the abscissa is equal in all figures.
Figure 8.36: Distribution functions of NO at \( x = 250 \) mm, in jet I (a), for \( y = 0.8 \) (upper), 21.7 (center) and 37.1 mm (lower), and jet II (b), for \( y = -3.2 \) (upper), 12.1 (center) and 20.6 mm (lower). The range of the abscissa is equal in all figures.

**Maximum instantaneous signal versus pure fuel**

We assume that in jet I the NO concentration at the burner exit is equal to the maximum instantaneous NO concentration at \( x = 50 \) \( (x/d_j = 8.3) \), given in Figure 8.34(a). In the case of jet I, the NO concentration, normalized with the exit NO concentration, represents the fuel mole fraction. For jet I, the mole fraction is almost equal to the mass fraction. We compare the centerline NO concentration at \( x = 150 \) mm, or \( x/d_j = 25 \), normalized with the largest NO concentration, with values for the fuel mass fraction at the same location reported in literature [8], [99]–[102].

Pitts[100] found that the average centerline mass fraction increased with Reynolds number due to a shift in virtual origin, and Pitts[8] found a decrease of average centerline mass fraction when fuel density is larger. According to Thring and Newby[9], this is because a jet issuing fluid with density \( \rho_i \) into coflowing fluid with density \( \rho_c \) develop with the axial distance normalized with an effective diameter, \( d_j^* = d_j / \sqrt{\rho_i / \rho_c} \). As the centerline mass fraction is inversely proportional to \( x/d_j^* \), we can map the values at the same distance \( x/d_j \) by multiplication of the mass fraction with \( \sqrt{\rho_c / \rho_i} \):

\[
\frac{Y_{cl}}{Y_0} \sqrt{\frac{\rho_c}{\rho_i}} \propto \frac{1}{x/d_j^*} \sqrt{\frac{\rho_c}{\rho_i}} = \frac{1}{x/d_j}. \tag{8.5}
\]

For \( x/d_j = 25 \), various reported values found for the average centerline mass fraction relative to the exit value of unity are given in Table 8.4, together with our average centerline NO concentration relative to the maximum instantaneous concentration at \( x = 50 \) mm. The value reported by Birch et al.[101] seems
Table 8.4: Reported values of average centerline fuel mass fraction at $x/d_j = 25$, relative to the exit mass fraction; the type of the jet, the ratio of jet and ambient fluid densities, $\rho_j$ and $\rho_c$, respectively, and the exit Reynolds numbers $Re_j$ are given. Also is given the relative centerline mass fraction, corrected for the effect of $\rho_j/\rho_c \neq 1$.

<table>
<thead>
<tr>
<th>reference</th>
<th>type</th>
<th>$\rho_j/\rho_c$</th>
<th>$Re_j$</th>
<th>$\frac{Y_{di}}{Y_0}$</th>
<th>$\frac{Re}{\rho_j} \frac{Y_{di}}{Y_0}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>[99]</td>
<td>air in air</td>
<td>1.00</td>
<td>54000</td>
<td>0.244</td>
<td>0.244</td>
</tr>
<tr>
<td>[101]</td>
<td>CH$_4$ in air</td>
<td>0.55</td>
<td>16000</td>
<td>0.256</td>
<td>0.345</td>
</tr>
<tr>
<td>[102]</td>
<td>CH$_4$ in air</td>
<td>0.55</td>
<td>4130</td>
<td>0.128</td>
<td>0.173</td>
</tr>
<tr>
<td>[8]</td>
<td>CH$_4$ in air</td>
<td>0.55</td>
<td>3950</td>
<td>0.135</td>
<td>0.182</td>
</tr>
<tr>
<td>[8]</td>
<td>C$_3$H$_8$ in CO$_2$</td>
<td>1.02</td>
<td>3950</td>
<td>0.173</td>
<td>0.171</td>
</tr>
<tr>
<td>[100]</td>
<td>C$_3$H$_8$ in air</td>
<td>1.55</td>
<td>3960</td>
<td>0.222</td>
<td>0.178</td>
</tr>
<tr>
<td>[100]</td>
<td>C$_3$H$_8$ in air</td>
<td>1.55</td>
<td>7930</td>
<td>0.238</td>
<td>0.191</td>
</tr>
<tr>
<td>[100]</td>
<td>C$_3$H$_8$ in air</td>
<td>1.55</td>
<td>11900</td>
<td>0.254</td>
<td>0.204</td>
</tr>
<tr>
<td>this study</td>
<td>N$_2$ in air</td>
<td>0.97</td>
<td>7700</td>
<td>0.156</td>
<td>0.158</td>
</tr>
</tbody>
</table>

to be too large, when compared to values of the other larger Reynolds number jets, presented by Pitts[100] and Becker et al.[99].

In our experiments of jet 1, the average centerline signal at $x = 150$ mm (or $x/d_j = 25$) is 1328 counts, and the maximum instantaneous signal at $x = 50$ mm is 8500 counts (Figure 8.34). The resulting estimation of the mass fraction at $x/d_j = 25$ is 0.156.

Comparing our result with that obtained in other jet with density ratio close to unity (C$_3$H$_8$ in CO$_2$[8]) we find that our mass fraction value is only 8 % smaller at $x/d_j = 25$; the agreement is slightly worse when the difference in Reynolds numbers is taken into account. We can also compare our result with that obtained in a jet with comparable exit Reynolds number ($Re_j = 7930[100]$), showing that our value is 17 % smaller, when the density ratio is taken into account. The difference is due to the fact that the maximum instantaneous signal at $x = 50$ mm ($x/d_j = 8.3$) cannot be determined very accurately from the distribution function, see also Figure 8.34(a). However, we find that at $x/d_j = 8.3$, the maximum concentration is close to the exit concentration.

**Shape of distribution functions**

The distribution functions of both isothermal jets are symmetric at centerline, and become skewed for larger radial distance. At centerline the shapes are approximately Gaussian. At the radial location where the average signal is 2/3 of the center line value (the function at center in the figures), the distribution functions are still fairly symmetric. Where the average signal is 1/3 of the centerline
value, the distribution functions have a positive skewness. At \( x = 50 \text{ mm} \) \( (x/d_j = 8.3) \), there is no indication of intermittency at this radial location, and the distribution functions can be described with a \( \beta \)-function. At \( x = 150 \text{ mm} \), a peak at zero signal due to intermittency is found at the radial location furthest from the centerline. For larger \( x \), this effect becomes more and more pronounced, but the shape of the distribution functions does not change much.

Dowling and Dimotakis[111] also present fuel concentration distribution functions in an ethylene jet issuing in nitrogen \( (Re_j = 5 \times 10^3, \rho_j/\rho_c = 1, d_j = 19 \text{ mm}) \), at centerline, at the radial location where the average concentration is 0.79 times the centerline value and where it is 0.43, for \( x/d_j = 20, 40, 60 \) and 80. For all \( x/d_j \), they found distribution functions with a slight negative skewness at centerline. At the second radial location the distribution functions were almost symmetric. At the third radial location, the distribution functions were bimodal, with a maximum near the average concentration and at zero concentration due to intermittency. Maximum values were twice the average concentration for the two radial locations closest to the axis and 2.5 times the average concentration for the largest radial distance. The distribution functions at the two radial locations closest to the axis are qualitatively equal to those of Figures 8.35 and 8.36. The third radial location of Dowling and Dimotakis[111] is closer to the axis than in Figures 8.35 and 8.36, and therefore in our results the maximum at finite concentration merged with the intermittency peak in our results.

Influence of primary air flow on the distribution functions for isothermal jets

We find that the shapes of the distribution functions are very similar for jet I and jet II. For larger \( x \), the standard deviation in jet II is somewhat larger, and consequently, the distribution functions are wider. For all \( x \), however, the maximum signal in jet II is larger than in jet I: only 7 \% larger at \( x = 50 \text{ mm} \) at centerline, where the jet widths are almost equal and the influence of the primary air flow is rather small, but 34 \% larger at \( x = 100 \text{ mm} \) where the influence of the primary air flow is first being noticed. The difference decreases with \( x \), for \( \geq 150 \text{ mm} \), and is 25 \% at \( x = 250 \text{ mm} \).

8.8.2 Influence of combustion on the distribution functions

We compare the distribution functions of NO fluorescence in flame III (Figures 8.37 to 8.39) with those in jet II (Figures 8.34(b) to 8.36(b)).

We find that the shape is significantly affected by combustion. At \( x = 50 \text{ mm} \), the average centerline value in flame III is 31 \% larger than in jet II, but the maximum instantaneous value is equal. At centerline of \( x = 50 \text{ mm} \), the distribution function in flame III is very skewed (with a negative skewness), see Figure 8.37. The most probable signal in flame III (10 \% larger than the average signal) is 44 \% larger than the most probable signal (= average signal) in jet II at centerline.
In the distribution functions, the tail extending towards small concentrations, is due to combustion products entrained into the fuel jet occasionally. In jet II, entrained air reaches the centerline much more frequently. Entrained hot fluid decreases the density and thus the NO concentration more than entrained cold fluid. An additional effect can play a role: the quenching of excited NO, which is strongest near the reaction zone. This quenching gives an extra decrease of signal. The distribution function shows that the hot products are found not very frequently at the centerline, for the probability of large signals is much larger. The standard deviation relative to the average signal is only 7 % smaller in the flame than in the isothermal jet, see Figure 8.29, so the influence of combustion on the standard deviation is in that case not very large. The largest NO concentrations are found in the cold fluid, and the maximum signal is equal in both jet II and flame III.

The temperature at centerline in flame III is only 20 K above room temperature, so the effect on the distribution function of larger signals because of larger Boltzmann factor is small.

It should be noted that the influence of entrainment of hot combustion products on the mass fraction distribution functions is different from the process described above: the fuel (or fuel tracer) mass fraction is affected less by entrainment of hot, and therefore light, fluid than by entrainment of cold and heavy fluid, see Schefer et al.[103], whereas the concentration is affected more by hot fluid decreasing the density.

At the second radial location at $x = 50$ mm, the small signals forming the tail in the centerline distribution function, become most probable, probably due to the decreased density. This gives a distribution function with positive skewness, whereas the distribution function in jet II at the same radial location was still symmetric.

At $x = 150$ mm, the distribution functions of jet II and flame III become similar in shape. The effects of larger signal due to Boltzmann factor increasing with temperature and of smaller signal due to smaller density apparently compensate, giving similar maximum signals at $x = 150$ mm.

At $x = 250$ mm, the signals in the flame are significantly smaller than in jet II, and the distribution functions at the second and third radial location are more skewed in flame III than in jet II, compare Figures 8.36(b) and 8.39.

### 8.8.3 Influence of exit Reynolds numbers on the distribution functions

We compare the distribution functions of NO fluctuations of flames I and III. The distribution functions of flame II have a shape between those of flame I and III and are not discussed separately. At $x = 50$ mm at centerline we find that in the distribution function the tail extending towards small signals in flame III (Figure 8.37) is not present in flame I, but that the most probable signal and the maximum signal are equal within the accuracy of the measurements (of
Chapter 8. Visualization and fluctuations of NO seed

Figure 8.37: Distribution functions of NO at $x = 50$ mm, in flame III, for $y = 0.5$ (upper), 2.3 (center) and 3.9 mm (lower). The range of the abscissa is equal in all figures.

Figure 8.38: Distribution functions of NO at $x = 150$ mm, in flame III, for $y = 1.6$ (upper), 5.5 (center) and 9.3 mm (lower). The range of the abscissa is equal in all figures.

flame III these values are both about 3 % larger). We therefore can conclude that the only difference at $x = 50$ mm at centerline is the larger probability of small signals, or in other words, that in flame I at $x = 50$ mm ($x/d_j = 8.3$) hot combustion products or air never reach the centerline, whereas in flame III they occasionally do. In flame II at the same location, there is some probability for signals less than, say, 6000 counts; as expected, this probability is larger than in flame I and smaller than in flame III.

At the radial locations where the average signal equals $2/3$ and $1/3$ of the centerline value, at $x = 50$ mm, the distribution functions of flames I, II and
8.8. Distribution functions of NO seed fluctuations

Figure 8.39: Distribution functions of NO at \( x = 250 \) mm, in flame III, for \( y = -0.9 \) (upper), 8.9 (center) and 15.1 mm (lower). The range of the abscissa is equal in all figures.

Figure 8.40: Distribution functions of NO at \( x = 50 \) mm, in flame I, for \( y = 0.4 \) (upper), 2.1 (center) and 2.9 mm (lower). The range of the abscissa is equal in all figures.
III are equal, with respect to the most probable signal, the maximum signal and the width, compare Figures 8.40 and 8.37.

The maximum signal in flame I decreases slower than in flame III, as can be seen by comparing the distribution functions $x = 150$, compare Figure 8.41 for flame I with Figure 8.38 for flame III. At $x = 50$ mm, the maximum signal of flame I is equal to that of flame III, at $x = 100$ mm it is 7 % larger, at $x = 150$ mm 100 %, at $x = 200$ mm 58 % and at $x = 250$ mm 21 %. The maximum signal in flame II differs at all axial locations less than 5 % from those in flame III, except for $x = 150$ mm, where the maximum signal in flame II is 14 % larger. Thus we find that the difference in the exit velocities of flames I to III has the largest effect on the maximum instantaneous signal at $x = 150$ mm; for smaller $x$, the maximum signals are comparable in these flames, for larger $x$ the maximum signal in flame I decreases slower than in flames II and III. This is in agreement with the observation that the fuel jet in flame I develops with $x$ less quickly than in the other flames with larger exit velocities.

The difference in distributions in flame I and III is largest at $x = 150$ mm. At $x = 250$ mm at centerline, the larger primary air velocity of flame IV gives a larger small-signal probability, compared to flame III. This indicates that combustion products or air reach the centerline more often when the primary air velocity is larger. The distribution functions of flame IV at $x = 50$ mm are shown in Figure 8.43. As was already observed in the previous section, the NO seeding concentration in flame IV was about 15 % larger, giving a distribution function similar in shape but extended towards larger signals. For larger $x$, the
8.8. Distribution functions of NO seed fluctuations

Figure 8.42: Distribution functions of NO at $x = 250$ mm, in flame I, for $y = 0.0$ (upper), 3.6 (center) and 7.3 mm (lower). The range of the abscissa is equal in all figures.

Figure 8.43: Distribution functions of NO at $x = 50$ mm, in flame IV, for $y = 0.4$ (upper), 2.2 (center) and 3.8 mm (lower). The range of the abscissa is equal in all figures.

difference in shape of the distribution functions between flames III and IV is rather small.

Preheating decreases the average concentrations (Figure 8.18), but gives rather large relative standard deviations (Figure 8.29); preheating, however, does not affect the shape of the distribution functions. The shape of the distribution functions of NO fluctuations in the preheated flames V and VI is comparable to those of the nonpreheated flames III and IV.
8.8.4 Joint distribution of concentration and gradient

We investigate the joint distribution of the NO signal and its radial gradient, as has already been done for the case of OH in Section 7.10. NO as fuel marker manifests completely differently from OH, which is found only in narrow regions for the range of $x$ covered by our measurements. We found a strong correlation between the OH concentration and its gradient.

Namazian et al.\textsuperscript{[104]} investigated the correlation between the fuel mass fraction and its dissipation in a methane jet issuing in air ($Re_j = 7 \times 10^3$, $d_j = 5.4$ mm) by means of two dimensional imaging of Raman scattering. The dissipation was defined as the scalar diffusion coefficient times the sum of the average squared axial gradient and twice the average squared radial gradient; it was assumed that the azimuthal and the radial components are equal. They found that the maximum average scalar dissipation is off-axis for $x/d_j < 15$; that in this region, off-axis the radial component of the dissipation is larger than the axial component (equal at centerline); that the dissipation is log-normally distributed; that there is no (or a weakly negative) correlation between the mass fraction and the dissipation at centerline, but that there is a rather strong positive correlation in the outer region of the shear layer ($y$ approximately equal to the jet width). The motivation for this is that "the scalar sharply increases at the boundary of the jet. At these boundaries, both the level and the gradient of the scalar are high, resulting in a finite degree of correlation between the two. In the central part of the jet, scalar levels are also high but the gradients are random and smaller than those found at the boundaries"\textsuperscript{[104]}. Many PDF models used to model turbulent diffusion flames, assume independence between scalar and gradient. As pointed out in Section 7.10, small scale processes are better described when the instantaneous gradients or scalar dissipation are included in the joint PDF.

In our NO measurements, the NO follows the fuel flow, and the joint distribution of signal and squared gradient serves as an indication of the relation between the mixture fraction and the scalar dissipation. We present the results of joint distribution of the NO signal and the squared radial gradient of jet II and flame III, at centerline and close to $y = u_{NO}/2$, for $x = 50$ mm and $150$ mm. Joint distributions of jet I are close to those of jet II, and are not discussed separately. In addition, we calculated the cross-correlation coefficient of $[S_F - \langle S_F \rangle]$ and $[(\partial S_F/\partial y)^2 - \langle (\partial S_F/\partial y)^2 \rangle]$, following Namazian et al.\textsuperscript{[104]}. The accuracy of the correlation coefficients is 0.05.

In Figure 8.44 the joint distribution of jet II at $x = 50$ mm is shown. Note that the squared gradient is set to a logarithmic scale. We find that at centerline the concentration and the squared gradient are not completely independent; the correlation coefficient was found to be 0.19. At $y = 5.6$ mm $\approx u_{NO}/2$ (Figure 8.44(b)), the relation between the concentration and squared gradient is more significant, and consequently, the correlation coefficient is somewhat larger, 0.24. Further off-axis, the correlation becomes larger, with a maximum of nearly 0.5 at
8.8. Distribution functions of NO seed fluctuations

Figure 8.44: Joint distribution functions of NO signal and the squared gradient, at $x = 50$ mm in jet II, for $y = 0.0$ (a) and 5.6 mm (b); $w_{NO}/2 = 4.5$ mm. Contours are at 5 (outer contour), 15, ... 55 out of 5000 measurements.

Figure 8.45: Joint distribution functions of NO signal and the squared gradient, at $x = 150$ mm in jet II, for $y = 1.0$ (a) and 11.0 mm (b); $w_{NO}/2 = 10.1$ mm. Contours are at 5 (outer contour), 15, ... 45 out of 5000 measurements.
Figure 8.46: Joint distribution functions of NO signal and the squared gradient, at \( x = 50 \) mm in flame III, for \( y = 0.0 \) (a) and 3.1 mm (b); \( w_{\text{NO}}/2 = 3.1 \) mm. Contours are at 5 (outer contour), 15, \ldots 45 out of 5000 measurements.

\( y = 10 \) mm \((r/x = 0.2)\). Figure 8.44(b) shows that the gradients remain rather large, even when the average signal is much smaller further from the axis. Close to the axis, the distribution function of the squared gradient, is not completely log-normal in shape, because when plotted logarithmically it is not completely symmetric (negative skewness).

The radial profiles of the cross-correlation coefficient are similar to those reported by Namazian et al.[104], although at \( x/d_j = 9 \) the centerline correlation of Namazian et al. is close to zero, whereas it is about 0.2 in our experiments at \( x/d_j = 8.3 \). This may be due to the use of gradients in two directions to derive the dissipation by Namazian et al., whereas we only used the radial gradient. For larger \( y \), the correlation coefficients of Namazian et al. increases, reaching a maximum value of 0.54 at about \( y \) about 1.6 times the jet width for \( x/d_j = 9 \). This maximum value is comparable to the maximum value obtained from our experiments.

At \( x = 150 \) mm the centerline correlation between the concentration and the squared gradient (Figure 8.45(a)) is smaller than at \( x = 50 \) mm, with a correlation coefficient of 0.04. At \( y = w_{\text{NO}}/2 = 10.1 \) mm, the correlation coefficient is 0.20.

At \( x = 50 \) mm in flame III the situation at centerline is different from that in both isothermal jets. There is negative correlation between signal and gradient (correlation coefficient \(-0.25\)), indicating that at centerline the gradients are smaller when the signal is larger; reported correlation coefficients of Namazian et al.[104] for a isothermal jet are never more negative than \(-0.04\) (a value found at
8.9. Length scales of NO seed fluctuations in isothermal jets and flames

Figure 8.47: Joint distribution functions of NO signal and the squared gradient, at \( x = 150 \) mm in flame III, for \( y = 0.0 \) (a) and \( 8.4 \) mm (b); \( w_{NO}/2 = 7.8 \) mm. Contours are at 5 (outer contour), 15, ... 55 out of 5000 measurements.

\( x/d_j = 5 \). In addition, in flame III the maximum correlation is larger (correlation coefficient 0.60 at \( y = 6.5 \) mm) than in both isothermal jets. Figure 8.46(b) shows that at \( y = 3.1 \) mm, the correlation is positive (correlation coefficient 0.29), which is larger than in jet II at the corresponding radial location. At \( x = 150 \) mm at centerline, there is a correlation of 0.20, larger than in the isothermal jets at \( x = 150 \) mm; at \( y = 8.4 \) mm, close to \( y = w_{NO}/2 \), there is a positive correlation of 0.29, again more pronounced than in jet II.

According to Figure 7.1, the reaction zone is at about \( y = 13 \) mm at \( x = 150 \) mm. At this radial location, we found significant positive correlation (in the order of magnitude of 0.3) between the concentration and the gradient at these radial locations, both in the isothermal jets and in flame III. Therefore, at the location of the reaction zone in a flame, there is a positive correlation between mixture fraction and scalar dissipation, also found by Namazian et al.[104].

8.9. Length scales of NO seed fluctuations in isothermal jets and flames

Length scales of velocity fluctuations represent the size of characteristic eddies in the turbulent flow. When derived from spatial correlation functions, the scales obtained are the Taylor micro scale and the integral scale of turbulence. Length scales of passive scalars give the size of the small and large eddies that transport this scalar. In the burner, the seed becomes homogeneously distributed, and the seed concentration does not reveal turbulent eddies in this flow. Hence, the length
scales of turbulent seed concentration fluctuations are due to mixing with flow
that does not contain seed, i.e. due to mixing by shear layer turbulence.

Length scales of OH fluctuations are discussed in Section 7.11. Definitions
of correlation coefficients and of length scales can be found in the same section.
In our measurements of NO fluctuations in isothermal jets, and to a lesser extent
in flames, the NO fluorescence to a good approximation represents a passive
scalar. The length scales derived from the measurements are lateral length scales.
The results are compared with the lateral isothermal jet length scale results
of Wygnanski and Fiedler[94] and the lateral heated jet length scale results of
Corrsin and Uheroi[95].

Wygnanski and Fiedler[94] measured length scales of turbulence in a
jet of air, issuing in still air $(Re_j = 10^5, d_j = 26.4$ mm, $x/d_j = 90$); Corrsin
and Uheroi[95] measured length scales of turbulent velocity fluctuations and of
temperature fluctuations in a heated jet $(T_j = 473$ K, $Re_j = 7.5 \times 10^4, d_j =
25.4$ mm, $x/d_j = 20$). Integral length scales $L_u/x = 1.57 \times 10^{-2}$ at centerline and
$3.04 \times 10^{-2}$ at $y/x = 0.15$ have been reported by Wygnanski and Fiedler[94];
an integral length scale $L_u/x = 1.32 \times 10^{-2}$ at centerline has been reported by
Corrsin and Uheroi[95]. For temperature fluctuations, Corrsin and Uheroi[95]
have reported $L_T/x = 1.52 \times 10^{-2}$ at centerline. The difference between $L_T$ and
$L_u$ was smaller than expected, regarding the difference in width of the average
velocity and temperature profiles. This was due to significant negative correlation,
decreasing the value of $L_T[95]$. The results of Corrsin and Uheroi are found at a
value of $x$ much closer to the nozzle than those of Wygnanski and Fiedler, and
self-similarity for the scales of Corrsin and Uheroi is not yet expected. The lateral
Taylor micro scales $\lambda_u/x$ found by Wygnanski and Fiedler at $x/d_j = 90$, range
from $3.2 \times 10^{-3}$ at centerline to $1.9 \times 10^{-3}$ at $y/x = 0.15$. At centerline, Corrsin
and Uheroi found $\lambda_u/x = 5.5 \times 10^{-3}$ and $\lambda_T/x = 8.5 \times 10^{-3}$.

Birch et al.[101] give results of the axial dependence of integral time
scales of fuel mass fraction fluctuations, obtained from an isothermal methane
jet, issuing in still air. Using Taylor's hypothesis, the time scales are converted to
longitudinal length scales. The axial centerline integral length scale of fuel mass
fraction fluctuations, divided by $x$ was found to be $4.3 \times 10^{-2}$. This longitudinal
value is $13\%$ larger than the scaled longitudinal integral length scale of turbulence
found by Wygnanski and Fiedler.

Integral length scales of fuel concentration, both in an isothermal methane
jet and in a lifted flame are presented by Schefer et al.[103], using a definition of
this scale as the separation between points that have a correlation coefficient of
0.5. It should be noted that the methane is not a conserved scalar in the flame.
Their measurements have been carried out at axial locations up to $18.5$ jet diam-
eters, where the centerline fuel is not yet consumed by combustion. They found
that the characteristic scales increase with downstream distance; that they are
larger in the mixing region than in the central jet; that in the mixing region the
longitudinal length scales are somewhat larger than the lateral length scales; that
due to combustion the length scales become noticeably larger (in radial direction roughly with a factor of 2.5 at centerline). The latter effect is explained by the "expansion of the eddies due to the higher temperatures in the flame" [103]. For this, the integral time scales should increase with a relatively smaller amount than the integral length scales.

In our measurements where NO is seeded to the fuel in flames, the NO is not conserved, like the methane concentration in the lifted flame presented by Schefer et al. [103]. Quenching in the region of the reaction zone and in the region of fresh air, decreases the NO fluorescence signal, thus decreasing the length of correlation. Despite this expected decrease of correlation length, we show that the results show the same trend as presented by Schefer et al. [103].

We investigate the influence of the primary air flow on the length scales and the influence of combustion, and we discuss the length scales in the different flames. We conclude with a comparison of the NO-seed length scales and the OH length scales.

8.9.1 Length scales in isothermal jets, influence of primary air flow

Integral scales

In an isothermal single jet, complete self-similarity was achieved by Wygnanski and Fiedler [94] for \( x/d_j > 50 \); their measurements were carried out at \( x/d_j = 90 \). This implies that in our measurements the jet flow is expected to be not self-similar at the most downstream station (\( x = 250 \) mm, or \( x/d_j = 41.7 \)), so the integral length scales obtained are expected not to scale with \( x \).

At centerline of jet I, the integral length scale of NO fluctuations, \( L_{NO} \), is 3.8 and 5.9 mm at \( x = 200 \) and \( 250 \) mm, respectively. This corresponds to \( L_{NO}/x = 1.9 \times 10^{-2} \) at \( x/d_j = 33.3 \) and \( 2.4 \times 10^{-2} \) at \( x/d_j = 41.7 \). We see that the scaled integral length scale increases with \( x \). Self-similarity is therefore not fully achieved for this range of \( x \).

At centerline of \( x = 250 \) mm (\( x/d_j = 41.7 \)), \( L_{NO}/x = 2.4 \times 10^{-2} \) and \( 1.2 \times 10^{-2} \), for jet I and jet II, respectively. (Note that \( L/x = \alpha \) corresponds to \( L/d_j = \alpha \times x/d_j \). Hence, \( L/x \) is to be compared for equal \( x/d_j \), except when \( L/x \) is constant.) Following Hinze [91], we multiply the centerline value of \( L_{NO}/x \) with a turbulent Schmidt number of 0.7, to compare \( L_{NO}/x \) with \( L_u/x \), and yield 1.7 and \( 0.8 \times 10^{-2} \) for jet I and jet II, respectively, at \( x/d_j = 41.7 \). The value obtained in jet I agrees well with the value for \( L_u \) found by Wygnanski and Fiedler for \( x/d_j = 90 \) (\( L_u/x = 1.57 \times 10^{-2} \)), and with those found by Corrsin and Uberoi for \( x/d_j = 20 \) (\( L_u/x = 1.32 \times 10^{-2} \)). In jet II, the (corrected) scaled centerline integral length scale of NO fluctuations is smaller than the reported values. This is due to a reduction of the integral length scales by the the primary air flow.

The integral length scale increases with radial distance, which was also observed by Wygnanski and Fiedler. The radial profiles of the integral length scale of both jet I and jet II are given in Figure 8.48, for \( x = 250 \) mm.
Figure 8.48: Integral length scales $L_{NO}$ of NO fluctuations at $x = 250$ mm ($x/d_j = 41.7$) in jet I (---), jet II (- -) and flame III (— —).

It is clear that the primary air flow significantly reduces the integral length scales of NO fluctuations, with a factor 1.5 and 2.0 at $x = 200$ and 250 mm, respectively. The turbulent primary air flow reduces the formation of large eddies in the shear layer, which also reduces the jet width with a comparable amount: jet II is 1.7 and 1.6 times narrower than jet I at $x = 200$ and 250 mm, respectively. The ratio $L_{NO}/w_{NO}$ is approximately constant for both isothermal jets, with a value of $(0.10 \pm 0.02)$.

Taylor micro scales
The difference between the Taylor micro scales of NO in jet I and jet II is relatively much smaller than the difference between the integral length scales in both jets: at $x = 50$ mm, the Taylor micro scales near the jet axis are only 10% larger in jet I than in jet II, at $x = 250$ mm they are 15% smaller. For $x = 100, 150$ and 200 mm, the differences are less than 3%, which is not significant. The accuracy of the Taylor micro scales is estimated from the scatter of the measurements in the radial profiles, yielding an accuracy of 4%. Taylor micro scales in jet I and jet II are shown in Table 8.6. Off-axis, the differences become more significant, see Figure 8.49 for $x = 250$ mm. In jet I, the Taylor micro scale increases with the radial distance, where the NO containing fluid from the central jet mixes with still air. In jet II, however, the Taylor micro scales show little radial dependence. This is because in jet II the mixing of NO is with a turbulent air flow. In the outer parts of the jet, Taylor micro scales are therefore smaller in jet II than in jet I.
8.9. Length scales of NO seed

Figure 8.49: Taylor micro scales $\lambda_{NO}$ of NO fluctuations at $x = 250$ mm ($x/d_j = 41.7$) in jet I (—), jet II (— —) and flame III (— — —).

At $x = 250$ mm, the centerline Taylor micro scales of NO in jet I and jet II, scaled with $x$, are $5.4 \times 10^{-3}$ and $6.3 \times 10^{-3}$. Multiplied with the turbulent Schmidt number of 0.7, this yields $3.8 \times 10^{-3}$ and $4.4 \times 10^{-3}$, for jet I and jet II, respectively. For $x/d_j = 90$, Wygnanski and Fiedler[94] found $\lambda_u/x = 3.7 \times 10^{-3}$, so the agreement of our centerline Taylor micro scale for jet I, scaled with $x$, with the value of Wygnanski and Fiedler is very good. For $x/d_j = 20$, Corrsin and Uberoi[95] found $\lambda_u/x = 5.5 \times 10^{-3}$ and $\lambda_T/x = 8.5 \times 10^{-3}$. At $x/d_j = 25$, comparable to the axial location used by Corrsin and Uberoi, the scaled centerline values of the Taylor scale of NO fluctuations are $1.1 \times 10^{-2}$ and $1.0 \times 10^{-2}$, respectively, which corresponds rather well with the value for $\lambda_T/x$ found by Corrsin and Uberoi.

The ratio of the integral length scale and the Taylor micro scales, both at centerline, is in the order of magnitude of 4, for jet I. Wygnanski and Fiedler[94] found a value of 4.2 at $x/d_j = 90$. Antonia and Bilger[112] also found a ratio of 4, for $x/d_j = 265$, in a jet with a much larger exit Reynolds number, $Re_j = 3.6 \times 10^4$. Our ratio for jet I at $x/d_j = 41.7$ agrees well with those for developed turbulent jets at much larger $x/d_j[94,112]$.

8.9.2 Influence of combustion on the length scales

Integral scales

We compare the integral length scale results and the Taylor micro scale results of flame III with those of jet II. The integral length scales in the flame are larger
than in the isothermal jet with primary air flow. For $x = 250$ mm, this is shown in Figure 8.48.

At centerline, the integral scales in flame III are 1.9, 3.0 and 2.1 times larger than in jet II, at $x = 150$, 200 and 250 mm, respectively. (For smaller $x$, the integral length scales have not been determined, because of convergence problems.) These factors agree with the factor 2.5 found by Schefter et al.[103]. Sisilian et al.[12] give integral scales of velocity fluctuations, both in an isothermal jet and in a turbulent nonpremixed flame. At $x/d_j = 50$, they found that the thermal expansion by combustion increases the average axial velocity with a factor of 4.2, whereas it decreases the integral time scale with a factor of only 1.6. As a result, the integral length scales increase with a factor 2.6.

The integral length scales in a flame are larger than in an isothermal jet (Figure 8.48), whereas the jet width is narrower (Figure 8.20). Hence, the relation between the integral length scale and the jet width is not constant under both isothermal and combusting conditions.

**Taylor micro scales**

The Taylor micro scales are also larger in the flame, except for $x = 50$ mm, where the centerline Taylor scales in the flame are almost equal. At $x = 100$ mm, the difference is still rather small, 4 %, but for $x = 150$ mm and larger, the difference becomes 32 %, 50 % and 33 %, for $x = 150$, 200 and 250 mm, respectively. Approximately the same increase is found off-axis. The differences at $x = 50$ and 100 mm are not significant, so we find that close to the nozzle the centerline Taylor micro scales are not significantly affected by combustion. We have also found that at centerline at $x = 50$ mm, the NO fluctuation distribution function in flame III differed from that in jet II at the same location, indicating a smaller entrainment in the flame. Hence, the smaller entrainment does not influence the small scale turbulent structure in the central part of the, flow close to the nozzle ($x \leq 100$ mm).

For $x = 250$ mm, the Taylor micro scales of both jet II and flame III are shown in Figure 8.49. There is a local minimum at centerline, both for the jet and for the flame. For increasing radial distance, the Taylor scales in the flame reach a maximum value. For larger $y (y \geq w_{NO}/2)$ the Taylor micro scales quickly decrease. This is observed in all flames at all axial locations. At the same radial location, the relative difference in standard deviation between jet II and flame III is much larger than the relative difference in radial gradient (both quantities are smaller in the flame), see also relation (7.3). It was already observed in Section 8.7 that the profiles of the standard deviation in flame III are significantly narrower than in jet II.
8.9. Length scales of NO seed

Table 8.5: Centerline values of the integral length scales of NO, $L_{NO}$, for flames I to VI, and for jet I and jet II, for $x = 200$, 250 and, when measured, 300 mm. The locations where the integration did not converge well, the obtained value is indicated with *, and the accuracy of these values is about 30 %, whereas the accuracy of the other measurements is about 10 %.

<table>
<thead>
<tr>
<th>flame</th>
<th>$x = 200$ mm</th>
<th>$x = 250$ mm</th>
<th>$x = 300$ mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>4.8*</td>
<td>4.3</td>
<td>4.0</td>
</tr>
<tr>
<td>II</td>
<td>3.5*</td>
<td>4.8</td>
<td>4.6</td>
</tr>
<tr>
<td>III</td>
<td>5.9</td>
<td>6.2</td>
<td>6.1</td>
</tr>
<tr>
<td>IV</td>
<td>3.8</td>
<td>3.1</td>
<td>3.0</td>
</tr>
<tr>
<td>V</td>
<td>3.4</td>
<td>4.2</td>
<td>−</td>
</tr>
<tr>
<td>VI</td>
<td>3.5*</td>
<td>2.8</td>
<td>−</td>
</tr>
<tr>
<td>jet</td>
<td>3.8*</td>
<td>5.9</td>
<td>−</td>
</tr>
<tr>
<td>II</td>
<td>2.6</td>
<td>2.9</td>
<td>−</td>
</tr>
</tbody>
</table>

8.9.3 Length scales in different flames

Integral scales

The integral length scales of the different flames show some interesting aspects. The shape of the integral length scale profiles are similar for all flames and for all $x$ measured, and an example is found in Figure 8.48. The values are given in Table 8.5, together with the values obtained in both isothermal jets. At some locations, the integration did not converge.

With increasing exit velocity, the centerline integral length scale increases: at $x = 250$ mm, this scale is 4.3, 4.8 and 6.2 mm, for flames I, II and III, respectively. We find largest integral length scales in the flame which is widest, yielding a relation $L_{NO}/w_{NO} = (0.27 ± 0.03)$.

We have seen that the primary air flow results in a decrease of the centerline integral length scale in the isothermal jets. A similar trend is seen in the flames, when the primary air velocity is increased: compare flames III and IV, and V and VI, respectively. At $x = 250$ mm, the centerline integral length scale is 6.2 and 3.1 mm in flames III and IV, respectively, and 4.2 and 2.8 mm in flames V and VI, respectively: a decrease due to larger primary air velocity of 50 % and 33 %. We see that the effect of the large primary air flow is smaller in the preheated flames.

Preheating decreases the integral length scales: when we compare flames III and V, and IV and VI respectively, using the values given above, we find that at $x = 250$ mm the integral scale in flame V is 32 % smaller than in flame III, and
Table 8.6: Centerline values of the Taylor micro scales of NO, \( \lambda_{NO} \), for flames I to VI, and for jet I and jet II, for \( x = 100 \) to 250 and, when measured, 300 mm. The accuracy is estimated as 4 %.

<table>
<thead>
<tr>
<th>flame</th>
<th>( x = 100 ) mm</th>
<th>( x = 150 ) mm</th>
<th>( x = 200 ) mm</th>
<th>( x = 250 ) mm</th>
<th>( x = 300 ) mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>1.39 ± 0.06</td>
<td>1.84 ± 0.07</td>
<td>2.31 ± 0.09</td>
<td>2.30 ± 0.09</td>
<td>2.20 ± 0.09</td>
</tr>
<tr>
<td>II</td>
<td>1.45 ± 0.06</td>
<td>2.00 ± 0.08</td>
<td>2.26 ± 0.09</td>
<td>2.03 ± 0.08</td>
<td>1.90 ± 0.08</td>
</tr>
<tr>
<td>III</td>
<td>1.54 ± 0.06</td>
<td>1.97 ± 0.08</td>
<td>2.27 ± 0.09</td>
<td>2.10 ± 0.08</td>
<td>2.05 ± 0.09</td>
</tr>
<tr>
<td>IV</td>
<td>1.52 ± 0.06</td>
<td>1.99 ± 0.08</td>
<td>2.35 ± 0.09</td>
<td>2.52 ± 0.09</td>
<td>2.77 ± 0.11</td>
</tr>
<tr>
<td>V</td>
<td>1.73 ± 0.07</td>
<td>2.02 ± 0.08</td>
<td>2.04 ± 0.08</td>
<td>2.02 ± 0.08</td>
<td>-</td>
</tr>
<tr>
<td>VI</td>
<td>1.80 ± 0.07</td>
<td>2.20 ± 0.09</td>
<td>2.43 ± 0.10</td>
<td>2.27 ± 0.09</td>
<td>-</td>
</tr>
<tr>
<td>jet</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td>1.52 ± 0.06</td>
<td>1.60 ± 0.06</td>
<td>1.50 ± 0.06</td>
<td>1.35 ± 0.05</td>
<td>-</td>
</tr>
<tr>
<td>II</td>
<td>1.49 ± 0.06</td>
<td>1.55 ± 0.06</td>
<td>1.53 ± 0.06</td>
<td>1.58 ± 0.06</td>
<td>-</td>
</tr>
</tbody>
</table>

that the integral scale in flame VI is 10 % smaller than in flame IV. Apparently, the large primary air flow already gives such a strong decrease of the integral length scale, that preheating does not have a very large influence.

**Taylor micro scales**

Again, the profiles of the Taylor micro scales in the different flames and at different \( x \) are similar in shape, and an example is shown in Figure 8.49.

In all flames, the centerline Taylor micro scale increases with \( x \) up to a certain axial distance: in flames I, II, III, V and VI, the centerline Taylor micro scale has a maximum at \( x = 200 \) mm; in flame IV, the scale increases with \( x \) until the last station at 300 mm. The values for \( x \geq 150 \) mm are given in Table 8.6.

As can be seen in Table 8.6, the differences between the Taylor micro scales in flames I to III are rather small, for every \( x \); the differences are smaller than 15 %, and in many combinations not significant. We have seen in Section 8.9.2 that for \( x \geq 150 \) mm the Taylor micro scales near the axis are larger in flame III than in the isothermal jet II, with a difference significantly larger than between jet I and jet II, see Figure 8.49. At \( x = 200 \) mm, the centerline Taylor micro scales of flames I–III differ less than 2 %, whereas they are about 52 % larger than the Taylor micro scales of jet I and jet II. For this region of \( x \), apparently, the combustion has an increasing effect on the Taylor micro scales, which is significantly larger than the influence of the exit velocities in flames I, II and III. The larger primary air velocity of flame IV gives centerline Taylor micro scales that are slightly larger compared to flame III, and the difference increases with \( x \), up to 40 % at \( x = 300 \) mm. Preheating gives somewhat larger scales at \( x = 50 \) and 100 mm. The Taylor
scales of flame v do not show an increase with \( x \geq 100 \text{ mm} \). Also in the preheated flames, the larger primary air velocity of flame vi results in Taylor micro scales that are about 10% larger than in the other preheated flame flame v.

Generally, we can state that close to the nozzle the Taylor micro scales in the isothermal jets and the nonpreheated flames are comparable, whereas the scales in the preheated flames are somewhat larger; that further downstream the mutual differences between the Taylor micro scales in the flames are much smaller than between the flames and the isothermal jets: the Taylor scales in the flames are about 50% larger than in the isothermal jets, whereas the Taylor micro scales of the different flames differ less than about 20%.

8.9.4 Comparison of length scales of NO and of OH

We now compare the length scale results of NO to those of OH, which are discussed extensively in Section 7.11. Remind that the Taylor scales of OH are not defined near the centerline in most flames for the range of \( x \) covered by the measurements, whereas the NO results are less reliable in the outer regions of the flame.

Integral scales

Integral length scales of OH have only been obtained in flames IV, V and VI, at the largest axial distances. Due to the character of the OH correlation functions, integration was not possible in other flames or closer to the nozzle. For NO, the integration converged for all flames, for \( x = 250 \text{ mm} \) and larger.

In Figure 8.50, the integral length scales of both NO and OH are shown. The relation between \( L_{\text{NO}} \) and \( L_{\text{OH}} \) of flames V and VI is qualitatively equal. With respect to these flames, there is a small difference in absolute value of the scales, slightly affecting the correspondence of \( L_{\text{NO}} \) and \( L_{\text{OH}} \).

We find that near the axis, the integral scales have comparable dependence on \( y \), but that the integral scale of OH is generally smaller. This is the case in all flames, at all locations. Near the jet axis, where \( L_{\text{NO}} \) and \( L_{\text{OH}} \) increase with \( y \), the difference is smallest. At \( x = 250 \text{ mm} \), \( L_{\text{NO}} \) is 1.7, 2.0 and 1.6 times larger than \( L_{\text{OH}} \), for flames IV, V and VI, respectively. As can be seen in Figure 8.50, the maximum values in the outer regions of the flame of \( L_{\text{NO}} \) and \( L_{\text{OH}} \) are also comparable, about 8 mm at \( x = 250 \text{ mm} \). This was also the case in the other flames.

At every radial location, the OH integral scales are smaller than the NO integral scales. This is because OH is strongly influenced by chemical reactions. The dependence of the OH signal (or concentration) on the mixture fraction is therefore much stronger than of the NO signal. Due to the chemical reactions OH is subjected to, OH exists in rather narrow regions, and the distances over which OH is transported by turbulent eddies is reduced. This results in a decrease of the OH integral length scales. The effect appears to be largest near the average
Figure 8.50: Integral length scales of NO, $L_{\text{NO}}$ (—) and OH, $L_{\text{OH}}$ (—-), at $x = 250$ mm ($x/d_j = 41.7$) in flame IV.

Location of the OH (or reaction) zone, this is where the OH integral scale profiles exhibit minima. At these locations, the NO integral scales are large, like in the outer regions of the OH profiles. The integral scales of NO have not been determined that far from the axis, because there the fluctuations in the NO signal were too small.

Taylor micro scales

To be able to compare the Taylor micro scales of OH and NO, we again select flames IV to VI, because in those flames, and only for larger $x$, the OH can be measured near the centerline, where the NO is present. In flames where this is not the case, the overlap in the regions of OH and NO is too small. For a flame with significant OH fluctuations at centerline, flame IV at $x = 250$ mm, an example of the relation between the Taylor micro scales of both NO and OH, is shown in Figure 8.51. In this figure it can be seen that both $\lambda_{\text{NO}}$ and $\lambda_{\text{OH}}$ are minimum where the average signals of NO and OH, respectively, are minimum; of course the radial locations of these local minima of the Taylor micro scales are different. The accuracy of the OH Taylor micro scales is estimated as 7%.

In flame IV at $x = 250$ and 300 mm, and in flame VI at $x = 250$ mm, the centerline values of $\lambda_{\text{NO}}$ are 10 to 17% larger than the centerline values of $\lambda_{\text{OH}}$, in flame V at $x = 250$ mm, $\lambda_{\text{NO}}$ is 14% smaller than $\lambda_{\text{OH}}$. This indicates that the centerline NO Taylor scale is not systematically larger, but also that the differences are not very large.

The centerline values of the Taylor scales of OH and NO do not differ
Figure 8.51: Taylor micro length scales of NO, $\lambda_{\text{NO}}$ (—) and OH, $\lambda_{\text{OH}}$ (+), at $x = 250$ mm ($x/d_j = 41.7$) in flame IV.

much, so both scales represent the same physical scale of turbulence. In regions of the flame outside the average location of the reaction zone, the NO Taylor micro scales become significantly smaller than the OH Taylor micro scales, due to the decreased NO signal fluctuations. In the outer regions, therefore, the OH Taylor micro scales give a better indication of the small scale scalar structure than the NO Taylor micro scales. Near the axis, the OH Taylor scales are not always defined, because in some flames (flames I–III) for the range of $x$ covered, centerline OH fluctuations are very small. The NO Taylor micro scales can be measured in all flames at centerline, and are, therefore, most appropriate to represent the small scale scalar structure in the central part of the flame. In flames where the OH scales can be determined at centerline, we find that the OH and NO Taylor scales yield values which differ less than about 20 %.

8.10 Conclusions

The measurements with NO seeded to the fuel allow for the following conclusions. In flames III, due to quenching the total flux of fluorescing NO has at $x = 250$ mm decreased with about 50 %, therefore the fluorescing NO is not conserved and the NO fluorescence cannot fully serve as a conserved scalar in flames. For the isothermal jets, it has been shown that NO is a good seed material.

The visualization of NO in flames, showed that the shear layer turbulence in flames I and II is not completely developed for the region of $x$ covered by the
measurements \((x < 175 \text{ mm})\). Large-scale structures are dominant in these flames. The other flames showed large scales breaking up into small scales much faster. Images showed that the contours at the center of the shear layer are much more convoluted than contours where the reaction zone is expected. This shows why there are turbulent structures in the shear layer that do not strongly influence the OH structure.

Besides the measurements in the flames, one dimensional measurements in isothermal jets of nitrogen seeded with NO, issuing in still air and with primary air flow, are presented. The width of the NO profiles in the isothermal jet without primary air flow agreed within 6 % with the width of mass fraction profiles found by Birch et al.[101] and with Becker et al.[99], and was at \(x = 250 \text{ mm} \) (or \(x/d_j = 41.7\)) 1.3 times larger than the widths of the axial velocity profiles obtained by Stroomer[2]. According to Forstall and Shapiro[110] this is agreement with a turbulent Schmidt number of 0.7. The jet width is represented by a linear approximation \(w_{\text{NO}} = (0.23 \pm 0.01)\left(x - (2.5 \pm 0.8)\right)d_j\).

The average NO profiles of the jet with primary air flow issuing from the annulus \((U_s/U_j = 0.2)\) are about 40 % narrower. Linearization yields \(w_{\text{NO}} = (0.12 \pm 0.02)(x + (4 \pm 3)d_j)\).

The NO profiles of the flames are even narrower, partly because of quenching of excited NO. In flames II–VI, the virtual origin is at 4 to 8 jet diameters upstream, which is further upstream than in the isothermal jets. The flame with lowest exit velocities showed a significantly slower decrease of the average centerline signal between \(x = 50\) and 100 mm than the other flames.

The standard deviation of the NO-seed concentration for the jet with primary air flow (jet II) is larger than for the jet without primary air flow (jet I). The profiles of the isothermal jets show off-axis maxima for \(x = 100 \text{ mm}\) and larger. The standard deviation relative to the average signal at centerline in the jet without primary air flow, \((0.222 \pm 0.017)\), is close to the reported values for larger \(x\): according to Dowling and Dimotakis[111] 0.23, to Becker et al.[99] 0.206 and to Pitts and Kashiwagi[102] 0.235. The relative standard deviations of the jet with primary air flow is close to that of the jet without primary air flow. The radial profiles of the relative standard deviation of the jet without primary air flow correspond very well with those found for the relative axial velocity standard deviation.

In all flames, the relative standard deviation of NO fluctuations is significantly larger than in the isothermal jets for larger \(x\). Due to instabilities, the relative standard deviation in flame I is larger than in the larger exit velocity flames II–IV, for \(x \geq 150 \text{ mm}\). At \(x = 150 \text{ mm}\), the relative standard deviation of flame I is 1.6 times large than that of flame III. For \(x = 50 \text{ mm}\), the relative standard deviation is smallest in the flame with the smallest velocities.

Probability density functions are equal in shape in both jets. At \(x = 50 \text{ mm}\), the centerline distribution function of flame III is significantly different from that of jet II. The most probable signal, and therefore the average signal, is
significantly larger. Apparently, at \( x = 50 \) mm, turbulent mixing in the flame is suppressed, with respect to that in jet II.

Joint distribution functions of signal and gradient, showed that at \( x = 50 \) mm in the isothermal jets there is a correlation between the NO-seed concentration and the squared gradient, indicating that both quantities are not independent. This correlation is about 0.2 at centerline and increases with radial distance, reaching a maximum of about 0.5. At \( x = 150 \) mm, the centerline correlation was found to be very small, so the concentration and squared gradient are almost independent. Again, there is a stronger positive correlation away from the axis. This agrees with the results of Namazian et al.\cite{104}. In flame III, there is a negative correlation of -0.25 at \( x = 50 \) mm at centerline, and a positive correlation for larger radial distance; near the centerline in flame III, the gradients appear to be small at \( x = 50 \) mm when the NO-seed concentrations are large. For larger \( x \), the centerline correlation is positive, and off-axis values are larger than in the isothermal jets. At rather strong correlation of 0.3 between the seed concentration and its squared gradient is found at radial locations where the reaction zone is on average located. Conserved scalar combustion models should therefore best not assume independence between mixture fraction and scalar dissipation, certainly not in the vicinity of the reaction zone.

Characteristic length scales have been derived from spatial (radial) correlation functions. At \( x = 250 \) mm \( (x/d_j = 41.7) \) in the isothermal jet without primary air flow, \( \mathcal{L}_{NO} / x = 2.3 \times 10^{-2} \). This value is in the same order of magnitude as \( \mathcal{L}_T / x = 1.52 \times 10^{-2} \), reported by Corrsin and Uberoi\cite{95} for \( x/d_j = 20 \). After correction for the turbulent Schmidt number of 0.7, yielding \( 1.6 \times 10^{-2} \), our result is in excellent agreement with the \( \mathcal{L}_u / x = 1.57 \times 10^{-2} \) found by Wygnanski and Fiedler\cite{94} for \( x/d_j = 90 \).

At \( x = 250 \) mm in the isothermal jet without primary air flow, \( \lambda_{NO} / x = 5.3 \times 10^{-3} \). After correction for non-unity Schmidt number this is very close to \( \lambda_u / x = 3.7 \times 10^{-3} \) reported by Wygnanski and Fiedler\cite{94}. At \( x = 150 \) mm \( (x/d_j = 25) \), close to the axial location used by Corrsin and Uberoi\cite{95}, we found a scaled centerline Taylor micro scale of \( 1.1 \times 10^{-2} \), comparable to \( \lambda_T / x = 8.5 \times 10^{-3} \) reported by Corrsin and Uberoi. The ratio \( \mathcal{L}_{NO} / \lambda_{NO} = 4 \) indicates that for \( x/d_j = 41.7 \) the turbulence of jet I is well developed.

The primary air flow decreases the integral length scale with a factor of about 2, and the Taylor micro scales with only 15 \% at \( x = 250 \) mm. Combustion significantly increases the integral length scales, with a factor of 2 to 3, yielding for example at \( x = 250 \) mm a centerline scale of 6.2 mm in flame III and 2.9 mm in the isothermal jet with primary air flow. Combustion also increases the centerline Taylor micro scales, with 30 to 50 \% for \( x \geq 150 \) mm, yielding scales in the range of 2.0 to 2.3 mm at \( x = 250 \) mm in flames I to III, compared to 1.6 mm in jet II. Close to the nozzle, the Taylor micro scales in the isothermal jets and in the nonpreheated flames are comparable, whereas the scales in the preheated flame are about 20 \% larger.
The centerline integral scales in flame IV are a factor of 2 smaller than in flame III at \( x = 250 \) and 300 mm, due to higher primary air flow. Integral scales are also decreased by preheating the flows, yielding for example centerline integral length scales at \( x = 250 \) mm of 4.2 mm in flame V, compared to 6.2 mm in flame III.

For larger \( x \), the Taylor micro scales in flames I–III do not differ much, with values in the range of 2.0 to 2.3 mm at \( x = 250 \) mm. For the range of \( x \) covered, the primary air flow is also not of large influence, although the difference increases with \( x \). For large \( x \), the effect of preheating on the Taylor micro scales is small.

When the scales of OH and NO are compared, we find that the integral length scales at centerline are in the same order of magnitude, but that the integral scales of OH are much smaller near the radial location of maximum OH concentration, where the influence of chemical reactions on the integral length scales of OH is strongest. The Taylor scales of OH and NO differ less than 20 % near the axis, for the flames where the Taylor micro scale of OH at centerline can be calculated. This difference is so small that both scales characterize the same physical scale of turbulence.
Chapter 9

General conclusions

In the previous chapters, the experimental results of OH and NO seeded to the fuel led to several conclusions. In this chapter, we summarize the conclusions and relate them to the aims of the study, presented in Chapter 1. One aim was to provide information on the spatial flame structure, in particular the reaction zone structure and the shear layer structure. This information consists of visualization and length scale results of turbulent concentration fluctuations. Another aim was to provide average concentrations, standard deviations and distribution functions of OH and of a nonreactive species. Finally, we aimed to uncover the shortcomings of the current turbulent combustion models.

9.1 Flame structure

Visualization of NO seeded to the fuel, revealed that in flame I, with the smallest exit velocities and an exit jet Reynolds number of $5 \times 10^3$, the shear layer between fuel and air is still developing for $x < 175$ mm ($x/d_j < 29.2$), with large periodic structures (wavelength about 10 mm). These instabilities suggest a Von Kármán vortex street. When the exit velocities are 40 % larger (flame II), the shear layer shows instabilities that start to dissipate into small-scale structures at $x \approx 100$ mm ($x/d_j = 16.7$). Turbulence is not completely developed in this flame for $x < 175$ mm. In the flames with larger exit velocities (flames III–VI), the shear layer seems well developed in the range covered ($25 < x < 175$ mm, or $4.2 < x/d_j < 29.2$).

The OH shows the location of the reaction zone, which is well outside the shear layer in the initial part of the flame. For small $x$, only some large-scale deformation of the OH zone is visible in flame I; for larger $x$, large-scale deformation (with scales in the order of magnitude of 10 mm) becomes more pronounced, but smaller scales of about 1 mm are not present. The smallest scales (in the order of magnitude of 0.1 mm) are not resolved. With larger exit velocities the deformation is by a wider range of scales, and additional increase of the primary air velocity (flame IV) gives a stronger deformation. Also the preheating gives a strongly deformed OH zone, because of the exit velocities.
being about 2.3 times larger due to thermal expansion. Close to the nozzle and primarily in flame IV with large primary air velocity, many holes are present in the OH structure, as evidence of local extinction. For larger $x$ the occurrence rate of the holes decreases. Preheating suppresses local extinction, because chemistry time scales decrease more rapidly with a temperature increase than the turbulence time scales (i.e. the Damköhler number increases).

The large primary air velocity of flame IV compared to flame III, reduces the visible flame length with nearly 20% because of increased turbulent diffusion. Also the larger velocities due to preheating increase turbulent diffusion and reduce the flame length by the same amount (flame V compared to flame III). The combination of large primary air velocity and preheated flows (flame VI) gives the shortest flame. The shear layer in flame I is not developed in the range of $x$ covered. This results in smaller entrainment of air into the fuel jet and hence in a NO-seed profile which is about 20% narrower than in the other flames, and also in a narrower flame.

By comparison of the average profiles of the axial velocity and of the NO-seed concentration, in an isothermal jet without primary air flow, we found that the NO-concentration profiles are 1.3 times wider, corresponding to a turbulent Schmidt number of 0.7. The width of the NO profiles in this jet is in very good agreement with values obtained by Birch et al.[101] and Becker et al.[99]; the jet width is represented by $w_{NO} = (0.23 \pm 0.01)(x - (2.5 \pm 0.8)d_j)$. A primary air flow from the annulus ($U_a/U_j = 0.2$) reduces the NO profile width with about 40%, yielding $w_{NO} = (0.12 \pm 0.02)(x + (4 \pm 3)d_j)$. The NO-seed profiles in the flames (II–VI) are about 20% narrower than in the jet with primary air flow. In flames II–VI, this virtual origin is at 4 to 8 jet diameters upstream, which is further upstream than in the isothermal jets.

The average OH zone width decreases when the exit velocities are increased, at $x = 50$ mm ranging from $(1.45 \pm 0.15)$ mm in flame I to $(0.98 \pm 0.15)$ in flame IV; preheating increases the average OH zone width to $(1.13 \pm 0.15)$ mm. At $x = 300$ mm, in all flames the average OH zone width is $(1.7 \pm 0.5)$ mm.

In isothermal jets, the primary air flow only has a small influence on the Taylor micro scales. For small $x$, the Taylor micro scales in the nonpreheated flames are close to the Taylor micro scales in the isothermal jets, see also Section 9.3. Taylor micro scales of turbulent fluctuations of OH concentration and NO seed are of the same order of magnitude, for not too small $x$; at $x = 300$ mm ($x/d_j = 50$), the scales are $(2.3 \pm 0.4)$ mm in all flames. For $x = 50$ and 100 mm, the Taylor micro scales of OH and NO seed are smallest in the flame with the smallest exit velocities (flame I) and largest in the flame with the largest exit velocities (flame VI); the scales of OH are in range 0.6 to 1.0 mm, the scales of NO in the range 1.4 to 1.8 mm (accuracy 7% and 4%, respectively). It is apparent that the Taylor micro scales are not very sensitive to the flame conditions.

Due to a primary air flow ($U_a/U_j = 0.2$) the integral length scales in the isothermal jets are reduced with a factor of about 2 at $x = 250$ mm ($x/d_j = 41.7$).
9.2. Concentrations and temperatures

Because the primary air flow also reduces the NO profile width, we obtain an approximately constant ratio of the centerline integral length scale and jet width \( L_{NO}/w_{NO} = (0.10 \pm 0.02) \). In the flames, the integral length scales are larger than in the isothermal jet with primary flow, see also Section 9.3. Near the radial location of maximum average OH concentration, which is the average location of the reaction zone, the integral length scales of OH are significantly smaller than the integral length scales of the nonreactive scalar, NO. This is because chemical reactions of OH decrease the distance for which the concentrations are correlated. At \( x = 300 \) mm \(( x/d_4 = 50) \), the integral length scale of OH is 1 to 2 mm in flames IV–VI at that radial location. In the outer regions at \( x = 300 \) mm, the integral length scales of both OH and NO seed are about \((8 \pm 2) \) mm in all flames. The integral length scales are not very sensitive to the flame conditions.

9.2 Concentrations and temperatures

In flame I, the maximum average temperature is 1946 K at \( x = 50 \) mm, decreasing with axial distance; at \( x = 250 \) mm, the maximum average temperature is 1552 K. Increase of the exit velocities with a factor 2 (flame III) decreases the maximum average temperature with 246 K at \( x = 50 \) mm, but does not significantly change the temperature (less than 40 K) at \( x = 250 \) mm. Increase of the primary air velocity (changing from flame III to flame IV) reduces the maximum average temperature with 310 K at \( x = 50 \) mm. The effect of preheating is only noticed for \( x \geq 100 \) mm, giving an increase in maximum average temperature of \((158 \pm 10) \) K at all \( x \) in flame V compared to flame III with equal mass flow rates.

The radial locations of maximum average OH concentration coincide with those of the maximum average temperature. Maximum average OH concentrations decrease with \( x \) in flames I–III. The maximum average OH concentration ranges in flame I from \( 11 \times 10^{15} \) cm\(^{-3} \) at \( x = 50 \) mm to \( 4.6 \times 10^{15} \) cm\(^{-3} \) at \( x = 300 \) mm (accuracy 10 %). Increase of the exit velocities with a factor of 2 (flame III) reduces the maximum average OH concentration at \( x = 50 \) mm to \( 5.1 \times 10^{15} \) cm\(^{-3} \). Due to stronger local extinction, an extra increase of the primary air velocity gives an extra decrease in the maximum average OH concentration to \( 2.8 \times 10^{15} \) cm\(^{-3} \) at \( x = 50 \) mm. For all flames, the maximum average OH concentration at \( x = 300 \) mm do not differ more than 35 %, a much smaller effect than closer to the nozzle, and the average value is \((4.0 \pm 0.5) \times 10^{15} \) cm\(^{-3} \).

The average NO seed concentration in flame I decreases slower than when proportional to \( x^{-1} \) for \( x \leq 100 \) mm, whereas in all other flames the decrease is faster than when proportional to \( x^{-1} \). This is due to the relatively small entrainment in flame I. The influence of the primary air flow is very weak on the average NO seed concentration (flame IV compared to III).

The standard deviations of OH concentration are very large, because OH manifests in narrow regions. At \( x = 50 \) mm, the relative standard deviation ranges
from 0.32 in flame I to 0.77 in flame IV. For larger $x$, the difference between the flames decrease, reaching $(0.77 \pm 0.06)$ at $x = 300$ mm in all flames.

Except for small $x$ in flame I, the distribution functions of OH fluctuations consist of a peak (with finite width) at zero concentration due to intermittency or local extinction, and a flat part for larger concentrations. Maximum concentrations are in the order of $20 \times 10^{15}$ cm$^{-3}$. In the preheated flames, this maximum concentration is slightly larger. Distribution functions of the peak concentration of every instantaneous radial profile, show that local extinction is absent in flame I, very rare in flame II, more probable in flame III, and very probable in flame IV. It is most probable around $x = 100$ mm in all flames. Preheating reduces local extinction. These distribution functions also show that the probability of peak concentrations in instantaneous radial profiles between about $1 \times 10^{15}$ and $5 \times 10^{15}$ cm$^{-3}$ are very rare compared to smaller or larger concentrations: when the concentration is below a concentration of about $5 \times 10^{15}$ cm$^{-3}$, local extinction occurs and OH disappears.

At $x = 50$ mm, the NO seed relative centerline standard deviation is smallest in flame I, 0.10, and largest in flame IV, 0.25. Due to periodicities in flame I, the relative standard deviation in this flame is 64 % larger than in flame III at $x = 150$ mm. This difference decreases with $x$, yielding a 30 % larger relative standard deviation at $x = 250$ mm. At $x = 300$ mm, the relative centerline standard deviation is in the range 0.34 to 0.51.

For $x = 50$ mm, the distribution functions of NO-seed fluctuations in flame I show that air or combustion products very rarely reach centerline; also in flame III this probability is rather small, giving a distribution function with negative skewness. For larger $x$, the distribution functions are symmetric at centerline and have a positive skewness off-axis. The distribution functions can be described with $\beta$-functions.

### 9.3 Influence of combustion on turbulence

At $x = 50$ mm, the centerline distribution function of NO-seed in the isothermal jets have a shape different from that in flame III. At $x = 50$ mm, the centerline probability of small concentrations as a result of entrainment, is reduced. For larger $x$, the difference is not noticed.

Close to the nozzle, Taylor micro scales of NO in isothermal jets and the nonpreheated flames are very close. In the preheated flames, the Taylor micro scales are about 20 % larger. For larger $x$, the Taylor micro scales in the flames are 30 to 50 % larger in the flames than in the isothermal jets. At $x = 250$ mm, the integral length scales of NO seed in the flames are a factor 2 to 3 larger than in the isothermal jet with primary air flow. It was also found that the primary air flow has large influence on the integral length scale: the centerline integral length scales in an isothermal jet with primary air flow are smaller by a factor of about
2 compared to an isothermal jet without primary air flow.

9.4 Relation with combustion models

We found that the primary air flow is very important for local extinction. Models should be able to describe local extinction, otherwise they will predict OH concentrations and temperatures that are far too large, especially when modelling a flame with large primary air velocity.

Although an exit jet Reynolds number of about 5000 would suggest otherwise, the shear layer in a flame with this Reynolds number shows periodicities for \( x < 175 \text{ mm} \), which is about 20% of the flame length. When modelling flames with Reynolds numbers in this order of magnitude or smaller, mixing between fuel and air cannot be assumed to be completely turbulent.

Joint distribution functions of the OH concentration and its squared radial gradient, show that there is a strong correlation between these two quantities. This is also the case for the concentration and squared radial gradient of NO seed, both in flames and in isothermal jets. The correlation increases with radial distance. At the location of the reaction zone, the correlation of the nonreactive scalar and its squared gradient is about 0.3. Combustion models should therefore not assume independence between the two quantities. When using conserved scalar PDF models, a better strategy is to include the joint distribution of the mixture fraction and scalar dissipation; when using PDF models with detailed chemistry, inclusion of the joint distribution function of species concentration and their dissipation is also important: the relation can be very strong, as is shown for the case of OH.
Chapter 9. General conclusions
Principal symbols

\begin{align*}
A_{ij} & \quad \text{Einstein coefficient for spontaneous emission} \\
B_{ij} & \quad \text{Einstein coefficient for stimulated emission} \\
B_v & \quad \text{1st coefficient for rotational energy} \\
c & \quad \text{velocity of light} \\
c_s & \quad \text{concentration of species } s \\
c_s' & \quad \text{peak concentration of species } s \\
D & \quad \text{molecular diffusion coefficient} \\
D_{ax} & \quad \text{Damköhler number } (x = -, 2, 3) \\
D_v & \quad \text{2nd coefficient for rotational energy} \\
d & \quad \text{(hydraulic) diameter} \\
E_x & \quad \text{molecular energy } (x = \text{el, rot, vib}) \\
E_l & \quad \text{laser pulse energy} \\
e & \quad \text{electronic level} \\
F_B & \quad \text{Boltzmann factor} \\
f & \quad \text{distribution function of fluctuations} \\
g_x & \quad \text{degeneracy } (x = \text{el, rot)} \\
I_\nu & \quad \text{spectral laser intensity} \\
J & \quad \text{rotational quantum number} \\
k & \quad \text{kinetic energy of turbulence} \\
L_x & \quad \text{integral length scale } (x = \text{u, T, s)} \\
L_f & \quad \text{flame length} \\
L_{OH} & \quad \text{size of largest eddies transporting OH} \\
l_{OH} & \quad \text{average OH zone width} \\
M & \quad \text{local molar mass} \\
M_s & \quad \text{molar mass of species } s \\
N_s & \quad \text{number density of species } s \\
N & \quad \text{total number density} \\
N_A & \quad \text{Avogadro number} \\
N_i & \quad \text{number density of rotational level } i \\
Q & \quad \text{quenching rate} \\
R & \quad \text{rotational relaxation rate} \\
R e_x & \quad \text{Reynolds number } (x = -, j, a, c) \\
r & \quad \text{radial distance} \\
S_F & \quad \text{fluorescence signal}
\end{align*}
Principal symbols

$Sc$  Schmidt number  (-)
$T_x$  temperature $(x = \cdot, j, a, c)$  (K)
$t$  time  (s)
$U_x$  average exit velocity $(x = j, a, c)$  (m/s)
$u$  axial velocity  (m)
$v$  vibrational quantum number  (-)
$w_x$  jet or flame width $(x = u, s)$  (m)
$X_s$  mole fraction of species $s$  (-)
$x$  axial location  (m)
$Y_s$  mass fraction of species $s$  (-)
$y$  traverse location  (m)

Greek symbols

$\delta$  radial separation  (m)
$\lambda$  wavelength  (m)
$\lambda_l$  laser wavelength  (m)
$\lambda_x$  Taylor micro scale $(x = u, T, s)$  (m)
$\nu$  wavenumber  (m$^{-1}$)
$\sigma_{x,ij,s}$  cross section of collisional energy transfer $(x = r, q)$  (m$^2$)
$\tau$  time scale  (s)
$\Phi$  equivalence ratio  (-)
$\phi_v$  volumetric flow rate  (m$^3$/s)
$\xi$  mixture fraction  (-)
$\rho$  density  (kg/m$^3$)
$\rho_s$  correlation coefficient for species $s$  (-)
$\chi$  scalar dissipation  (s$^{-1}$)
$\omega_e$  1st coefficient for vibrational energy  (m$^{-1}$)
$\omega_{e,x_e}$  2nd coefficient for vibrational energy  (m$^{-1}$)

Subscripts

a  at the annulus exit
bb  broadband detected
c  at the coflow exit
ch  of chemistry
cl  at centerline
el  electronic
F  fluorescence
f  of the flame
Principal symbols

G  of the camera gate
i  of energy level $i$
$j$  at the central jet exit
$j$  of energy level $j$
l  laminar
$q$  of quenching
$r$  of rotational relaxation
rot  rotational
$S$  of the fluorescence signal
$s$  at the center of the shear layer
$s$  of chemical species $s$
sat  at saturation
$sl$  single emission-line detected
split  of spin splitting
st  at stoichiometry
$T$  of temperature
t  turbulent
$u$  of axial velocity
vib  vibrational
vis  visible
$x$  longitudinal
$y$  lateral
$2$  of two-body reactions
$3$  of three-body reactions

Superscripts

$^\text{•}$  corrected for density difference
$^0$  before laser excitation
Principal symbols
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Summary

In this thesis, a study on turbulent fluctuations in natural gas diffusion flames is reported. In particular the fluctuations of the reactive species OH and of a nonreactive species following the fuel flow are considered. Because of the large natural gas reserves of The Netherlands, natural gas is used in many heating appliances, in industry and for domestic use. Several combustion reactions cannot considered to be infinitely fast, and interaction between turbulence and chemistry becomes important. Examples are the formation of NO and of soot, and the existence of local flame extinction. Concentration fluctuation measurements of selected species provide information on this interaction. Radical concentrations can be measured instantaneously and nonintrusively with laser induced fluorescence detection. We selected OH, which is present near the narrow zone in diffusion flames, and NO seeded to the fuel, to study shear layer mixing of fuel and air.

In a diffusion (or nonpremixed) flame, the reaction zone is found at locations where the ratio of fuel and air are within close limits around stoichiometry. In the initial part of the flame this zone is at radial locations well outside the region where the shear layer mixing is most intense; further downstream, the reaction zone moves into the shear layer and the influence of shear layer mixing on the reaction zone becomes larger. OH serves as a chain carrier, attacking fresh fuel molecules and preserving the chain reaction of combustion. In diffusion flames, OH is mainly found close to the reaction zone. When time scales of turbulent mixing are smaller than those of the fastest chemical reactions, or, equivalently, when the Damköhler number is smaller than unity, local flame extinction can occur.

The physical principle on which our nonintrusive concentration measurements have been based, is laser induced fluorescence (Chapter 3). Molecules of the species selected, are excited with strong laser pulses (of 5 ns duration and 100 ms separation), and the subsequent spontaneous emission is detected. This spontaneous emission is proportional to the concentration (or number density) of the species. Collisional energy transfer processes influence the fluorescence signal. This influence can only be corrected for in turbulent flames when local temperatures and major species concentrations are also known, or can be reduced by detecting a single line in the spectrum of the fluorescence emitted. A disadvantage
is that the use of a spectroscope for this purpose allows for point measurements only. Because spatial flame structure is also very important, we have used one- and two-dimensional detection in addition, which is inevitably associated with broadband detection. We have estimated the systematic influence of the collisional energy processes on the broadband detected signals.

The burner used (Chapter 4), consisted of a central 6 mm fuel jet, surrounded by premixed pilot flames and a primary air flow issuing from an annulus with 30 mm hydraulic diameter. Fuel was Groningen natural gas, consisting mainly of methane (81 % by volume) and nitrogen (14 % by volume). The burner was placed in a low velocity coflow and the flame was confined by an octagonal glass flame chamber (0.58 m side to side, 0.9 m high). Exit Reynolds numbers of the flames investigated, range from about 5000 to 10,000 for the fuel flow, and 4400 to 16,000 for the primary air flow. The natural gas and primary air flows have been facultatively preheated to about 700 K.

Setups have been constructed (Chapter 5) to detect OH concentration fluctuations, for point measurements with single emission-line detection, for one-dimensional measurements with broadband detection and for two-dimensional qualitative visualization measurements. OH concentration have been set to an absolute basis with a reference flame, a laminar premixed flat flame. We have found that OH fluorescence was about 70 % of the saturation level in the case of one-dimensional detection. NO has been seeded to isothermal jets and the fuel of flames, to investigate the influence of the primary air flow on the jet and of combustion on the turbulent shear layer mixing. Initial volume fraction of NO was about 0.7 %. NO fluctuations are measured with one- and two-dimensional detection, like in the case of OH. NO fluorescence was almost completely saturated (99.5 %).

The laminar premixed flat flame (Chapter 6), fuelled with methane and air, has been studied under lean (Φ = 0.88) and rich (Φ = 1.17) conditions. Axial temperature profiles have been measured with thin thermocouples. Results appeared to be rather sensitive to the correction for radiation losses, but in the post-flame zone, temperature measurements agree within about 40 K to numerical full-chemistry simulations of the flame. In this model, heat radiation losses of H₂O and CO₂ were included, yielding temperature and OH concentration decreasing with x. Relative OH concentrations, measured with both single emission-line detection and broadband detection, follow the predicted profiles very well.

Visualization of OH (Chapter 7) has shown the influence of shear layer turbulence on deformation of the reaction zone. The lowest exit velocity flame shows only large scale deformations of about 10 mm, and this only for larger x. Increase of the exit velocities gives deformation of the OH zone by smaller scales
also. Large primary air velocity gives frequent local extinction around \( x = 100 \) mm, and strong deformation of the OH zone for larger \( x \). Preheating reduces local extinction and gives more turbulent, shorter flames, with strongly deformed OH structure.

At \( x = 50 \) mm \((x/d_j = 8.3)\) in the flame with the smallest exit velocities and exit Reynolds numbers of about 5000, maximum average temperatures and maximum average OH concentrations are largest, about 2000 K and \( 11 \times 10^{15} \) cm\(^{-3}\), respectively. Both decrease with \( x \), to 1552 K and \( 4.8 \times 10^{15} \) cm\(^{-3}\), respectively, at \( x = 250 \) mm \((x/d_j = 41.7)\). Increase of the exit velocities with a factor two, gives a decrease of maximum average temperature and OH concentration of 246 K and a factor two, respectively, at \( x = 50 \) mm. The influence is rather small for larger \( x \).

Distribution functions show large influence of intermittency and, at several locations, of local extinction. Maximum OH concentrations are about \( 20 \times 10^{15} \) cm\(^{-3}\). We also derived distribution functions of the peak concentration of every radial profile, showing the effect of local extinction separately from intermittency. Local extinction is most intense in the nonpreheated flame with large primary air velocity, decreasing the average OH concentration at \( x = 50 \) and 100 mm with a factor 1.8 and 2.5, respectively, due to the larger primary air velocity.

Comparison of single emission-line detected OH and broadband detected OH showed similar distribution functions. The maximum concentrations, obtained with the single emission-line detection, have been found to be about 20 % larger than those obtained with broadband detection. This difference is attributed to the large influence of shot noise on the weak signals of single emission-line detection, and not to the influence of collisional energy transfer on the broadband detected OH measurements. Hence, this latter influence appeared to be rather small.

Length scales of OH fluctuations have been determined from spatial correlation of one-dimensional OH profiles. Only at the locations with the strongest OH structure deformation, integral length scales have been derived, yielding about 8 mm in the outer regions and about 1 mm close to the average location of the reaction zone. Taylor micro scales in all flames were found to be about 1 mm at \( x = 50 \) mm \((x/d_j = 8.3)\), and 2 mm at \( x = 300 \) mm \((x/d_j = 50)\).

In flames, the total flux of the fluorescing NO seed fluorescence is found to decrease with 50 % in the range \( x = 50 \) to 250 mm due to the influence of collisional energy transfer (quenching) (Chapter 8). Hence, NO fluorescence cannot fully represent a conserved scalar. In the isothermal jets, the flux of fluorescing NO molecules did not change more than 10 % in this range of \( x \), which is not significant.

Visualization of NO seeded to the fuel, showed periodicities like in a Von Kármán vortex street, in the shear layer in the flame with smallest exit velocities \((Re_j \approx 5000)\), for \( x < 175 \) mm. Also in the flame with 40 % larger exit velocities,
the shear layer does not become completely developed in this range of $x$. In all other flames, periodicities have not been observed.

Average and standard deviation profiles of NO seed in a single jet of nitrogen are similar to those reported in literature. Presence of a primary air flow issuing from the annulus with $U_a/U_j = 0.2$ gives average NO profiles which are about 40% narrower than in the absence of the primary air flow.

Taylor micro scales in the isothermal jets are close to those in the flames for small $x$, and are about 1.5 mm, but they are about 50% larger in the flames for larger $x$, about 2.3 mm. Taylor micro scales of NO seed and of OH are comparable for $x = 250$ mm at radial distances with significant amounts of OH and NO. Integral length scales of NO and OH are comparable in the outer regions of the flame, about 8 mm at $x = 300$ mm ($x/d_j = 50$). In the central part of the flame, the OH integral length scales are significantly smaller. OH takes part in chemical reactions, and its concentration is not conserved. Hence, near the reaction zone, the largest distance over which OH concentrations are correlated, decreases.

The results lead to some conclusions on the influence of combustion on turbulence. Considering the centerline distribution functions of NO-seed in both an isothermal jet with primary air velocity and a jet with equal exit velocities at $x/d_j = 8.3$, we found a decreased probability of small NO concentrations in the flame. These small concentrations, more probable in the isothermal jet, are a result of entrainment. This revealed that combustion suppresses entrainment close to the nozzle, resulting in a 30% larger average centerline concentration in the flame at this axial location. For larger $x$, this effect has not been noticed.

Combustion gives an increase in the integral length scales and Taylor micro scales of a factor of about 2 and 1.5, respectively, at $x/d_j = 41.7$.

The results also lead to some recommendations for combustion modelling. Local extinction appeared to be very sensitive to the primary air flow. A reduction in temperature of 300 K and a reduction of the average OH concentration with a factor of about 2 are found for small $x$ when the primary air velocity is increased from $U_a/U_j = 0.20$ to 0.36. Therefore, the model must be able to predict local extinction, for otherwise the predicted temperatures and OH concentrations are far too large. Joint distribution functions of the NO-seed concentration and its squared local gradient, and of the OH concentration and its squared radial gradient, showed a significant correlation between the concentrations and gradients of both species, with a value of 0.3 near the reaction zone for the case of the NO-seed, and even stronger correlation of OH concentration and OH radial gradient. PDF models should therefore not assume independence between for example the mixture fraction and the scalar dissipation rate.
Samenvatting (summary in Dutch)

In dit proefschrift wordt een studie besproken van turbulente fluctuaties in diffusievlammen van aardgas. De aandacht gaat voornamelijk uit naar de concentratiefluctuaties van de reactieve stof OH en van een niet-reactieve stof die de brandstofstroom volgt. Nederland heeft een grote aardgas reserve. Daarom wordt aardgas in Nederland voor veel toepassingen gebruikt, zowel in de industrie als thuis. Van verschillende verbrandingsreacties kan men niet aannemen dat ze oneindig snel verlopen en dan gaat de interactie tussen turbulentie en chemie een rol spelen. Voorbeelden van dergelijke trage processen zijn de vorming van NO en van roet, en lokale vlamuitdovend. Door van bepaalde stoffen de concentratiefluctuaties te bepalen, kan men inzicht krijgen in deze interactie tussen turbulentie en chemie. Met laser geïnduceerde fluorescentie kunnen radicaalconcentraties instantanen gemeten worden zonder daarbij de stroming te beïnvloeden. OH is gekozen omdat het in diffusievlammen slechts aanwezig is bij de (dunne) reactiezone. Door NO toe te voegen aan de brandstofstroom kan de turbulente menglaag tussen brandstof en lucht onderzocht worden.

In een diffusievlam ligt de reactiezone op de plaats waar de mengverhouding tussen brandstof en de lucht dicht bij stoichiometrie is. In het eerste gedeelte van de vlam ligt deze reactiezone buiten het gebied van met de sterkste turbulente menging; verder stroomafwaarts dringt de reactiezone steeds verder door in de turbulente menglaag, waardoor de invloed van de turbulentie in deze menglaag op de verbranding steeds sterker wordt. Bij de verbranding is OH een zogenaamde kettingdrager, die verse brandstof moleculen aanvalt en de kettingreactie in stand houdt. In diffusievlammen komt OH voornamelijk dicht bij de reactiezone voor. Als de tijdschalen van turbulente menging kleiner zijn dan die van de snelste chemische reacties, of anders gezegd, wanneer het Dammöhler getal kleiner is dan één, kan in de vlam lokale uitdoving optreden.

Onze metingen zijn gebaseerd op het principe van laser geïnduceerde fluorescentie (Hoofdstuk 3). Moleculen van de geselecteerde stof worden door een sterke laserpuls aangeslagen. De pulst duurt in ons geval 5 ns en heeft een herhalingsfrequentie van 100 ms. De daarop volgende spontane emissie kan worden gedetecteerd, en is evenredig met de concentratie van de stof. Energieoverdracht door inelastische botsingen kan het fluorescentiesignaal beïnvloeden. In turbu-
lente vlammen kan hier niet voor gecorrigeerd worden als niet gelijktijdig de lokale temperaturen en concentraties van alle belangrijke stoffen gemeten worden. De invloed kan beperkt worden door slechts één lijn uit het fluorescentiespectrum te detecteren. Een nadeel van deze enkelijn-detectie is dat er een spectrocoop voor gebruikt moet worden, waardoor alleen puntmetingen mogelijk zijn. Omdat ook de ruimtelijke structuur van de vlam belangrijk is, hebben we ook één- en tweedimensionale detectie gebruikt, waarbij breedband-detectie onvermijdelijk is. Daarom hebben we een afschatting gemaakt van de systematische invloed van de energieoverdracht door inelastische botsingen op het signaal bij breedband-detectie.

De gebruikte brander (Hoofdstuk 4) bestond uit een centrale brandstofuitlaat met een diameter van 6 mm, omringd door voorgemengde stabilisatievlammetjes. Daaromheen was een uitlaat voor de primaire lucht met een hydraulische diameter van 30 mm. Als brandstof is Gronings aardgas gebruikt, dat voornamelijk bestaat uit methaan (81 vol.% en stikstof (14 vol.%). De brander stond in een windtunnel voor een secundaire luchtstroom met lage snelheid. De vlam was van de omgeving afgeschermd door een acht hoekige glazen vlamkast (0.58 m breed en 0.9 m hoog). De Reynoldsgetallen van de onderzochte vlammen waren aan de uitlaat van de brander tussen 5000 en 10.000 voor de aardgasstraal, en tussen 4400 en 16.000 voor de primaire-luchtstraal. Het aardgas en de primaire lucht kon voorverwarmd worden tot ongeveer 700 K.

Er zijn opstellingen (Hoofdstuk 5) gebouwd om OH concentratiefluctuaties te meten, voor puntmetingen met behulp van een spectrocoop, voor ééndimensionale detectie met breedband-detectie en voor tweedimensionale visualisatie. OH concentraties zijn geijkt door de signalen te vergelijken met het signaal van een voorgemengde laminaire vlakke vlam als referentie. Fluorescentie van OH bleek voor 70 % verzadigd te zijn. NO is aan stikstof van een isotherme straal toegevoegd en aan de brandstof van vlammen, om de invloed te onderzoeken van de primaire luchtstroom op de straal en van verbranding op de turbulente menging. De volume fractie van het toegevoegde NO was ongeveer 0.7 %. Net als bij OH, zijn NO fluctuaties zowel met één- en tweedimensionale detectie bepaald. Fluorescentie van NO bleek bijna volledig verzadigd te zijn (99.5 %).

De voorgemengde laminaire vlakke vlam (Hoofdstuk 6), werd gevoed met methaan en lucht, en is onderzocht onder arme ($\Phi = 0.88$) en rijke ($\Phi = 1.17$) condities. Axiale temperatuurprofielen zijn gemeten met een dun thermokoppel. De resultaten bleken nogal gevoelig voor de stralingscorrectie, maar in de hete gassen boven de vlam kwamen de temperaturen binnen 40 K overeen met numerieke berekeningen met volledige chemie van de vlammen. In het model zijn verliezen door straling van $H_2O$ en $CO_2$ opgenomen, waardoor de temperatuur afneemt met de afstand tot het branderoppervlak. Relatieve OH concentraties,
gemeten met enkellijn-detectie en breedband-detectie, komen zeer goed overeen met de berekeningen.

De visualisatie van OH (Hoofdstuk 7) toonde de invloed van de turbulentie op de reactiezone. In de vlam met de kleinste uitstroomsnelheden blijken alleen grootschalige deformaties van de OH zone voor te komen, ongeveer 10 mm groot, en dit alleen voor grotere afstand van de brander. Vergroting van de uitstroomsnelheden resulteert in deformatie van de OH zone door ook kleinere schalen. Een grote uitstroomsnelheid van de primaire-luchtstroom geeft sterke lokale uitdoving, met name rond $x = 100$ mm, en sterke deformatie van de OH zone voor grotere afstand van de brander. Voorverwarming van de stromingen vermindert lokale uitdoving en geeft kortere en meer turbulentele vlammen, met sterk vervormde OH structuur.

In de vlam met de kleinste uitstroomsnelheden en Reynolds getallen bij de uitlaat van ongeveer 5000, is bij $x = 50$ mm ($x/d_j = 8.3$) de maximaal gemiddelde temperatuur ongeveer 2000 K en de maximaal gemiddelde OH concentratie $11 \times 10^{15}$ cm$^{-3}$. Beide nemen af met $x$, tot 1552 K en $4.8 \times 10^{15}$ cm$^{-3}$ bij $x = 250$ mm ($x/d_j = 41.7$). Vergroting van de uitstroomsnelheden met een factor twee, geeft een 246 K lagere maximaal gemiddelde temperatuur en een factor twee lagere maximaal gemiddelde OH concentratie bij $x = 50$ mm. Verder stroomafwaarts zijn de verschillen klein.

Verdelingsfuncties van OH concentratiefluctuaties tonen een grote invloed van intermittentie en, op verschillende locaties, van lokale uitdoving. De maximale OH concentratie is ongeveer $20 \times 10^{15}$ cm$^{-3}$. We hebben ook de verdelingsfuncties afgeleid van de piek-concentratie in elk radieel concentratieprofiel. Deze verdelingsfuncties tonen het optreden van lokale uitdoving, onafhankelijk van intermittentie. Lokale uitdoving is het sterkst in de niet-voorverwarde vlam met grote uitstroomsnelheid van de primaire-luchtstroom, waardoor, vergeleken met de vlam met kleinere uitstroomsnelheid van de primaire lucht, de OH concentratie op $x = 50$ en 100 mm met respectievelijk een factor 1.8 en 2.5 afneemt.

De verdelingsfuncties verkregen met enkellijn-detectie en met breedbanddetectie zijn vergelijkbaar. Alleen de maximale concentratie, bepaald met enkellijn-detectie bloeke 20 % groter te zijn dan die bepaald met breedband-detectie. Dit verschil is te wijten aan de grote invloed van ruis veroorzaakt door het geringe aantal gedetecteerde fotonen bij enkellijn-detectie, en is niet te wijten aan de invloed van energieoverdracht door inelastische botsingen op het signaal verkregen met breedband-detectie. De laatste invloed bloeke dus vrij klein te zijn.

Lengteschalen van OH fluctuaties zijn verkregen uit de functies van ruimtelijke correlatie van ééndimensionale OH profielen. Alleen als de OH structuur sterk gedefinieer was konden de integrale lengteschalen bepaald worden. Deze integrale lengteschalen bleken 8 mm te zijn in het buitengebied van de vlam en ongeveer 1 mm op de plaats waar de gemiddelde concentratie maximaal is. In alle vlammen bleken Taylor microschalen ongeveer 1 mm te zijn op $x = 50$ mm. 
$(x/d_j = 8.3)$, en 2 mm op $x = 300$ mm $(x/d_j = 50)$.

In de vlammen neemt de totale deeltjesstroom van fluorescerende NO moleculen met 50 % af van $x = 50$ tot 250 mm, door energieoverdracht door inelastische botsingen (Hoofdstuk 8). In vlammen representeert NO fluorescentie dus geen behouden groothed. In de isotherme stralen bleek totale deeltjesstroom van fluorescerende NO niet meer dan 10 % te veranderen. Deze variatie is niet significant.

Visualisatie van NO als brandstofindicator liet periodiciteiten zien in de menglaag van de vlam met de kleinste uitstroom snelheden $(Re_j \approx 5000)$, voor $x < 175$ mm. Deze periodiciteiten lijken op een Von Kármán wervelstraat. Ook in de vlam met 40 % grotere uitstroomsnelheden bleek de menglaag niet volledig turbulent te zijn voor deze axiale afstanden. In de andere vlammen zijn geen periodiciteiten gevonden.

De profielen van de gemiddelde NO concentratie en van de standaard deviatie van NO bleken in een isotherme jet zonder primaire luchtstroom goed overeen te komen met de profielen die in de literatuur genoemd worden. Als er een primaire luchtstroom aanwezig is met $U_a/U_j = 0.2$, is de breedte van de gemiddelde NO profielen 40 % kleiner dan die in de straal zonder primaire luchtstroom.

Bij de vergelijking van de verdelingsfuncties van NO fluctuaties in vlammen en isotherme stralen bleek dat dicht bij de brander de inmenging van lucht door verbranding verminderd wordt. Verder stroomafwaarts is dit niet geconstateerd.

Dicht bij de brander zijn de Taylor microschalen in isotherme stralen ongeveer gelijk aan die in de vlammen, ongeveer 1.5 mm, maar voor grotere afstand tot de brander zijn ze ongeveer 50 % groter in de vlammen, ongeveer 2.3 mm. De Taylor microschalen van NO en OH zijn vergelijkbaar bij $x = 250$ mm en bij radiële afstanden waar de hoeveelheid OH en NO voldoende groot zijn. Integrale lengteschalen van NO en OH zijn vergelijkbaar in het buitengebied van de vlam, ongeveer 8 mm bij $x = 300$ mm $(x/d_j = 50)$. In het centrale gebied van de vlam zijn de integrale lengteschalen van OH significant kleiner. OH neemt deel aan chemische reacties en de concentratie is niet behouden. Daardoor neemt in de buurt van de reactiezone de afstand waarover OH concentraties gecorreleerd zijn, af.

De resultaten leiden tot enkele conclusies met betrekking tot de invloed van verbranding op turbulentie. Bij vergelijking van de verdelingsfuncties van NO fluctuaties in de isotherme straal met primaire luchtstroom en in een vlam met vergelijkbare snelheden, vonden we bij $x/d_j = 8.3$ op de symmetrie-as een afgenomen kans op kleine concentraties in de vlam. Deze kleine concentraties, die dus vaker voorkomen in de isotherme straal, zijn het gevolg van inmenging van lucht. We vonden dus dicht bij de brander in de vlam een afgenomen in-
menging van lucht en productgassen, wat op de as een 30 % grotere gemiddelde concentratie tot gevolg heeft. Verder stroomafwaarts is dit niet waargenomen.

Door verbranding nemen de integrale lengteschalen en Taylor microschalen toe, met respectievelijk een factor 2 en 1.5, bij $x/d_j = 41.7$.

De resultaten leiden ook tot enkele aanbevelingen voor modellering van verbranding. Lokale uittoving blijkt erg gevoelig te zijn voor de primaire luchtstroom. Een vergroting van de uitstromensnelheid van de primaire lucht van $U_s/U_j = 0.20$ tot 0.36 geeft een verlaging van de maximaal gemiddelde temperatuur van 300 K en een verkleining van de maximaal gemiddelde OH concentratie met een factor van ongeveer 2. Het is dus noodzakelijk dat het gebruikte model lokale uittoving kan voorspellen, omdat anders de voorspelde temperatuur veel te hoog en OH concentratie veel te groot zijn. Gecombineerde verdelingsfuncties van de NO concentratie en haar radiële gradient, en van de OH concentratie en haar radiële gradient, toonden in de buurt van de reactiezone een significante correlatie tussen de concentraties en de gradient, met een waarde voor correlatie tussen de NO concentratie en gradient van 0.3, en nog sterkere correlatie tussen de OH concentratie en gradient. Men kan daarom in modellen waarbij gebruik wordt gemaakt van verdelingsfuncties, beter niet veronderstellen dat bijvoorbeeld de mengselfractie en de scalaire dissipatiesnelheid onafhankelijk zijn.
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Curriculum vitae

december 24, 1966    Born in Zaandam

1979-1985         Gymnasium $\beta$ at the Zaanlands Lyceum, Zaandam

1985-1990         Study Applied Physics at Delft University of Technology, graduated \textit{with honours} in the Section Optics, on the subject: "Construction of a continuously tunable semiconductor laser with external cavity"

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