MODELING SOOT FORMATION IN DIESEL ENGINES
USING CONDITIONAL MOMENT CLOSURE

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Abstract

In this study numerical simulations of diesel spray combustion with emphasis on soot formation have been performed using multi-dimensional first order conditional moment closure (CMC) coupled with a reduced \( n \)-heptane chemical mechanism, representing the first application of a semi-empirical soot model within the framework of CMC for spray combustion and for diesel engines.

Validation and application of the model have been conducted at three different experimental test facilities: (1) model validation at the Sandia constant-volume chamber for quasi-steady fully developed \( n \)-heptane sprays, (2) model application at the Wärtsilä marine spray combustion chamber for diesel sprays representative for large two-stroke marine diesel engines and (3) model application at the Sandia heavy-duty diesel engine with intermittent diesel sprays.

**Sandia constant-volume chamber:**

A detailed analysis of \( n \)-heptane autoigniting sprays under diesel engine conditions has first been conducted, including comparison with experimental data of ignition delay (ID) and flame lift-off length (LOL) for different ambient oxygen volume fractions (8-21\%) and ambient densities (14.8 and 30 kg/m\(^3\)). ID was in general over-estimated, particularly for low reactivity cases. Flame LOL was found to agree well for all conditions. The soot volume fraction distribution during the quasi-steady period (3-6 ms) has been compared with experimental data. In general, high soot volume fraction location and semi-quantitative distribution have been well described. Furthermore, for the same test cases, the influence of the combustion model has been assessed by comparing the CMC model with the direct integration (DI) approach (i.e. neglecting turbulent fluctuations) shedding light upon conditions where turbulence-chemistry interaction (TCI) plays a major role.

**Wärtsilä marine spray combustion chamber:**

Numerical simulations of autoigniting diesel sprays with an injector orifice diameter of 0.875 mm from a two-stroke marine diesel engine reference experiment have been performed. Simulation results are compared with
experimental data by means of macroscopic spray development under non-reacting conditions as well as ID time, ignition location and quasi-steady flame LOL for different ambient temperatures ranging between 730 and 910 K.

**Sandia heavy-duty diesel engine:**
The model has been subsequently applied to the Sandia optically accessible heavy-duty diesel engine for five different operating conditions including variations in ambient oxidizer dilution, start of injection (SOI) and intake temperature. Simulation results have been compared with experimental data by means of non-reactive fuel vapor distribution, apparent heat release rates (AHRR), temporal evolutions of quantitative in-cylinder soot mass and natural luminosity. The model was found to well reproduce traces of AHRR with a slight underestimation of the premixed portion of combustion and it was capable to reproduce semi-quantitative trends of soot mass for all five cases. A post-processing methodology for a consistent comparison of 3D computed soot distribution with the 2D measured natural luminosity has further been proposed.

Overall, the CMC framework coupled with a two-equation soot model has demonstrated excellent prediction capabilities for diesel spray combustion over a wide range of applications and is a highly promising candidate for studying soot emissions in the full complexity of diesel engines.
Zusammenfassung


**Sandia Hochdruckzelle**


**Wärtsilä Hochdruckzelle**

Numerische Simulationen von Dieselsprays mit einem Injektordurchmesser von 0.875 mm wurden durchgeführt und validiert anhand von experimentellen Daten eines Referenzversuchsträgers repräsentativ für Zweitakt-SchiffsDieselmotoren. Die untersuchten Größen sind die nicht-reaktive makroskopische
Zusammenfassung

Sprayentwicklung, Zündverzug, Zündort und stationäre Lift-off Länge für einen Lufttemperaturbereich von 730 bis 910 K.

Sandia Heavy-Duty Dieselmotor


Die CMC-Methode kombiniert mit einem semi-empirischen Russmodell hat eine hervorragende Voraussagekraft für die Simulation von Dieselsprayverbrennung über einen breiten Bereich von Applikationen aufgezeigt, und stellt somit eine vielversprechende Methodik für die Analyse von Russbildung in Dieselmotoren dar.
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1 Introduction

1.1 Motivation

Combustion of fossil fuel is a key technology as it constitutes the main source of energy today but also in the foreseeable future. Diesel engines are among the highest efficiency energy conversion devices and, in conjunction with their high reliability, operational flexibility and low production and maintenance costs are therefore heavily relied upon for power generation, seaborne and on-road transport for freight as well as for passenger cars. The success of the quality controlled, non-premixed operation is however aggravated by significant in-cylinder emissions due to rich and lean regions of the combustion zones leading to substantial amounts of soot and nitric oxides.

In recent years, driven by increasingly stringent legislations for pollutant emissions, the development of diesel engines has seen a formidable reduction of gaseous and particulate emissions. These have been achieved through more complex engine configurations including in-cylinder strategies and aftertreatment devices. Since emission related processes lie at the end of the chain of combustion events, physical understanding of all affecting phenomena is essential in order to identify the most promising concepts for pollutant abatement. Computational fluid dynamics (CFD) has become an important tool for supporting experimental investigations in engine research. Numerical simulations allow for deep analysis of all in-cylinder processes; however model validation is a prerequisite to ensure reliable and accurate predictions.

Optically accessible test rigs and engines in conjunction with the emergence of cycle-resolved measurement techniques and high-speed imaging in recent years has significantly improved the situation with respect to the availability of large data-sets for validation of models under well-controlled conditions with quantifiable measurement uncertainties.

This work is aimed at the validation of such models in order to provide an improved understanding of the in-cylinder processes and to further develop simulation tools towards reliable predictions for wide range of operating
conditions and hence aiding in the development of future lowest emission diesel engines.

1.2 Objective of this study

The ultimate objective of this work is to simulate soot formation in a real diesel engine. The pre-existing conditional moment closure framework has been extended to account for soot formation by implementing a semi-empirical two-equation soot model in the CMC context. The performance of the CMC framework for modeling auto-igniting sprays has been assessed at three different optically accessible test facilities encompassing the entire spectrum of diesel engines sizes, viz. automotive, heavy-duty and marine diesel engines.

1.3 Structure of the thesis

The thesis is organized as follows. First, a brief literature review focused on modeling of turbulent combustion and soot formation is given. Chapter 3 presents the relevant numerical methodology employed. Results have been subdivided along the three test facilities considered: (I) Sandia constant-volume chamber, (II) Wärtsilä marine spray combustion chamber and (III) Sandia heavy-duty diesel engine.

In the first part, the soot model has been validated for different \( n \)-heptane sprays and the influence of the combustion model under diesel engine conditions has been assessed with respect to flame structure and soot relevant quantities. In the second test facility the CMC framework is applied to diesel sprays relevant for large two-stroke marine diesel engines and the influence of the ambient temperature is studied. In the third results part the previously validated model is finally applied to a heavy-duty diesel engine for five different reference cases. Finally, Chapter 7 briefly recapitulates the main conclusions of the study and suggestions for future work are provided.
2 Literature review

2.1 Turbulent combustion modeling

Modeling of turbulent combustion is a particularly challenging task due to many chemical reactions interacting with diffusion and turbulent motion involving a large range of time and length scales in an often multi-phase environment. Nonetheless, turbulent combustion modeling has seen a formidable development in the last decades. Various books and review articles presented a comprehensive overview of the progress, e.g. [2-4]. This Section seeks to summarize advances and state-of-the-art modeling aspects with focus on non-premixed combustion encountered in typical engineering relevant applications such as diesel engines.

The simulation of combustion consists of numerically solving conserving equations for mass, momentum, energy and chemical species resulting in a large set of partial differential equations. The physical domain is discretized using different techniques like finite volumes, finite differences and spectral elements. In turbulent flows, a multitude of eddy-structures with different sizes are characteristic. The smallest length scale is described by the Kolmogorov scale being proportional to Re$^{-3/4}$. Thus, for higher turbulence levels the minimal flow length scale becomes smaller and the number of grid points needed for the full resolution of the problem increases rapidly. For real diesel engines, the resolution of all turbulent scales is prohibitive with current computational power available. In this case, not all scales are resolved and a turbulence model is therefore required.

Depending on model accuracy requirements and computational expense constraints, three different methods for the description of turbulent flows are introduced: direct numerical simulation (DNS), large-eddy simulation (LES) and Reynolds-averaged Navier-Stokes (RANS).

DNS solves the Navier-Stokes equations for all flow scales directly on the computational grid and a turbulence model is not required. DNS is a powerful tool for the understanding of fundamental processes, e.g. interaction between chemistry and turbulence, and for supporting the development of predictive
models as well as the interpretation of measurements. High-order schemes are generally used for spatial discretization and time integration and simple geometries are usually considered. The increasing availability of high-performance computing clusters is steadily increasing the range of applicability of DNS towards larger and/or more complex problems [5]. These include e.g. simulations of 3D domains instead of 2D, simulations of higher Reynolds numbers, inclusion of more complex fuels, more complex chemical mechanisms and simulations of higher ambient pressures as well as multi-phase flows.

The RANS approach is the most inexpensive method. The entire spectrum of turbulence is modeled and transport equations describe the evolution of the mean flow and the RANS solution has the physical meaning of an ensemble average of the instantaneous quantities over a large number of flow realizations. Information concerning turbulent fluctuations of the flow are lost. Due to the averaging of the Navier-Stokes equations unclosed terms representing the influence of turbulent transport processes on the mean flow appear. Standard practice for the closure of these terms is the k-ε model with different variants where additional transport equations for turbulent kinetic energy and its dissipation are solved and Reynolds stresses are closed with a turbulent viscosity based on k and ε. The grid resolution requirement is massively reduced compared to DNS as mean flow properties vary over length scales much larger than the Kolmogorov length scale, and typical grid sizes in the order of 1 mm are generally used. This makes RANS an ideal candidate for the simulation of high Reynolds number flows encountered in industrial applications at a modest computational cost.

LES is an intermediate approach between DNS and RANS and is becoming increasingly a widely used tool also for application relevant configurations. In LES, large turbulent scales are resolved and smaller scales are modeled. A spatial filtering of the Navier-Stokes equations is applied. The filter size is generally set to be the characteristic grid size. The resolution requirements for LES are not unambiguously defined. Following Pope [6], as a rule of thumb 80 percent of the energy should be resolved. However, different levels of resolution are commonly employed and according to Rutland [7] are subdivided into: scientific and engineering LES. For typical diesel spray simulations, high-fidelity LES has a grid size of about 0.1 mm [8] and engineering LES in the range of 0.15-0.3 mm. An advantage of LES compared to RANS is the ability to capture unsteadiness of processes such as cycle-to-cycle variability, e.g. [9], or the accurate prediction of recirculation zones in gas turbine burners [10]. The general trend for diesel engines simulations is clearly towards LES. However
some time is still needed for the development of different LES-specific sub-models such as the spray models which have been originally developed in the framework of RANS [7].

2.1.1 Chemistry model

The description of chemical models for conventional diesel fuels derived from petroleum sources is highly complex as the fuels are composed of hundreds to thousands of compounds. Models that consider all these components are currently computationally prohibitive. Thus, the real fuel is modeled with a simplified surrogate fuel mixture consisting of a reduced number of pure components matching the chemical and physical characteristics of the target fuel. For petroleum-based diesel, five primary classes of components are commonly identified: n-alkanes, iso-alkanes, cycloalkanes and aromatics [11]. For higher hydrocarbons (e.g. $n$-heptane or $n$-dodecane), even detailed mechanisms [12] of single component are still prohibitive from CFD applications. These models are usually employed for perfectly stirred reactor configurations.

This section is therefore intended to review reduced chemical models commonly used for multidimensional CFD simulations for diesel engine relevant configurations. Despite various diesel surrogates mixtures, pure $n$-heptane is the most used due to its similar cetane number as diesel fuel but also because of the vast availability of compact reduced mechanisms in the range of 30-60 species [13-18]. Note that the LLNL detailed $n$-heptane mechanism [19] has 561 species and 2539 reactions. The ERCv2 model [13] developed at University of Wisconsin has seen a wide application for engine simulations, e.g. [20-23]. The mechanism has 29 species and 52 reaction and was originally validated for HCCI combustion. The mechanism by Liu et al., [15] has been successfully applied for diesel sprays in constant-volume vessels [24-26] as well as in diesel engines [27]. Another mechanism encountered in diesel simulations is the one by Golovichev et al., [14] applied for diesel sprays, e.g. [28]. Pei et al. [25] performed $n$-heptane spray simulations with the composition PDF method and compared the performance of three chemical mechanisms [13, 15, 18] and for the given configuration the mechanism by Lu [18] was found to perform at best. During the ECN2 Workshop [29] various chemical mechanisms using different combustion models have been directly compared and there was not a conclusive trend of a higher model performance for the more complex chemical model. From the perspective of additional soot emission modeling, $n$-heptane/toluene blend [30, 31] has also seen considerable application motivated by the fact that
diesel fuel contains considerable amounts of fuel-bounded aromatics components, which are expected to influence soot formation [11].

2.1.2 Turbulence-chemistry interaction

When it comes to simulation of reactive flow with sufficiently high Reynolds numbers RANS or LES methods are employed solving equations for averaged (filtered) quantities. The chemical rate of reaction spans a broad range of timescales and in some cases those timescales overlap with the physical timescale of the flow [4]. In this case the chemistry cannot be treated separately from turbulence. Here the notion of turbulence-chemistry interaction (TCI) is introduced and is generally related to the closure of the chemical source term, which represents one of the main challenges in turbulent combustion modeling during its development in the last four decades [32]. The inherent difficulty is that chemical reactions take place at the molecular level and that the chemical source term is strongly non-linear obeying a classical Arrhenius-type expression with exponential temperature dependence. As in RANS and LES not all spatial scales are resolved, some fluctuations are still present within the cell. Thus, the evaluation of the mean reaction rate using the mean composition and temperature within the cell can lead to considerable errors [4]. This poses a tremendous challenge and different modeling strategies have been proposed. An overview of main TCI closures with representative applications for diesel engines follows.

It is worth nothing that currently, contrary to non-premixed flames, for diesel engines there is no clear consensus about the necessity of a TCI closure yet. In recent years with the advent of more detailed measurements with more stringent set of validation data this question is being addressed in more detail. The Engine Combustion Network (ECN) [29] seeks to provide a common platform for the comparison of numerical as well as experimental activities from different groups worldwide focusing on well-defined target conditions. At the recent ECN2 Workshop models with a TCI closure were overall found to perform better. However, no decisive conclusion about the importance of TCI closure for diesel spray has been achieved and the neglect of TCI remains questionable.

For the auto-ignition process in diesel engines, the conventional modeling approach in the past has been to solve transport equations for the ensemble-averaged reacting scalar mass fractions using chemical source terms with semi-empirical Arrhenius expressions [33], but completely neglecting turbulent fluctuations. One justification for this widely adopted “Shell model” was that the chemistry of auto-ignition is slow relative to the turbulent timescale. An
additional pragmatic reason was that, at that time, no comprehensive turbulent reacting flow models were available, which could be used for problems for a wide range of Damköhler numbers. In this, relatively dated, conventional approach, after auto-ignition the model switched to a turbulent combustion model for the non-premixed part of the diesel combustion process (e.g. Magnussen’s Eddy-Dissipation) [34, 35].

With the development of the flamelet model in the 90’s, more advanced turbulent combustion theories with different TCI closure strategies started to appear in the field of diesel engine CFD. These include e.g. perfectly stirred reactor (no model), partially stirred reactor (PaSR) [36], generalized Flame Surface Density model [37], Flamelet/Progress variable models [38], Flamelet Generated Manifold method [39], Flamelet [40], transported PDF [41] and CMC [42].

The simplest TCI closure is no closure, i.e. the mean chemical source term is evaluated using directly the mean temperature and composition neglecting turbulent fluctuations. In the literature this approach is referred to by different names, e.g. perfectly stirred reactor, well-mixed, direct integration, CHEMKIN, detailed chemistry and no model. In the engine community this method is not unlikely to be encountered and, despite the conceptual limitations, numerous successful applications for diesel engines have been reported, e.g. [20, 22, 43, 44]. The applicability of the well-mixed model for diesel combustion has been recently discussed in [45]. Following results reported in [46] where the role of flame propagation has been investigated, it was argued that diesel combustion is mainly governed by volumetric heat release (except at the lift-off position) and a TCI closure is of secondary importance.

An extended version of the well-mixed model is the Chalmers PaSR model which is based on the idea that each computational cell can be divided into a reacting part and a non-reacting part. The reacting part is treated as a perfectly stirred reactor and the sub-grid diffusion with the non-reacting part is due to micro-mixing approximated with the help of a micro-mixing time as suggested in the interaction with the mean (IEM) approach typically used in PDF methods. The PaSR is a semi-sophisticated TCI closure with considerable popularity for auto-igniting sprays, e.g. [28, 47, 48].

One of the most used TCI closure is the laminar flamelet model, originally introduced by Peters as steady laminar flamelet method (SLFM) [49] in a RANS context and extended to LES subsequently [50]. The flamelet theory is applicable to high Damköhler numbers and therefore an intrinsic separation of
scales is applied. The thickness of the flame is small compared to the size of the eddy, thus the flame is treated as a one-dimensional laminar flame sheet embedded in the turbulent flow. The flamelet model is a conserved scalar approach (the same as CMC); for non-premixed combustion the gas-phase mixture fraction is used as conditioning quantity, describing the state of mixing. The flamelet equation for chemical species is a pure reactive-diffusive equation accounting for micro-mixing and arbitrary chemistry. Contrary to the CMC method, convective terms as well as turbulent fluxes in physical space are absent.

For the simulation of diesel engines various model developments have been presented compared to the original SLFM formulation, as the latter does not account for transient phenomena like autoignition. The main model increment was the extension to unsteady flamelet known as representative interactive flamelet (RIF) [40] where one flamelet is representative for the entire domain but the flow field is updated at every time step and thus a temporal evolution of the scalar dissipation rate is considered. Further developments are due to the Eulerian particle flamelet model (EPFM) [51] subdividing the computational domain and assigning these domains to different flamelet histories associated with Eulerian markers. For a diesel engine, given a sufficient number of flamelets, the EPFM was found to provide considerably better predictions than RIF.

The flamelet model has also been applied to diesel engines operating with a multiple injection configuration [52-54]. For this scope a two-mixture fraction formulation (two-dimensional flamelet) representing a three-feed system has been derived using a three-scale asymptotic analysis [52]. The foundation of the model has been recently tested in a priori analysis using 2D-DNS simulation of a three-feed system where the DNS solution for scalar dissipation rate has been imposed to the two-dimensional flamelet model [55]. The latter was found to accurately represent the interaction of the multiple stream mixing and chemistry. Despite its importance, an accurate closure for the cross scalar dissipation rate is currently not existing and simplified expressions are commonly employed [52-54].

A further extension of the flamelet model is the flamelet/progress variable (FPV) method introduced in [38]. All detailed chemical processes are mapped to two independent tracking scalars: the mixture fraction, which describes the state of mixing of fuel and oxidizer, and the progress variable, which tracks the global extent of reaction at the given mixture. The FPV approach has been successfully applied for diesel spray combustion [24].
A similar method is the flamelet generated manifold [39], where solutions of one-dimensional flamelet equations are pre-computed, the so-called flamelet manifolds, with different prescribed values of scalar dissipation rate, pressures, temperatures and oxidizer dilutions for a given detailed chemical mechanism. Look-up tables with the mean chemical source term are then constructed as a function of means and variances of mixture fraction and progress variable. For auto-igniting sprays the model has seen considerable success in a constant-volume vessel given an appropriate tabulation strategy, e.g. [8, 56], and for real diesel engines the definition of pressure dependent manifolds are necessary [57].

The transported PDF method presented in [41] and later reviewed in [6, 58] is a powerful TCI closure approach that solves the transport equation for the one-point, one-time Eulerian joint PDF of velocity and composition or alternatively composition only. The main advantage compared to previous TCI closure strategies is that no closure problems arising from averaging of one-point nonlinear terms in the governing equations such as chemistry and radiation are encountered. On the other hand the micro-mixing term needs to be modeled. Various mixing models have been proposed, mainly the exchange with the mean (IEM), the modified curl (MC) and the Euclidean minimum spanning tree (EMST). The transported PDF method has the largest validity range and can be applied irrespective from the premixedness of the problem, i.e. premixed, non-premixed and partially-premixed. This generality of the model comes with the price that it is also the most expensive one. The solution of the large problem is solved numerically using stochastic methods such as the Monte Carlo based Lagrangian stochastic particle method. The PDF method is an ideal candidate for the investigation of TCI as well as turbulence-radiation interaction (TRI) [59]. The composition PDF method has seen application with highly promising results for HCCI [60] and diesel engines [61]. More recently the model has been applied for \( n \)-heptane and \( n \)-dodecane spray combustion under diesel engine conditions with excellent results [21, 25, 62].

The CMC method used in this study will be discussed in detail in section 3.2.1.

2.2 Soot modeling

Modeling of soot formation and oxidation, in particular in diesel engines, is an extremely challenging task, as soot processes are heavily affected by previous processes, e.g. liquid spray atomization, fuel evaporation, fuel/oxidizer mixing field evolution, autoignition and flame characteristics and soot precursor
evolution. Therefore, for an accurate prediction of soot behavior all the aforementioned processes must be accurately described.

This section is aimed at reviewing various aspects of soot modeling. First, the generic soot phenomenology is summarized. Then, various soot modeling approaches with different complexities are presented, from empirical up to detailed models.

2.2.1 Soot phenomenology

Soot is a solid graphite-like structure mainly consisting of carbon with a minor part of hydrogen. The density of soot is reported to be 1.84 ± 0.1 g/cm³ [63]. Soot is formed during combustion of hydrocarbons under substoichiometric conditions or high temperature pyrolysis. The emission of soot from a diesel engine or a generic flame is determined by the competition between particle formation and oxidation. A schematic representation of processes involved during soot formation is displayed in Figure 2-1. In the literature, processes are commonly identified as: pyrolysis, inception, surface growth, coagulation, agglomeration and oxidation. The characteristic size of molecules and particles increases from left to right; the typical size of a nascent soot particle is around 1-2 nm. The temporal evolution of the processes follows as well the same direction.

![Figure 2-1: Schematic representation of the fundamental soot formation mechanisms (adopted from [64]).](image-url)
In the following, a brief review of soot formation mechanisms is provided. Here, it is important to mention that most of the fundamental soot related work has been conducted for atmospheric or even sub-atmospheric flames. For higher pressures encountered in diesel engines the knowledge of fundamental mechanisms is much more limited due to the lack of experimental data [65].

Particle inception is the transition of gas-phase species to solid particles and is probably the least understood part of the soot formation process. Various key gaseous precursors to soot have been proposed, e.g. polyacetylenes, ionic species or PAH. In the literature, the vast majority attribute soot to form via PAHs [66].

In the flame, as hydrocarbons pyrolyze, primarily smaller hydrocarbons are produced and acetylene is the most abundant species. The growth of the gaseous species prior to soot formation is mainly governed by the well-known HACA (hydrogen-abstraction-acetylene-addition) mechanism pioneered by Frenklach and coworkers [66, 67]. Of particular importance during the formation and growth of aromatic species is the formation of the first aromatic ring (benzene) from small aliphatics, as is believed to be the rate-limiting step towards higher aromatics rings. A detailed reaction mechanisms for PAH formation is reported in [67]. The production of complex aromatic components (e.g. pyrene) is seen to be sensitive to a large range of elementary reactions. Bottleneck in the formation of PAH can be promoted by the kinetic reversibility in the HACA mechanism, allowing other paths to compete, e.g. propargyl recombination, stepwise dehydrogenation of cyclohexane [68]. Oxidation of aromatics is parallel to the formation of PAH. Oxidation is mainly due to molecular oxygen and OH seems to be unimportant [66]. PAH oxidation is particularly important in the early phase of PAH growth when considerable O₂ is still present. The latter is rapidly consumed in fuel-rich environments sustaining aromatics growth.

Following Wang [68] different paths are distinguished for particle nucleation. One is a quasi-two-dimensional growth of PAHs into curved fullerene-like structures and the other one involves the physical coalescence of PAHs into stacked clusters and finally coalescence into crosslinked three-dimensional structures. The nucleation process and the respective accumulation of particle mass seems to be a combination of chemical reaction with gaseous precursors simultaneously with the growth of particles size by collision among PAH clusters [66]. As a consequence, the soot number density function is often bimodal as observed experimentally [69]. The first mode consists of small
particles formed by persistent nucleation from PAH and the second derives from larger particle grown by collision and surface reactions.

Once soot particles have been formed, they can grow by two mechanisms: coagulation and surface growth. The latter is the process of adding mass to the nucleated particle. Surface growth is responsible for the increase in soot mass whereas the number of particle remains unaffected. Acetylene plays a major role for the chemical surface reaction in the context of the HACA mechanism as is the case for the PAH growth. Experimental studies observed that initially the particles look spherical and later acquire a fractal shape [70]. The initial coagulation consists of a coalescent growth forming new spherical particles. The process is described by the Smoluchowski master equation. Depending on the Knudsen number, different coagulation regimes are distinguished: at low pressures coagulation takes place in the free-molecular regime, while at high pressures in the continuum regime. Later in the process soot particles agglomerate forming chain-like structures of spherules with a diameter of about 30-50 nm [71]. Soot oxidation is a chemical surface reaction and takes place during the entire period as displayed in Figure 2-1. Oxidation occurs primarily as a result of O₂ and OH. Other oxygenated species such as O, H₂O and NO₂ may be important under some conditions [72].

For what concerns experimental data availability for diesel engine relevant conditions, traditionally soot is measured in the exhaust and thus no information about the temporal soot evolution is available. One of the main challenges in soot modeling in diesel engines is that most of the in-cylinder soot is oxidized and only a small part is retrieved in the exhaust. In recent years, optically accessible test rigs and engines in conjunction with the emergence of cycle-resolved measurement techniques and high-speed imaging has significantly improved the situation with respect to the availability of large data-sets for validation of models under well-controlled conditions with quantifiable measurement uncertainties. Various measurements techniques are employed for reactive diesel sprays, these include active methods such as laser induced fluorescence (LIF) of CH₂O, OH, PAH [73, 74], and soot laser induced incandescence (LII)[75], and passive methods such as OH* chemiluminescence, two-colour thermometry, natural luminosity as well as Schlieren imaging.

2.2.2 Semi-empirical models

Semi-empirical models solve a set of equations (one or two) with reaction rate constants calibrated against experimental data. Soot formation and oxidation
processes are described phenomenologically and different model complexities have been proposed in the past decades.

Each sub-process (i.e. inception, surface growth, coagulation and oxidation) is generally described by a single reaction/step. One of the most simple but still considerably popular in the diesel engine community is the one-equation two-step model by Hiroyasu and co-workers [76]. The model considers a soot mass formation step linked directly to the fuel vapor concentration and an oxidation step via molecular oxygen. However, the model needs specific calibration for different engine operating conditions.

Two-equation soot models are widely used for CFD applications, where transport equations for the soot mass and number density are solved, corresponding to a reduced method of moments where the first two moments (M0 and M1) of the particle size distribution (PSD) are solved. The PSD variance (M2) is assumed to be zero and therefore particles are assumed to be spherical and mono-dispersed. To relax this assumption a three-equation formulation can be employed where a log-normal shape of the particle size distribution is commonly assumed [77, 78] as originally proposed by [79].

Prominent two-equation models have been proposed by e.g. Leung et al. [80], Lindstedt [81] and Fusco et al. [82], and various derivates denoted as multi-step or phenomenological soot models are used, e.g. [83, 84]. Soot inception is generally based on acetylene only in a one-step reaction. More complex inception paths such as via PAHs belongs to the class of detailed models and are discussed in section 2.2.3. Inception has a minor contribution to soot mass, however is the unique source for the particle number. The size of a fresh nucleated particle is a model parameter expressed in terms of the number of elementary carbon atoms that a new particle consists of. Leung et al. [80] proposed 100 C-atoms corresponding to a sphere with 1.24 nm diameter. Alternatively Lindstedt [81] proposed 60 C-atoms.

Also soot surface growth is a function of acetylene only and depends on the square root of the specific soot surface area, describing the availability of active sites at the particle surface for the acetylene adsorption. The square root function is generally motivated by particle aging as the particle becomes larger and therefore the reactivity is reduced. A linear dependency with respect to the surface area has also been alternatively proposed [81] but is less common.

Particle coagulation is described by the Smoluchowski master equation. In semi-empirical models only the first two moments are considered, i.e. spherical and mono-dispersed particles. Collision frequency for limit cases of free-molecular
(Kn>>1) and continuum (Kn<<1) coagulation are employed [85]. For the transition regime of coagulation an harmonic mean of the limit values can be applied [79]. In general, semi-empirical soot models account for coalescent coagulation and the agglomeration is not considered on the contrary to more advanced particle dynamics approaches discussed in the next section.

Soot oxidation is a surface reaction and the soot mass is reduced whereas the number of particle remains unaffected. The most widely used model for O\textsubscript{2} oxidation is the one by Nagle and Strickland-Constable (NSC) [86] or alternatively with a single reaction [80]. Oxidation by OH is also commonly integrated as a single step reaction following e.g. [87].

### 2.2.3 Detailed models

Detailed models attempt to describe the detailed elementary chemical reaction and physics of soot formation, which is subdivided into gas-phase (PAHs formation) and surface (surface growth and oxidation) chemistry and particle dynamics (coagulation). In the following, these modeling components are summarized separately.

As the PAH formation takes place at the gas-phase, the PAH chemistry is integrated with the fuel oxidation chemistry. A typical fuel that has seen a wide application is acetylene. Soot precursor chemistry leads to the formation and growth of larger PAHs until nucleation occurs, i.e. transition from gaseous to solid phase. Detailed PAH mechanisms have around hundred species and hundreds of reactions validated for shock tubes, laminar premixed and counterflow diffusion flames [67, 88]. The mechanisms describe the fuel pyrolysis and oxidation, benzene formation and PAH mass growth and oxidation. Blanquart et al. [89] developed a detailed mechanism with emphasis on soot formation for the high temperature combustion of engine relevant fuels ranging from methane to iso-octane. The mechanism is composed of 149 species and account for all major pathways of PAH formation. Also reduced PAH growth mechanisms have been recently developed. Bisetti et al. [90] performed the first two-dimensional DNS of an n-heptane/air turbulent non-premixed flame with soot formation including a reduced version of the PAH mechanism from Blanquart et al. [89] with 47 species and 290 reactions. Xi and Zhong [91] reduced the detailed PAH mechanim by Wang and Frenklach [67] for n-heptane combustion with PAH formation up to pyrene to 48 species and 76 reactions and the model was successfully applied in a diesel engine by [20]. PAH molecules grow and primary particle are nucleated. The transition is chosen by four or more fused aromatic rings.
With the advent of nucleated particle there is the necessity to describe its dynamic and chemical interaction with the gas-phase. The evolution of the soot number density function (NDF) is governed by the population balance equation (PBE), which has a high dimensionality and a direct solution is intractable and various numerical techniques can be used to obtain an approximate solution as reviewed in [92]. Two classes of methods are distinguished: one solves for a set of mean quantities of the NDF (moment-based method) and the other solves for an approximate form of the NDF.

For the solution of the approximate NDF there are, again, mainly two strategies. One is using a Monte Carlo based statistical method, which is the more accurate but also computationally more intensive. This technique is limited to simple configurations such as homogeneous reactors. The second is the sectional method, where the NDF is discretized in bins along the particle size coordinate. A transport equation for every size bin has to be solved, and typically 25-50 bins are required [93, 94], making the method considerably expensive for calculation.

On the other hand, the method of moments is computationally the most efficient approach for solving the PBE. In principle, the knowledge of all the moments is equivalent to knowing the distribution itself. In practice, the properties one considers is determined by just the first few moments. Equations are solved for every moment considered and complete information about the population is lost. The moment equation for the highest moment is unclosed as the next higher moment is required to compute the source term of the lower moment. In the literature, three different closure models are usually the most popular practices: the method of moments with interpolative closure (MOMIC) [95], the direct quadrature methods of moment (DQMOM) [96] and the more recent hybrid method of moments (HMOM) [97]. MOMIC is the most widely used approach, the equation for the highest moment is closed by logarithmic polynomial interpolation. Despite its simplicity MOMIC is unable to account for bimodal soot NDF as put forward in [97]. DQMOM is numerically more complex than MOMIC but is capable to account for multivariate distributions of the NDF. A linear system must be inverted to obtain the source term for the transport equation. However, depending on the shape of the NDF, the inversion may be ill-posed. Motivated by the shortcoming of the first two methods of moments, Mueller et al. [97] proposed a hybrid method combining the advantages the these two approaches, i.e. numerical robustness and bimodal mode of the NDF. The model has been successfully applied in the framework of LES and DNS for non-premixed flames [90, 98].
Soot particles have a much higher density (or representative molecular weight) than gaseous species and therefore the molecular mass diffusivity of soot is much lower and approaches zero. This results in a Lewis number much higher than unity. The preferential diffusion effect of soot has been an intense topic of debate in the last decade in the context of non-premixed flames. Conceptually, the effect of differential diffusion is expected to decrease for higher Reynolds numbers as the contribution of the molecular diffusivity becomes negligible with respect to the turbulent transport [42, 99]. For low pressure gaseous flames, different closures have been proposed in the context of mixture fraction based models: for flamelets [100] and CMC [101, 102]. In general, inclusion of preferential diffusion showed improved agreement with experimental soot distribution for both methods RANS [103] and LES [104]. The peak soot volume fraction was observed to increase by a factor of two [103]. More recently, DNS investigations of soot formation have started to shed light upon governing processes around soot dynamics [78, 90, 105] and will further support model development. However, in real diesel engine the importance of differential diffusion is still unclear.

An important physical phenomenon for combustion processes, in particular with the presence of soot particles, is radiative heat transfer, which is a formidable challenge when it comes to be coupled with turbulent reactive flows. The radiative transfer equation (RTE) is an integro-differential equation with six independent variables: three spatial coordinates, two angular coordinates (direction of propagation) and one spectral variable. Time dependence is generally neglected as a radiation beam travels at the speed of light, which is higher by several order of magnitude compared to typical flow velocities [106]. The direct solution is intractable and various techniques can be used, these include a ray tracing, a spherical harmonics method, a discrete ordinate method (DOM) or a Monte Carlo method. Starting from the full dimensionality of the problem, various simplifications may be applied, e.g. gray medium properties or optical thin formulation. The latter may apply for low levels of soot and ambient density where radiation self-absorption is negligible. Due to the high levels of soot in diesel engines, an optically thin assumption does not apply, thus radiation for diesel engines is generally neglected [20, 22, 31, 77, 107]. Alternatively, Yoshikawa and Reitz [108] employed the DOM approach to investigate the effect of radiation on diesel engine combustion.

Analogous to the interaction between turbulence and chemistry, turbulence-radiation interaction (TRI) arises from the highly non-linear coupling between fluctuations of radiation intensity and fluctuations of temperature and chemical
composition of the medium. The influence of TRI has been widely investigated for various premixed and non-premixed flames, e.g. [109, 110]. Here the composition PDF method has demonstrated to be highly suitable for the TRI closure. Accounting for TRI increases considerably the radiative heat loss and as a consequence reduces local gas temperature [111].
3 Methodology

In this chapter, first, a brief overview of CMC related works that have been carried out in the literature is given. Then, the numerical methodology employed throughout this work is presented and it is subdivided into CFD and CMC related governing equations, depending whether equations are solved in conserved scalar space or not. CFD governing equations describe the evolution of the two-phase flow field including the liquid fuel evaporation and mixing as well as the distribution of the gas-phase mixture fraction and its variance. In the CMC governing equations section the CMC model equations and the respective modeling closures are presented as well as the soot model.

3.1 Overview CMC activities

Combustion modeling by means of conditional moment closure (CMC) was motivated by the non-linear behavior of the turbulent reactive source term and the corresponding need for an accurate closure of this term. From its derivation by Klimenko [112] and Bilger [113] in the early 90’s the CMC method has seen a wide development and application for which a comprehensive review can be found in [42] and more recently in [2]. CMC was originally employed for a number of different single-phase turbulent reactive problems in a RANS context, such as non-premixed attached flames [114, 115], lifted turbulent flames [116-118], piloted turbulent flame [115] and autoigniting jets of methane [119, 120] and n-heptane [121]. In a LES context LES-CMC has seen application for lifted flames [122], bluff-body stabilized flames [123, 124], autoigniting jets of hydrogen [125, 126], piloted flames with extinction and re-ignition [127, 128] and spray flames [129].

Soot formation has been studied using a two-equation model for different gaseous non-premixed flames e.g. methane [103, 104], propane [130] and ethylene [131].

CMC has also been successfully used for two-phase flows at conditions relevant for diesel engine applications. Spray auto-ignition has been studied in detail for
different generic test rigs such as the one installed at Sandia [26, 132, 133], Aachen [134, 135], ETH [136]. For what concerns real diesel engines, simulations have been performed at the ETH heavy-duty diesel engine [137, 138] where heat release rates and NOx emissions have been compared with experiments. Recently, soot as well as NOx formation have been studied at the Sandia heavy-duty engine [27, 139]. For the latter work, various results are presented in section 6.

More recently, CMC has been used for premixed flames with the progress variable as conditioning quantity [140, 141].

3.2 CMC related governing equations

3.2.1 CMC equations

The conditional moment closure is a conserved scalar approach and belongs to the class of presumed PDF methods. The CMC governing equations have been derived independently by Klimenko [112] and Bilger [113] in the early 90’s. In the case of non-premixed combustion the gas-phase mixture fraction is used as the conditional quantity. Transport equations for temperature and species mass fraction are derived conditionally on the mixture fraction. For a detailed derivation of the CMC governing equations, the reader is referred to Klimenko and Bilger [42] and more particularly to Mortensen and Bilger [142] for the governing equations for spray combustion. A brief presentation of the equations follows. The conditional expectations of temperature, $T_{\alpha}$, and of the $\alpha$-th species mass fraction, $Q_{\alpha}$, are introduced as

$$Q_{\alpha}(\eta, \mathbf{x}, t) = \langle \mathbf{Y}_{\alpha}(\mathbf{x}, t) | \xi(\mathbf{x}, t) = \eta \rangle$$

$$Q_{T}(\eta, \mathbf{x}, t) = \langle T(\mathbf{x}, t) | \xi(\mathbf{x}, t) = \eta \rangle$$

where $\eta$ is the sample space variable of the mixture fraction. The two arguments separated by the vertical bar in the angular brackets represent the ensemble average of the quantity on the left side for the case of fulfilment of the condition on the right side of the vertical bar.

In a RANS context, following Bilger’s decomposition approach [113], an instantaneous scalar is decomposed into a conditional mean and a fluctuation as
Methodology

\[ Y_a(x, t) = Q_a(\eta, x, t) + Y_a''(x, t) \]  

(3)

This expression is substituted into the unconditional Favre averaged species conservation and after some rearrangement the following expression can be obtained

\[
\frac{\partial Q_a}{\partial t} + (u_i | \eta) \frac{\partial Q_a}{\partial x_i} = \langle N | \eta \rangle \frac{\partial^2 Q_a}{\partial \eta^2} + \langle w_a | \eta \rangle + e_r + e_o
\]

(4)

with

\[
e_o = \left( \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Q_a}{\partial x_i} \right) + \rho D \frac{\partial \xi}{\partial x_i} \left( \frac{\partial}{\partial \eta} \frac{\partial Q_a}{\partial \eta} \right) \right) | \xi[x, t] = \eta
\]

(5)

\[
e_r = -\left( \rho \frac{\partial Y''}{\partial t} + \rho u_i \frac{\partial Y''}{\partial x_i} - \frac{\partial}{\partial x_i} \left( \rho D \frac{\partial Y''}{\partial x_i} \right) \right) | \xi[x, t] = \eta
\]

(6)

The closure of the conditional velocities, \( \langle u, | \eta \rangle \), the conditional scalar dissipation rate, \( \langle N | \eta \rangle \), and the chemical source terms, \( \langle w_a | \eta \rangle \), will be discussed in Section 3.2.2.

With the assumption of high Reynolds number the term \( e_Q \) tends to zero and assuming unity Lewis number for all species and temperature and neglecting fluctuations in density and diffusivity, the following set of governing equation for conditional species mass fraction and conditional temperature are obtained [42]:

\[
\frac{\partial Q_a}{\partial t} + (u_i | \eta) \frac{\partial Q_a}{\partial x_i} - \langle N | \eta \rangle \frac{\partial^2 Q_a}{\partial \eta^2} + \frac{1}{\rho \tilde{P}(\eta)} \frac{\partial}{\partial x_i} \left[ \langle \rho u_i Y'' | \eta \rangle \tilde{P}(\eta) \right] = \langle w_a | \eta \rangle
\]

(7)

\[
\frac{\partial T_a}{\partial t} + (u_i | \eta) \frac{\partial T_a}{\partial x_i} = \langle N | \eta \rangle \frac{\partial^2 T_a}{\partial \eta^2} + \langle N | \eta \rangle \left[ \frac{1}{\langle c_p | \eta \rangle} \left( \frac{\partial \langle c_p | \eta \rangle}{\partial \eta} + \sum_{a=1}^{N} \langle c_{p,a} | \eta \rangle \frac{\partial Q_a}{\partial \eta} \right) \right] \frac{\partial Q_a}{\partial \eta}
\]

\[
- \frac{1}{\rho \tilde{P}(\eta)} \frac{\partial}{\partial x_i} \left[ \langle u_i T'' | \eta \rangle \tilde{P}(\eta) \right] + \frac{1}{\langle c_p | \eta \rangle} \left( \frac{\partial \langle \rho | \eta \rangle}{\partial t} \right) + \langle \rho | \eta \rangle \langle c_p | \eta \rangle + \langle \rho | \eta \rangle \langle c_p | \eta \rangle
\]

(8)

The conditional species equation is similar to the unconditional species equation with from left to right: rate of change, convection, micro-mixing, turbulent fluxes and chemical source. The CMC equations are very similar to the flamelet equations apart from the appearance of transport in physical space.
The energy equation has an analogous form with the species ones despite that it is solved in terms of temperature and therefore the composition heat capacity is employed to link enthalpy to temperature. In the temperature equation the rate of pressure change cannot be neglected for closed geometry with elevated variations in ambient pressure as is the case in diesel engines. The conditional thermal radiation, \( \langle w_{RAD} \mid \eta \rangle \), is considered as a temperature sink. For simulation of diesel engines, where high levels of soot volume fraction (several ppm) are present, an optically thin formulation does not apply and therefore radiation has been neglected.

### 3.2.2 CMC sub-models

The gradient fluxes assumption is used for the closure of conditional turbulent fluxes for any quantity \( \Phi \),

\[
\langle u_j^* \Phi^* \mid \eta \rangle = -D_{ij} \frac{\partial Q_\Phi}{\partial x_j} \tag{9}
\]

where \( D_{ij} \) is the turbulent diffusivity and is expressed with mean flow field quantities as

\[
D_{ij} = \frac{\mu_i}{\text{Sc}_i} \]

The conditional velocity \( \langle u_i \mid \eta \rangle \) was modeled with a linear correlation \([42]\) as

\[
\langle u_i \mid \eta \rangle = \bar{u}_i - \frac{D_{ij}}{\bar{\xi}^2} \frac{\partial \bar{\xi}}{\partial x_j} (\eta - \bar{\xi}) \tag{10}
\]

Conditional density is computed using the ideal gas law as

\[
\langle \rho \mid \eta \rangle = \frac{PW_\eta}{\gamma Q_\eta} \tag{11}
\]

The rate of change of pressure term is modeled assuming constant pressure in mixture fraction space resulting in

\[
\frac{1}{\langle \rho \mid \eta \rangle} \frac{\partial P}{\partial t} = \frac{1}{\langle \rho \mid \eta \rangle} \frac{\partial P}{\partial t} \tag{12}
\]
The closure of the chemical source term is applied at first order, neglecting conditional fluctuations. Conditional mean reaction rate depends only on the mean conditional composition and temperature and is expressed as

\[
\langle w_a | \eta \rangle = \omega_a (Q_a, Q_T, P)
\]

\[
\langle w_r | \eta \rangle = -\sum_{a=1}^{N} h_a \langle w_a | \eta \rangle
\]  

(13)

A first order closure presupposes that fluctuations in reaction rate are mostly due to mixture fraction fluctuations and therefore fluctuations of conditional value are small. For situations where scalar dissipation fluctuations are large, local extinction can occur and therefore for the same mixture fraction a considerable scattering of the temperature and composition is observed. For those cases a second order closure has been proposed [143] and generally slightly improved predictions have been achieved for e.g. non-premixed flames [144] and auto-igniting \( n \)-heptane plume in a heated co-flow air [121]. However, second order CMC increases the computational cost considerably and it is common practice to employ first order CMC.

The conditional scalar dissipation rate is responsible for the micro-mixing and is modeled with the amplitude mapping closure (AMC) [145].

\[
\langle N | \eta \rangle = N_0 G(\eta)
\]

\[
G(\eta) = \exp \left( -2 \left[ \text{erf}^{-1} (2\eta - 1) \right]^2 \right)
\]

\[
N_0 = \frac{\tilde{\chi}}{\int_0^1 G(\eta) \tilde{P}(\eta) d\eta}
\]

(14)

The function \( G(\eta) \) exhibits a bell-shaped profile with a maximal value of 1 at \( \eta = 0.5 \). The amplitude of the conditional scalar dissipation rate is scaled with the mean scalar dissipation rate \( \tilde{\chi} \), which is modelled using mean turbulence quantities as:

\[
\tilde{\chi} = c_x \frac{\tilde{\varepsilon}}{k} \xi^{\sigma^2}
\]

(15)
$c_\chi$ is a model constant set to 2.0 as was the case in previous CMC spray studies, e.g. [134].

3.2.3 Soot model

The two-equation semi-empirical soot model by Leung et al. [80] has been adopted. The focus in this study was not to develop a new soot model, but to assess the suitability of the CMC framework for modelling soot over a wide range of conditions representative of diesel engine combustion. Transport equations for conditional soot mass fraction $\langle Y_S | \eta \rangle$ and conditional soot number density $\langle N_S | \eta \rangle$ are solved in the same way as for reactive species including terms accounting for convection and diffusion in space as well as molecular mixing. Moreover, the soot chemical source term, $\langle w_S | \eta \rangle$, includes soot sub-processes for the conditional soot mass fraction and particle number density as follows:

\begin{align}
\langle w_{Y_S} | \eta \rangle &= \langle w_{Y_S, \text{inc}} | \eta \rangle + \langle w_{Y_S, \text{gro}} | \eta \rangle + \langle w_{Y_S, \text{oxidO}} | \eta \rangle + \langle w_{Y_S, \text{oxidOH}} | \eta \rangle \\
\langle w_{N_S} | \eta \rangle &= \langle w_{N_S, \text{inc}} | \eta \rangle + \langle w_{N_S, \text{coag}} | \eta \rangle
\end{align}

where terms accounting for simultaneous soot inception, surface growth, oxidation by O$_2$ and OH, and particle coagulation are considered. The individual chemical reactions and the corresponding rates are summarized in Table 3-1 and Table 3-2, respectively.

As soot is a solid, high-density material it is subjected to a low mass diffusivity resulting in a high Lewis number. In the literature various models have been proposed for differential diffusion for low pressure gaseous flames, e.g. [100-102], however under diesel engine conditions no closure has been validated yet. Therefore, in this study, unity Lewis number for soot has been assumed.

Thermophoretic forces have also been neglected following conclusions reached in [78] where DNS was used to study non-premixed atmospheric ethylene jet flames and it was concluded that soot is essentially convected with the flow field. In this study, due to the very high injection velocity (in the order of 500 m/s) the relative importance of thermophoretic diffusion velocity is expected to decrease. More complete models should however assess the influence of both these effects in the future.
Particle inception (reaction I) and surface growth rate (reaction II) were both assumed to be first order function of acetylene concentration only. Production and consumption of chemical species present in the soot model (C\textsubscript{2}H\textsubscript{2}, O\textsubscript{2}, OH, H, H\textsubscript{2} and CO) has been considered. A primary particle consists of 100 C-atoms [80]. The soot surface growth rate is proportional to the square root of the specific surface area, \( S_{\text{Soot}} \) (in m\textsuperscript{2}/m\textsuperscript{3}), which is introduced as:

\[
S_{\text{Soot}} = \pi d_p^2 \rho S = \pi \left( \frac{6 Y_S}{\pi \rho_S N_S} \right)^{2/3} \quad (18)
\]

where \( d_p \) is the diameter of the particles which are assumed to be spherical and mono-dispersed. The soot density, \( \rho_S \), was set to 2000 kg/m\textsuperscript{3} according to Leung et al. [80].

### Table 3-1: Soot chemistry mechanism [80].

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Reaction rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>(I) ( C_2H_2 \xrightarrow{R_{\text{inc}}} 2C_{(s)} + H_2 )</td>
<td>( R_{\text{inc}} = k_{\text{inc}}[C_2H_2] )</td>
</tr>
<tr>
<td>(II) ( C_2H_2 + nC_{(s)} \xrightarrow{R_{\text{gro}}} (n + 2)C_{(s)} + H_2 )</td>
<td>( R_{\text{gro}} = k_{\text{gro}}[C_2H_2] )</td>
</tr>
<tr>
<td>(III) ( C_{(s)} + 0.5O_2 \xrightarrow{R_{\text{oxidO}_2}} CO )</td>
<td>( R_{O_2} = k_{\text{oxidO}_2}[O_2] )</td>
</tr>
<tr>
<td>(IV) ( C_{(s)} + OH \xrightarrow{R_{\text{oxidOH}}} CO + H )</td>
<td>( R_{\text{oxidOH}} = k_{\text{oxidOH}}[OH] )</td>
</tr>
</tbody>
</table>

### Table 3-2: Soot formation and oxidation reaction rate constants in Arrhenius form, \( k_j = A \cdot T^b \cdot \exp(-T_a/T) \cdot S_{\text{Soot}}^c \). Units are in kg, kmol, m, s, K. From Leung et al. [80].

<table>
<thead>
<tr>
<th>( k_j )</th>
<th>A</th>
<th>b</th>
<th>( T_a )</th>
<th>c</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(I) Inception ( k_{\text{inc}} )</td>
<td>1.0e4</td>
<td>0</td>
<td>21100</td>
<td>0</td>
<td>[80]</td>
</tr>
<tr>
<td>(II) Surface growth ( k_{\text{gro}} )</td>
<td>6.0e3</td>
<td>0</td>
<td>12100</td>
<td>0.5</td>
<td>[80]</td>
</tr>
<tr>
<td>(III) Oxidation by ( k_{\text{oxidO}_2} )</td>
<td>1.0e4</td>
<td>0.5</td>
<td>19800</td>
<td>1</td>
<td>[80]</td>
</tr>
<tr>
<td>(IV) Oxidation by ( k_{\text{oxidOH}} )</td>
<td>0.36</td>
<td>0.5</td>
<td>0</td>
<td>1</td>
<td>[103]</td>
</tr>
</tbody>
</table>
Particle coagulation is the unique process which acts as a sink for the particle number and it is denoted as:

$$nC_{(s)} \rightarrow C_{n(s)}$$  \hspace{0.5cm} (19)

Following Leung et al. [80], the coagulation rate for a spherical and mono-dispersed distribution is expressed as follows:

$$\langle w_{N_s,\text{coagulation}} | \eta \rangle = -c_a \left( \frac{24R}{\rho_s N_A} \right)^{\frac{1}{2}} \left( \frac{6}{\pi \rho_s} \right)^{\frac{1}{6}} T^{\frac{1}{2}} \left( \rho \langle Y_s | \eta \rangle \right)^{\frac{1}{6}} \langle N_s | \eta \rangle^{\frac{11}{6}}$$  \hspace{0.5cm} (20)

where $N_A$ is the Avogadro number, $R$ is the universal gas constant and $C_a$ is a model constant set to 3 according to Ma et al. [146]. Particle agglomeration is not considered and the computed particle number has the meaning of a mean primary particle size.

Soot oxidation by molecular oxygen and OH (reaction III and IV Table 3-1 in Table 3-2 and respectively) acts at the particle surface and therefore oxidation reduces the soot mass, but not the particle number.

Soot radiation has been considered with an optically thin assumption, where radiation is considered for major gaseous species (CO$_2$, CO, H$_2$O and CH$_4$) according to [147] and soot with absorptivity properties reported in [148]. The conditional radiation heat transfer $\langle w_{\text{RAD}} | \eta \rangle$ follows

$$\langle w_{\text{RAD}} | \eta \rangle = -4 \sigma \alpha_{\text{soot}} \left[ \langle T | \eta \rangle^4 - T_{\text{Wall}}^4 \right]$$  \hspace{0.5cm} (21)

where $\sigma$ is the Stefan-Boltzmann constant. $\alpha_{\text{soot}}$ is the soot particle mean absorptivity coefficient assumed to be $\alpha_{\text{soot}} = 2370 \frac{1}{m K} \cdot f_{\text{soot}} \cdot T$ [148].

$T_{\text{Wall}}$ is the wall temperature and it was set to 450 K. The influence of this term is not large due to the fourth power dependency of the radiative power on temperature. A more complex radiation model, e.g. discrete ordinate method [149], has not been attempted here. For the heavy-duty diesel engine simulation presented in Section 6 soot radiation has been deliberately neglected as due to the high levels of soot an optical thin formulation does not apply.
3.2.4 CMC solver and boundary conditions

The CMC equations, were discretized using finite differences. Diffusive terms are solved using second-order central differences and convective terms with a first-order upwind scheme. Conserved scalar space is discretized in 101 nodes which were clustered around the stoichiometric mixture fraction $\eta_{st}$. Physical space was divided with different resolution for the three test facilities and details about the geometry and CFD as well as CMC grids are provided in the respective results sections (cf. sections 4.2, 5.2 and 6.2). In general a constant CMC grid resolution of 1 by 2 mm, in radial and axial directions respectively, was employed. To assess the sensitivity of the results, on the one hand for the Sandia constant-volume configuration a TVD scheme has been tested in lieu of up-wind and the impact of the axial resolution has further been assessed (27 and 108 nodes instead of 54), resulting in less than 1 percent difference in the total soot mass.

The CMC equations were integrated with the stiff integrator VODPK [150]; in order to reduce the number of ODEs which have to be solved simultaneously, a full operator splitting of transport in physical and in $\eta$-space as well chemistry was used as described in Wright et al. [134] and De Paola et al. [137]. The CFD temporal integration is performed with the standard implicit Euler formulation and 5 internal CMC time steps are employed per CFD time step, here set equal to $1 \times 10^{-6}$ s. Conditional species mass fractions were initialized assuming adiabatic frozen mixing in which species mass fractions are linearly distributed in mixture fraction between their values at the oxidizer and fuel side. The temperature profile is computed based on a linear distribution of mixture enthalpy between oxidizer and fuel values.

3.3 CFD related governing equations

3.3.1 Gas phase

The RANS formulation consists on time or ensemble averaging of the instantaneous transport equations for mass momentum and species. A generic instantaneous property is split into an ensemble average and a fluctuation and is referred to Reynolds averaging. For variable density flows a density weighted averaging, denoted as Favre averaging, is commonly applied.

Conservation equations for mass, momentum and energy are written as
Methodology

\[
\frac{\partial \bar{p}}{\partial t} + \frac{\partial}{\partial x_j}(\bar{\rho} u_j \bar{v}) = S_m \quad (22)
\]

\[
\frac{\partial (\bar{p} \bar{v})}{\partial t} + \nabla \cdot (\bar{p} \bar{v} \bar{v}) = -\nabla \bar{p} + \nabla \bar{v} + \bar{p} g - \nabla \cdot (\bar{p} \bar{v}^* \bar{v}^*) \quad (23)
\]

\[
\frac{\partial \bar{p} \tilde{h}}{\partial t} + \nabla \cdot (\bar{p} \tilde{h} \tilde{h}) = -\nabla \cdot (\bar{p} \tilde{v}^* \tilde{v}^*) + \frac{\partial \bar{p}}{\partial t} + \tilde{v} \cdot \nabla p + \tau : \tilde{v} \tilde{v} + \nabla \cdot \tilde{J} + \bar{q} \quad (24)
\]

Following the Boussinesq assumption for turbulence viscosity, the Reynolds stresses \( \bar{p} \tilde{v}^* \tilde{v}^* \) are described using the viscous tensor \( \tau \) considering a Newtonian fluid

\[
\bar{p} \tilde{v}^* \tilde{v}^* - \frac{2}{3} \bar{p} k I = -\mu_t \left[ 2 \tilde{S} - \frac{2}{3} \nabla \cdot \tilde{v} I \right] \quad (25)
\]

Where \( \tilde{S} \) is the stain tensor, defined as

\[
\tilde{S} = \frac{1}{2} \left( \nabla \tilde{v} + \nabla \tilde{v}^* \right) \quad (26)
\]

The turbulent viscosity \( \mu_t \) is estimated as

\[
\mu_t = \bar{p} C_\mu \frac{\tilde{k}^2}{\tilde{\varepsilon}} \quad (27)
\]

The RNG (Renormalization Group) variant of the \( k-\varepsilon \) model [151] is used here for the calculation of the Favre averaged turbulent kinetic energy \( \tilde{k} \) and its dissipation rate \( \tilde{\varepsilon} \) for which two additional transport equations are solved as

\[
\frac{\partial (\bar{p} \bar{k})}{\partial t} + \nabla \cdot (\bar{p} \bar{v} \bar{k}) - \nabla \cdot \left( \bar{p} \frac{V_T}{Sc_{\tilde{\varepsilon}}} \nabla \bar{k} \right) = -\left( \tilde{\tau} : \nabla \tilde{v} \right) - \bar{p} \bar{\varepsilon} \quad (28)
\]

\[
\frac{\partial (\bar{p} \bar{\varepsilon})}{\partial t} + \nabla \cdot (\bar{p} \bar{v} \bar{\varepsilon}) - \nabla \cdot \left( \bar{p} \frac{V_T}{Sc_{\tilde{\varepsilon}}} \nabla \bar{\varepsilon} \right) = -c_{e_k} \bar{p} \bar{\varepsilon} \left( \tilde{\tau} : \nabla \tilde{v} \right) - c_{e_k} \bar{p} \frac{\tilde{\varepsilon}^2}{\tilde{k}} - c_{e_k} \bar{p} \bar{\varepsilon} \nabla \tilde{v} - R \quad (29)
\]
with an additional term, R, representing the effect of mean flow distortion on the
dissipation rate

\[
R = \frac{C_\mu \tilde{\eta}^3 \left(1 - \frac{\tilde{\eta}}{\eta_0}\right)}{1 + \beta \tilde{\eta}^3} \frac{\bar{\rho} \tilde{\varepsilon}^2}{k}
\]  

(30)

where

\[
\tilde{\eta} = S \frac{k}{\bar{\varepsilon}}
\]  

(31)

The distribution of velocity, temperature, turbulence quantities within the
boundary layer is described by standard wall function [152].

The unconditional species mass fractions in the CFD code are computed by
special-purpose subroutines by convoluting the conditional averages with the
mixture fraction PDF; the latter is presumed to be a beta function. Therefore, in
the CFD code no transport equations for species are required.

Furthermore, in order to reconstruct the mixture fraction PDF, transport
equations for the mean mixture fraction and its variance were solved as:

\[
\frac{\partial \bar{\rho} \tilde{\xi}}{\partial t} + \nabla \left[ \bar{\rho} \tilde{u}_j \tilde{\xi} - \left( \bar{\rho} D_{\tilde{\xi}} + \frac{\mu_t}{Sc_{\tilde{\xi}}} \right) \nabla \tilde{\xi} \right] = \dot{S}_d
\]

(32)

\[
\frac{\partial \bar{\rho} \tilde{\xi}^{*2}}{\partial t} + \nabla \left[ \bar{\rho} \tilde{u}_j \tilde{\xi}^{*2} - \left( \bar{\rho} D_{\tilde{\xi}^{*2}} + \frac{\mu_t}{Sc_{\tilde{\xi}^{*2}}} \right) \nabla \tilde{\xi}^{*2} \right] = \frac{2 \mu_t}{Sc_{\tilde{\xi}^{*2}}} \left( \nabla \tilde{\xi}^{*2} \right)^2 - \bar{\rho} \tilde{\xi}^{*2}
\]

(33)

where \( \dot{S}_d \) is the mean mixture fraction source term arising from droplet
evaporation. In the mixture fraction variance equation, the influence of
evaporating droplet on the variance has been neglected, since the influence of
this term was investigated by Borghesi et al. [132] for the same experimental
setup as in this study and a marginal influence on the spray ignition delay time
and lift-off height was observed.
3.3.2 CFD-CMC interfacing

The CMC code is full two-way coupled with the flow field solver Star-CD [152]. In Figure 3-1 a schematic of the solution procedure is illustrated. The CFD solver provides the flow field solution until start of injection. When the first injected fuel enters the domain, the CMC solution is initialized according to adiabatic mixing as described in section 3.2.4. The flow field provides parameters for the CMC solution at every time step, these are mainly scalar dissipation rate, distribution of mean mixture fraction and its variance, and pressure. CMC advances the solution of the governing equations for conditional species and conditional temperature at the CMC resolution using a fractional steps technique with five to ten sub-steps. The mean composition at the CFD resolution is retrieved by convoluting the conditional composition with the presumed beta-function for the given CFD cell. The mean composition is then returned to the flow field solver for every CFD cell.

3.3.3 Spray model

The evolution of the dispersed liquid phase is described in a Lagrangian-Eulerian framework where conservation equations for mass, momentum and energy of the liquid fuel droplets are solved in a Lagrangian manner and
exchange of mass, momentum and energy between phases are coupled with the
gas phase balance equations. As the spray consists of several millions of droplets,
modeled droplets have been collected into so-called droplet parcels, each
consisting of a number of droplets with the same properties, i.e. diameter,
temperature. Droplets are assumed to be spherical and the temperature is
considered to be homogeneous in the entire droplet volume.

**Momentum**

In the simulation, typically few 10,000 fuel parcels are singularly tracked
according to

\[
\frac{d\mathbf{x}_d}{dt} = \mathbf{u}_d
\]  

(34)

The evolution of the droplet velocity \(\mathbf{u}_d\) is retrieved using the droplet momentum
equation as

\[
m_d \frac{d\mathbf{u}_d}{dt} = F_{dr} + F_p + F_b
\]  

(35)

The forces acting to the droplet are the drag, pressure and body forces, \(F_{dr}\), \(F_p\)
and \(F_b\), respectively. A spherical droplet travelling in the surrounding gas field
experiences a drag force, which is given by

\[
\mathbf{F}_{\text{drag}} = \frac{1}{2} C_d \rho A_d |\mathbf{v} - \mathbf{v}_d| (\mathbf{v} - \mathbf{v}_d)
\]  

(36)

the drag coefficient, \(C_d\), is computed using the standard correlation from [152]

\[
C_d = \begin{cases} 
\frac{24}{\operatorname{Re}_d} \left( 1 + 0.15 \operatorname{Re}_d^{0.687} \right) , & \operatorname{Re}_d \leq 10^3 \\
0.44 , & \operatorname{Re}_d > 10^3 
\end{cases} 
\]  

(37)

where two regimes depending from the droplet Reynolds number, \(\operatorname{Re}_d\), are
distinguished. The latter is defined as
The effect of turbulent dispersion has been included using the stochastic model. A droplet in a turbulent flow experiences local velocities fluctuations due to turbulence that are not included in the RANS velocity field. Therefore, the mean velocity has been augmented by a random velocity fluctuation. The latter is assumed to be isotropic and to obey a Gaussian PDF with zero mean and a standard deviation of $(2k/3)^{1/3}$ [152].

**Mass**

The liquid mass rate of change is driven by droplet evaporation and condensation as follows

$$\frac{dm_d}{dt} = -A_d F_m$$ (39)

where $A_d$ is the droplet surface area, and the droplet evaporation rate, $F_m$, is calculated from

$$F_m = K_g \ln \left( \frac{p_t - p_{v,\infty}}{p_t - p_{v,s}} \right)$$ (40)

with $p_t$, $p_{v,s}$, and $p_{v,\infty}$ are the total gas pressure and vapor pressure at the droplet surface and at its surrounding, respectively.

The mass transfer coefficient, $K_g$, is derived from the following relationship

$$K_g = \frac{Sh D_m}{R_m T_m D_d}$$ (41)

in which the Sherwood number, $Sh$, is a function of the droplet Reynolds number, $Re_d$, and the Schmidt number, $Sc$ following [153]
\[ Sh = 2 \left( 1 + 0.3 \, \text{Re}_d^{1/2} \, \text{Sc}^{1/3} \right) \]  \hspace{1cm} (42)

and for the mean film temperature, \( T_m \), the 1/3 rule is applied as

\[ T_m = \frac{2T_d + T_g}{3} \]  \hspace{1cm} (43)

The mass relaxation time \( \tau_m \) can be expressed as

\[ \tau_m = \frac{m_f}{A_d |F_m|} = \frac{\rho_d D_d}{6 |F_m|} \]  \hspace{1cm} (44)

**Energy**

The droplet energy balance correlates the change of the droplet internal energy with the surface heat transfer from the gas phase the evaporation as

\[ m_d c_{p,d} \frac{dT_d}{dt} = -A_d q_d^* + h_{fg} \frac{dm_d}{dt} \]  \hspace{1cm} (45)

where \( c_{p,d} \) is the droplet specific heat capacity, \( h_{fg} \) is the fuel latent heat at the temperature \( T_d \) and the surface heat transfer between the two phases, \( q_d^* \), is proportional to their temperature difference according to

\[ q_d^* = h \left( T_d - T_g \right) \]  \hspace{1cm} (46)

where the heat transfer coefficient between the phases, \( h \), is calculated from

\[ h = \frac{k_g \, NuZ}{(e^Z - 1) D_d} \]  \hspace{1cm} (47)

in which the Nusselt number, \( Nu \), is expressed as [153]
\[ Nu = 2 \left( 1 + 0.3 \text{Re}^{1/2} \text{Pr}^{1/3} \right) \]  \hfill (48)

where Pr is the Prandl number and \( Z \) is derived from

\[ Z = \frac{-c_p \left( \frac{dm_d}{dt} \right)}{\pi D_d k_g Nu} \]  \hfill (49)

\( k_g \) is the gas phase conductivity.

Properties of fuel parcels needed for the description of the aforementioned processes are evaluated as a function of temperature, these are: density, latent heat, specific heat capacity, viscosity, conductivity, surface tension and saturation pressure.

The atomization has been modeled with the so-called ‘blob’ model \[154\], where large droplets with the diameter of the orifice are injected and the spray cone angle has to be prescribed. Information about the internal nozzle dynamics are not considered here. Droplets are injected with equal probability of velocity direction within the spray cone. The measured temporal rate of injection has been employed as a boundary condition for the simulation. Alternatively, for the marine spray combustion chamber the rate of injection has been estimated from the measured pressure drop across the injector body, since no direct measurements of rate of fuel injected were available.

The secondary break-up is treated with the model by Reitz-Diwakar \[154\] which distinguishes two modes of break-up: the bag and the stripping break-up.

The first is caused by non-uniform pressure field around the droplet, which yields to a disintegration of the droplet when surface tension forces are overcome. The second is caused by tangential viscous forces that provoke Kelvin-Helmholtz instabilities leading to a stripping of droplets from the surface.

For every droplet parcel, the droplet break-up rate is expressed as a relaxation equation for the instantaneous droplet diameter, \( D_d \), towards a stable diameter, \( D_{d,\text{stable}} \), following

\[ \frac{dD_d}{dt} = \frac{D_d - D_{d,\text{stable}}}{\tau_{bu}} \]  \hfill (50)
Where $\tau_{bu}$ is the characteristic break-up time. Note that break-up reduces the droplet diameter but not the mass, i.e. the number of droplets contained in a parcel is therefore increased to fulfill this requirement.

The criterion for the occurrence of bag break-up is based on the droplet Weber number, $We$

\[
We = \frac{\rho_d |v_d - v|^2 D_d}{2 \sigma_d} \geq C_{b1}
\]

(51)

which is defined as the ratio between aerodynamic forces and the tension of the droplet. Bag break-up takes place when $We$ exceeds the critical value $C_{b1}$. The default value of the model constant $C_{b1}=6$ has been used.

The corresponding characteristic time is calculated as

\[
\tau_{bu} = \frac{C_{b1} \rho_d^{1/2} D_d^{3/2}}{4 \sigma_d^{1/2}}
\]

(52)

The stable diameter after break-up is computed as the diameter when $We=C_{b1}$.

At higher $We$ numbers the stripping break-up occurs when the following criterion is satisfied

\[
\frac{We}{\sqrt{Re_d}} \geq C_{s1}
\]

(53)

where $C_{s1}$ is a model constant and was set to its default value of 0.5. The stripping break-up time is modeled as

\[
\tau_{bu} = \frac{C_{s1}}{2} \left( \frac{\rho_d}{\rho} \right)^{1/2} \frac{D_d}{2 |u-u_d|}
\]

(54)

In diesel sprays, in particular for larger orifices, the vast majority of the droplets are in the stripping regime due to the high values of relative velocities. As a
consequence, the spray was observed to be more sensitive with respect to model constants for the stripping regime.

The inter-droplet collision is based on the work by [155], where three principal collision modes are considered: coalescence, separation and bouncing. The model describes statistically the collision of the uniform assumed droplets within the cell and depending on the regime the new number of droplets is recomputed. Droplet wall impingement is described by the model of Bai and Gossman [156] accounting for four types of regimes: droplet stick, spread, rebound and plashing. For test cases considered in this study, practically no droplet reaches the wall. The model has been considered for completeness.
4 Results I: Sandia constant-volume chamber

4.1 Experimental test facility

Experimental data available from the optically accessible constant-volume combustion vessel installed at the Sandia National Laboratories simulating quiescent diesel engine conditions have been considered for the first validation of the numerical model. A schematic of the vessel is given in Figure 4-1 along with optical diagnostic set-ups. The combustion chamber has a cubic shape with 108 mm side length with optical access provided by side-port quartz windows. The common-rail fuel injector is mounted at the centre of a metal side-port and the spray is directed into the centre of the chamber.

Figure 4-1: Schematic of the combustion vessel and optical setup for soot measurements (adopted from [75]).
Results I: Sandia constant-volume chamber

For a detailed description of the experimental setup and the measurement techniques the reader is referred to \[29, 75\] and references therein. A broad range of experimental conditions have been measured, including variations in fuel, fuel pressure, nozzle orifice diameter, temperature, density, and oxygen content of the oxidizer.

In this study, simulations only with \(n\)-heptane fuel with a single-hole co-axial injector with an orifice diameter of 0.1 mm have been carried out. In total 13 different test cases are considered as summarized in Table 4-1.

<table>
<thead>
<tr>
<th>Case</th>
<th>Gas temperature (K)</th>
<th>Gas density (kg/m(^3))</th>
<th>(O_2) (% by vol.)</th>
<th>Injection duration (ms)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>1000</td>
<td>14.8</td>
<td>21</td>
<td>6</td>
</tr>
<tr>
<td>A2</td>
<td>1000</td>
<td>14.8</td>
<td>15</td>
<td>6</td>
</tr>
<tr>
<td>A3</td>
<td>1000</td>
<td>14.8</td>
<td>12</td>
<td>6</td>
</tr>
<tr>
<td>A4</td>
<td>1000</td>
<td>14.8</td>
<td>10</td>
<td>6</td>
</tr>
<tr>
<td>A5</td>
<td>1000</td>
<td>14.8</td>
<td>8</td>
<td>6</td>
</tr>
<tr>
<td>B2</td>
<td>1000</td>
<td>30</td>
<td>15</td>
<td>6</td>
</tr>
<tr>
<td>B3</td>
<td>1000</td>
<td>30</td>
<td>12</td>
<td>6</td>
</tr>
<tr>
<td>B4</td>
<td>1000</td>
<td>30</td>
<td>10</td>
<td>6</td>
</tr>
<tr>
<td>B5</td>
<td>1000</td>
<td>30</td>
<td>8</td>
<td>6</td>
</tr>
<tr>
<td>C1</td>
<td>900</td>
<td>14.8</td>
<td>21</td>
<td>6</td>
</tr>
<tr>
<td>C2</td>
<td>900</td>
<td>14.8</td>
<td>15</td>
<td>6</td>
</tr>
<tr>
<td>C4</td>
<td>900</td>
<td>14.8</td>
<td>10</td>
<td>6</td>
</tr>
<tr>
<td>S3</td>
<td>1000</td>
<td>14.8</td>
<td>12</td>
<td>1.8</td>
</tr>
</tbody>
</table>

The nine cases where soot distribution is reported are the A and B series, all with 1000 K ambient temperature with oxygen sweeps at both low and high ambient densities. An additional oxidizer variation is considered at 900 K for the low density cases. The last operating configuration considered (S3) employs, contrary to the rest, substantially shorter injection duration with 1.8 ms against 6 ms. This makes it ideally suited for studies of the spray jet transient after end of injection, including air entrainment within the experimental measurement domain.
Definitions of criteria for model comparison with experiment

The measurement of ID is based on the pressure with a correction for the speed of sound, whereas LOL is determined by means of OH\* chemiluminescence, and soot volume fraction under quasi-steady conditions is measured with planar laser induced incandescence (PLII) with a calibration against laser extinction measurements. Simulated ID and LOL were both defined according to the ECN recommendations [29]: ID was defined as the time after SOI when the maximal rate of change of peak temperature within the domain occurs, while the flame LOL has been determined as the minimal axial distance from the injector tip where a threshold value of two percent of the peak OH mass fraction is present.
4.2 Numerical considerations

As a symmetric spray arrangement is considered, a quasi-2D CFD grid with one degree angle was used with a homogeneous resolution of 0.5 x 0.5 mm\(^2\) size in the first 15 mm radially along the entire axial domain, while in the outer part a constant cell size of 1 x 1 mm\(^2\) is employed. This mesh resolution is in agreement with previous studies on grid sensitivity at the same experimental setup, i.e. [48, 157] and is in line with ECN recommendations [29]. The width of the domain has been adjusted in order to match the chamber volume.

Physical space was divided into 20 by 54 CMC nodes in radial and axial direction, respectively. For the region with radius up to 15 mm from the injector axis, a constant CMC grid resolution of 1 by 2 mm, in radial and axial directions respectively, was employed.

The thermo-physical properties of \(n\)-heptane droplets (density, viscosity, latent heat, heat capacity, surface tension and saturation pressure) are computed as a function of temperature. The measured injection rate is imposed for the simulation. Heat transfer at the heated walls has been neglected.

![Figure 4-2](image)

**Figure 4-2:** Two-dimensional axis-symmetric CFD grid of the Sandia constant volume chamber. Grid resolution is 0.5 x 0.5 mm\(^2\) in the first 15 mm radially from the injector axis.
4.3 Validation of soot model for steady n-heptane sprays

4.3.1 Ignition delay and lift-off length

Figure 4-3 compares ID time (left) and flame LOL (right) for the CMC model (red lines) against experimental data (squares) for ambient oxygen variations at three different ambient conditions: at 1000 K with 14.8 kg/m³ (upper) and 30 kg/m³ (middle) and at 900 K with 14.8 kg/m³ (lower). Note that results obtained with the DI method (blue lines) have been already integrated in Figure 4-3 but results are going to be discussed in Section 4.3.1 in the context of the influence of the combustion model.

In general, the impact of the oxidizer dilution on the ID is well reproduced. The chemical mechanism employed tends to overestimate ID for lower oxidizer reactivities, i.e. for low ambient oxygen contents, temperatures and densities. At the higher ambient density the chemical model showed very good agreement. This trend is consistent with former studies employing the same chemical mechanism, i.e. [25, 26, 132, 158]. The accuracy of various n-heptane chemical mechanisms has been compared in the framework of ECN2 workshop [29]. The chemical mechanism employed in the present work [15] was found to perform similarly compared to more complex mechanisms.

Once ignition occurs, the flame stabilizes upstream until a quasi-steady location is reached, the so-called lift-off length. As illustrated in Figure 4-3, the CMC model captures the correct trend with an increase in LOL by diminishing oxygen content as well as the influence of ambient gas density. For the lower ambient temperature (900 K) LOL is slightly underestimated for the low oxygen cases. Overall, relative errors at 1000 K were within 24.4 and 21.6 percent of the experimental values for the low and high pressures, respectively and within 17 percent at 900 K. Time evolutions of the LOL using the same modeling approach have been previously shown by Borghesi et al. [132] and a flame stabilization due to a convective-reactive balance at the anchoring point of the flame base was observed for all ambient oxygen considered. Since in this study the same balance of terms in the temperature equation is found at the LOL, these are not repeated here.
Results I: Sandia constant-volume chamber

Figure 4-3: Ignition delay (left) and flame lift-off length (right) as a function of oxygen content for different ambient densities and ambient temperatures.

(a) 14.8 kg/m$^3$ and 1000 K

(b) 30 kg/m$^3$ and 1000 K

(c) 14.8 kg/m$^3$ and 900 K
4.3.2 Flame structure

4.3.2.1 Unconditional averages
Due to the long injection duration of 6 ms, the spray reaches a quasi-steady behaviour for the LOL and for the soot distribution until end of injection. In order to investigate the soot distribution in physical space, it is important to examine the location of the stoichiometric mixture fraction during this period because soot is mainly present in the fuel rich zone. In the following section the influence of the ambient oxygen concentration on the flame structure is studied. Quantities present in the soot model (temperature, mass fraction of C₂H₂, O₂ and OH) at the 14.8 kg/m³ gas density for two different ambient oxygen mole fractions (21 % and 10 % O₂) during the quasi-steady period are considered.

Case A1 with 21 % O₂ is used as the conventional diesel engine condition without EGR and the case A4 with 10 % O₂ reproduces high EGR-rate conditions as shown in Figure 4-4.

First of all, the stoichiometric mixture fraction of case A4 is roughly half of that of case A1 due to the different ambient composition (0.0304 vs. \( \xi_{ST} = 0.061 \), respectively). As a consequence, the extent of the stoichiometry region in physical space is larger in both axial and radial directions and a considerably broader flame can be observed. As expected, the lower amount of oxygen affects the flame with a considerable decrease in temperature. For both cases, the OH mass fraction nicely follows the stoichiometric isocontour until the flame base. Ignition takes place downstream of the LOL and the reaction zone travels back along the stoichiometric contour, as was also observed by Borghesi et al. [132].

The acetylene distribution is, as expected, in the fuel rich zone as a marker of where soot is conceptually assumed to be formed [159]. For the 10 % O₂ case, the mass fractions of OH and C₂H₂ are roughly a factor of 15 and 3 lower compared to the 21 % O₂ case, respectively. Therefore the rates of all soot processes, i.e. soot formation and oxidation, are expected to slow down. This fact makes it difficult to predict intuitively the change in the net soot amount.

Simulations showed that an ambient pressure increase does not significantly change the mean flame temperature (not shown here). With a higher gas density, the spray penetration length becomes lower due to the increased aerodynamic forces acting on the fuel droplets. For the 10 % O₂ case, an increase of approximately 50 % in acetylene mass fraction is observed (not shown here).
Figure 4-4: Spatial distribution of relevant quantities for flame characterization at 5 ms after SOI for case A4 (a), upper) and case A1 (b), lower). From left to right: mean mixture fraction, temperature, mass fractions of C2H2, O2, OH and CH2O, respectively. The black line denotes the location of the stoichiometric mixture fraction $\xi_{ST}$. The locations A-E indicated in (b) are discussed in Figure 4-5.
Higher gas density corresponds to higher molar concentrations of all reactants and therefore an increased reaction rate. In this case, enhanced soot formation rate is expected and therefore more soot is formed as will be shown in the next section.

4.3.2.2 Conditional averages

An analysis of the conditional flame structure is carried out for the A1 case. The conditional temperature and acetylene reach a quasi-steady distribution in the fuel rich region approximately 3 ms after SOI (not shown here). Figure 4-5 shows the mixture fraction PDF and the conditional quantities responsible for soot processes at locations (A-E) illustrated in Figure 4-4 (b) at 5 ms after SOI. Profiles in mixture fraction space confirm a quasi-developed flamelet structure at the various locations, where the oxygen has been consumed and the temperature profile approaches a fully developed diffusion flame peaking at stoichiometry as well as OH profiles, which were almost identical at all selected locations. As can be seen in the profiles of the soot precursor acetylene, non-zero values are present only in the fuel rich zone with a maximum at an equivalence ratio of around 2.5. Profiles were roughly the same for all locations selected except at 30 mm axial distance from the injector where acetylene is not yet formed completely. The mixture fraction PDF shows a decrease in mean value and variance with increasing axial and radial distance from the injector due to conserved scalar mixing with the ambient field.
Figure 4-5: Mixture fraction PDF (a), conditional temperature (b), conditional species mass fractions of C$_2$H$_2$ (c), oxygen (d) and OH (e) for case A1 at 5 ms after SOI at the selected locations indicated in Figure 4-4 (A-E, mixture fraction for case (b), lower). The dashed vertical lines denote the stoichiometric mixture fraction.
4.3.3 Soot formation

4.3.3.1 Soot reference case A1
The A1 case was chosen for as reference case because the phenomenological soot model used was developed and validated at this oxygen mole fraction [80]; although, the ambient pressure was 1 bar compared to the approximately 42 bar of the A1 reference case of this study. Another motivation for the choice of the A1 case as a reference is the fact that during the quasi-steady period soot is entirely visible within the experimental observation domain. Soot model constants shown in Table 3-2 are used for all computations presented. Assumption of a first order function of acetylene concentration for particle inception rate and surface growth rate is widely used in the literature [71].

In Figure 4-6, a comparison between simulations and experiments of the soot volume fraction is shown. Soot, as expected, is present in the fuel rich zone and the predicted location of the soot region agrees well with the experiment. Peak soot load was over-predicted and roughly double compared to the measurement. Simulated soot distribution shows a quasi-steady behavior between 3 and 6 ms in agreement with the experiment.

Figure 4-6: Comparison of soot volume fraction (in ppmv) distribution for the case A1. Simulation left and experiment (right).
4.3.3.2 Soot model sensitivity analysis

In the following, the sensitivity of the phenomenological soot model has been analyzed for the reference case A1 with respect to various soot model constants. The pre-exponential factor for inception rate, surface growth rate, oxidation rate by OH and O₂, the constant for coagulation rate \( c_c \) and the number of carbon atoms of a primary soot particle have been varied by a factor of two, one at a time.

The soot concentration peak value using the reference model constant setting was over-predicted (cf. Figure 4-6); therefore a reduced formation rate and an enhanced oxidation rate were examined. The soot volume fraction along the spray axis and the radial distribution at the axial location of peak soot load \((Z=60 \text{ mm})\) have been considered in order to quantify the sensitivity of the various soot model parameters, for which results are shown in Figure 4-7. The soot model is most sensitive with respect to the surface growth rate constant, where with half of the reference value the peak soot volume fraction decreases almost by a factor of three. However, for the examined range of parameter variation, the location of soot itself is not affected. As can be seen in Figure 4-7, all other model constant variations do not show strong sensitivity regarding the soot concentration magnitude. Variations of those constants by a factor of two affect the peak soot volume fraction at most by 8 % for the case of doubled soot oxidation rate by OH.

![Figure 4-7: Influence of different soot model parameters on soot volume fraction (in ppmv) along the spray axis (left) and radially at Z=60 mm for case A1 at 5 ms after SOI.](image)

The amount of C-atoms required to form a primary soot particle was changed from 100 to 60 as proposed by Lindstedt [81]. An increase in soot load was observed, although sensitivity was weak (cf. Figure 4-7). Ma et al. [146] performed a sensitivity analysis with respect to various soot model constants for
a turbulent non-premixed ethylene/air flame. A much weaker sensitivity with respect to variations in coagulation and number of C-atoms per primary particle was found when using surface growth rate to be a square root function of soot surface area, as in this study, instead of a linear correlation as proposed in e.g. Lindstedt [81]. This is in agreement with what is observed in the present investigation. The soot surface growth process is the dominant source for soot mass increase. Inception rate does influence the number of particle, but not the mass considerably.

Since the emphasis of this study was not to carry out model constant calibration to obtain accurate predictions for all experimental conditions, all results presented in the remainder of this study are computed with model constant as proposed in Table 3-2. This approach allows for the assessment of the predictive capabilities of the soot model embedded in the CMC framework for the different ambient conditions reported in Table 4-1.

4.3.3.3 Unconditional quantities

Figure 4-8 shows the computed soot volume fraction distributions in comparison with the experiment for all conditions investigated. Note that experimental data are available only up to approximately 85 mm axial distance from the injector. For the cases with low O₂ contents the sooty region continues further downstream and results can only be compared qualitatively.

For the prediction of the soot region, a correct description of the flame structure is of vital importance. The CMC formulation showed good agreement with the experiments with respect to ignition delay and lift-off height for all conditions considered. Regions of soot presence and location of peak soot concentration are well captured by the model for almost all conditions. By diminishing the ambient oxygen concentration the soot region shifts more downstream from the injector and it becomes broader due to the reduced stoichiometric mixture fraction; this effect is in agreement with the experiment. The separation of the sooty region from the lift-off height is also well reproduced.

To facilitate the comparison for all nine cases considered, Figure 4-8 illustrates the evolution of the maximal soot volume fraction (left) and its axial location (right). Experimental values are drawn with squares and CMC results in red denoted as CMC-CMC. Note that simulation results with the DI (DI-DI) and with a hybrid formulation (CMC-DI) are also displayed in Figure 4-8 but are discussed later in Section 4.4.3.1 where the influence of the combustion model on soot formation is analysed.
Results I: Sandia constant-volume chamber

Figure 4-8: Evolution of peak soot volume fraction (left) and peak soot axial position (right) for different ambient densities: (a) 14.8 kg/m³ and (b) 30 kg/m³.

A reduced peak soot volume fraction when diminishing the oxygen availability was correctly captured for both ambient densities considered. For the 14.8 kg/m³ density cases, the soot volume fraction was over-predicted for all cases. The axial location of the soot peak is well reproduced. An enhanced soot load by increasing the ambient density was correctly forecasted: at the higher density conditions, the quantitative soot distribution was very well predicted for all ambient oxygen concentration except at 8 % O₂, where soot was over-predicted. A possible explanation for this good agreement at the higher ambient density could be the correct prediction of the ignition delay for all oxidizer content considered.

Soot formation modeling at the same experimental setup has also been investigated by various research groups with different approaches: Azimov et al. [160] employed the ECFM-3Z combustion model coupled with the two-step empirical soot model by Hiroyasu et al. [161]. A poor prediction capability for soot location under different oxygen concentration was reported. D’Errico et al.
[162] adopted the same empirical soot model aforementioned with the perfectly stirred reactor (PSR) combustion model. Soot distribution for case A1 and A4 were compared with experiments. For the first case, LOL was well reproduced, but soot location was not reproduced correctly, because peak soot load was not at the spray axis and high soot concentration was present until end of the visible domain, corresponding to approximately 85 mm. For the case with 10% O$_2$ the flame LOL was abundant over-predicted and, as a consequence, soot distribution was not described accurately. Vishwanathan and Reitz [20] showed soot distribution during the quasi-steady period of the spray for a broad range of conditions (8 out of 9 cases listed in Table 4-1). In general, trend of soot volume fraction distribution under different oxidizer content was well captured. In contrast to the lower gas density cases, at the higher one location of peak soot load was slightly under-predicted.

Figure 4-9 shows the mean particle diameter distribution at 5 ms after SOI for three different cases: A1, A4 and B2. In the soot model employed, a primary soot particle consists of 100 C-atoms, which corresponds to a sphere with approximately 1.24 nm diameter [80]. This size is found where first soot is formed; afterwards mean particle size is progressively increased due to surface growth and particle coagulation. Note that a higher mean soot particle diameter does not correspond necessarily to a higher soot concentration.

![Figure 4-9](image)

**Figure 4-9:** Computed mean soot particle diameter (in nm) spatial distribution at 5 ms after SOI for three different cases.
For the reference case A1, a maximal soot mean particle diameter of approximately 21 nm is found, which is in agreement with simulation findings pointed out by Vishwanathan and Reitz [20]. There, the slightly increase of the maximum primary soot particle size when diminishing the oxygen concentration is in contrast to the present results. An increased particle size by enhancing the gas density is however in agreement with Vishwanathan and Reitz [20].

Mainly motivated by legislation and potential health risks, the engine community notifies an increasingly interest on the particle size characterization. Very recent experimental work is ongoing for the characterization of soot particle size within diesel sprays. Measurements have been performed with laser induced incandescence (LII) [163] and with transmission electron microscope (TEM) analysis in a constant volume chamber [164] as well as in a diesel engine [165]. In the near future, the availability of such detailed measurement data will further support the validation of current models and a direct analysis of the various operating parameter will be facilitated.

It can hence be concluded, that despite the simplicity of the soot model employed and considering only one single inception species, the modelling approach followed in this study was able to predict semi-quantitatively soot distributions; for most cases, the discrepancy was within a factor of 2. Considering that this investigation presents a first application of a two-equation soot model in multi-dimensional CMC for two-phase combustion and high pressures, these results can be considered very encouraging given the wide range of conditions.

### 4.3.3.4 Conditional quantities

Conditional soot mass fraction (left) and soot number density (right) for the 21 % O2 case at (A-E) locations indicated in Figure 4-4 (b) is shown in Figure 4-10. In all selected locations soot mass fraction is present as expected only in the fuel rich region with a peak at an equivalence ratio of approximately 2.

The soot number density shows a similar shape, but still with a non-zero value in the fuel lean region. This is due to the numerical formulation of soot oxidation in the two-equation model used: oxidation by O2 and OH does decrease particle mass but not the number; therefore a non-zero number of particles with negligible mass is present in the fuel lean region, which does not influence soot volume fraction.
Figure 4-11 illustrates the conditional soot mass fraction and number density quantities and the corresponding contribution of the various source terms at the location of peak soot load at 5 ms after SOI for the 21 % and 10 % O₂ cases (A1 and A4, respectively). Selected locations were both on the spray axis and at 60 and 90 mm from the injector for the 21 % (left) and 10 % O₂ case (right), respectively.

Figure 4-10: Conditional soot mass fraction and number density for case A1 at 5 ms after SOI at the selected locations indicated in Figure 4-4 (A-E, mixture fraction for case (b), lower). The dashed vertical lines denote the stoichiometric mixture fraction.
Results I: Sandia constant-volume chamber

Figure 4-11: Conditional soot mass fraction and number densities (upper), source term contributions for conditional soot mass fraction (middle) and conditional number density (lower) at 5 ms after SOI at the location of maximal soot concentration for case A1 (left, z=60 mm, r=0 mm) and case A4 (right, z=90 mm, r=0 mm). The dashed vertical lines denote the stoichiometric mixture fraction.
In the source terms balance diagrams, only relevant contributions have been included. For the conditional soot mass fraction at the selected locations, the contribution of particle inception, axial and radial turbulent diffusion and radial convection was negligible. Conditional soot mass fraction and number density for the lower $O_2$ concentration (b) exhibit the same shape as in the higher $O_2$ case (a), but a lower magnitude by a ratio of about 12 and 3.5, respectively. The conditional source terms for the soot mass fraction at the location of the peak soot load for the two different ambient compositions, (c) and (d), show qualitatively the same shape, evidencing that soot related processes occur at the same equivalence ratio. Surface growth is the main soot source and it is mainly balanced by axial convection. Molecular diffusion of soot towards stoichiometry is counteracted by oxidation by oxygen and OH. It is interesting to note that, for the 21% $O_2$ case, oxidation by $O_2$ and OH are of comparable importance, whereas at 10% $O_2$ oxidation by OH becomes negligible due to the over-proportional decrease of OH concentration compared to ambient oxygen decrease (Figure 4-4). The importance of soot oxidation by OH is in agreement with findings reported in Kronenburg et al. [103].

Between Figure 4-11(c) and (d) there is a factor of about 40 in magnitude of ordinates-axis scale, which illustrates the much weaker soot related processes with lower oxidizer availability.

Concerning the soot number density with the different $O_2$ concentrations, Figure 4-11 (e) and (f), a similar distribution of the source terms is observed and a scale magnitude ratio of 13 is reported. Here, the dominant source is particle inception, which is mainly balanced by particle coagulation. Diffusion in conserved scalar space and axial convection have a weaker impact compared to the soot mass fraction.

**4.3.3.5 Unconditional soot terms**

The influence of ambient oxygen concentration on the various soot related phenomenology is visualized in Figure 4-4, while Figure 4-12 shows the contribution of the most important terms responsible for soot mass fraction evolution in physical space for the same two ambient oxygen concentrations. The unconditional soot terms are retrieved by integrating the various conditional soot terms over the mixture fraction PDF.
Results I: Sandia constant-volume chamber

Figure 4-12: Contribution of main unconditional soot terms for 10% and 21% O₂ at 14.8 kg/m³ and at 5 ms after SOI (in [1/s]). The black line denotes the stoichiometric mixture fraction. Oxidation by O₂ and OH and axial convection are visualized as a sink.
In both cases the main source for soot mass fraction is the surface growth which corresponds essentially to the presence of acetylene. Surface growth is mainly counteracted by oxidation, molecular diffusion, axial convection and radial diffusion, which act as a sink in the high sooty region.

The relative importance of soot oxidation by OH at the higher O<sub>2</sub> concentration is reported in the previous section. Note that oxidation by OH acts at higher mixture fraction compared to the oxygen induced oxidation (Figure 4-11 (b)), consequently it takes place more inside the sooty region as can be observed in Figure 4-12 (b). On the other hand, oxygen oxidizes the soot more around stoichiometry. The sensitivity of soot oxidation for the 21% O<sub>2</sub> case was shown in Figure 4-7. Oxidation by O<sub>2</sub> is strong in the region where first soot is formed (Z= 35-40 mm) and therefore an increased oxidation rate by O<sub>2</sub> slightly postpones axially the start of soot formation. On the other hand, a greater oxidation by OH anticipates the end of axial soot region and reduces more the peak soot volume fraction, because of its strong oxidation in the region of peak soot location.

### 4.3.4 Comparison with conceptual model

Idicheria and Pickett [73] and Pickett and Siebers [166] presented a revised conceptual model of diesel spray combustion compared to the well known model by Dec [159]. Based on n-heptane spray measurements at the Sandia constant-volume pressure vessel, Idicheria and Pickett [73] showed schematically the spatial distribution in the jet of a) formaldehyde by means of Planar Laser Induced Fluorescence (PLIF); b) high heat release rate zones (OH* chemiluminescence); c) Poly-Aromatic Hydro-carbons (PAH, also by means of PLIF) and d) soot (PLII) with 10 and 21 % oxygen mole fraction at a constant density of 14.8 kg/m<sup>3</sup>. For both oxidizer dilution levels, three different sooting propensities with increasing ambient temperature were observed: no-, low- and moderate soot conditions. In the following, measurements with 1000 K ambient temperature (case A4 and A1) are used for validation of the predicted spray structure. Figure 4-13 compares the regions of formaldehyde, high heat release rate, PAH and soot with 10 % (left) and 21 % (right) ambient oxygen: The measured PAH distribution is compared to the computed acetylene species, which has been considered as the unique soot precursor. The numerical region of various species has been defined by the isocontour where an arbitrary value of 25 % of the related peak mass fraction is found. The location of the diffusion flame is defined as the high temperature region along the 25 % ambient oxygen
isoline. In general, the species distributions in the spray structure are well reproduced: At the lower oxygen concentration the flame becomes broader and processes are shifted upstream. The location of the diffusion flame and formaldehyde species at the flame base is qualitatively in good agreement. Idicheria and Pickett [73] reported a triple flame structure at the flame base (not shown here). Borghesi et al. [132] investigated flame propagation and stabilization mechanism after autoignition, where a convective-reactive balance at the anchoring point was reported. The ability of multi-dimensional CMC to describe triple flame behavior in auto-igniting sprays has been previously demonstrated by Wright et al. [134].

The axial distance of the first acetylene appearance in comparison to the first measured PAH is considerably under-predicted. This can be intuitively expected, since in reality soot inception is a multi-step process including PAH [66]. For pure paraffinic fuels (no pre-existent PAH) formation of acetylene, PAH and soot is expected to occur in a sequential order.

![Figure 4-13: Comparison of spray conceptual structure for cases A4 (left) and A1 (right). Experimental figures modified from [73].](image)
Nonetheless, the soot distribution for both cases is well predicted, confirming the suitability of acetylene as the unique inception and growth species for \textit{n}-heptane fuel in the employed modelling framework. Both the axial shift between the soot precursor (gas-phase) and the solid-phase soot as well as the separation of the lift-off position and the start of soot formation are well reproduced. Vishwanathan and Reitz [167] performed a similar comparison between their simulations and the spray conceptual model. The authors used the chemical mechanism by Patel et al. [13] with a PSR method coupled with the two-equation soot model by Tao et al. [83], which is of comparable complexity as the one by Leung et al. [80] employed in this study. Their predictions of the spatial soot distribution showed high values too close to the lift-off height and the separation between soot and the lift-off length was considerably under-predicted. Improved soot results for the test cases listed in Table 4-1 were subsequently presented by the same authors [20]. The general trend of LOL and soot load was well predicted for all conditions considered, however no comparison of ignition delays for the autoigniting sprays for all test cases were given allowing for a joint comparison of both models’ performance.
4.4 Influence of combustion model

4.4.1 Flame structure

In order to analyse the differences arising from the combustion model used, Figure 4-14 displays a comparison of the quasi-steady flame structure for the A2 reference case for the DI (upper) and CMC (lower) models at 5 ms after start of injection (SOI). Iso-contours of quantities relevant for soot behaviour are considered, these are from left to right: mean MF, mean temperature, and mass fraction of oxygen, OH and acetylene, and volumetric heat release rate (HRR). The latter is drawn in logarithmic scale. The black lines represent the stoichiometric iso-contour. Note that, due to the symmetric spray arrangement, only half of the spray is visualized.

The comparison between the two models reveals that, as expected, the mean MF distribution is approximately the same, as fuel evaporation and scalar transport are treated in the same way. Marginal differences are caused by the different temperature distributions, which influence the fuel evaporation. The main difference between the two computed flames lies in the spatial distribution of oxygen. With DI the oxygen is completely consumed at the stoichiometric iso-line and no oxygen is left in the fuel rich region. On the other hand, the CMC method predicts a certain stratification also in the rich region. This broadening is expected in turbulent combustion and is due to the description of the MF through a PDF. However, oxygen is completely consumed in MF space for some MF. The DI case results in a much thinner layer of OH along stoichiometry as well as a thinner layer with high chemical activity as revealed by the HRR distribution. The peak OH mass fraction predicted by DI is approximately twice as high as the corresponding CMC value, but in a smaller volume; as a consequence, the resulting field totals are of roughly the same order.

As expected, for both models the highest HRR is found at the LOL; downstream along the stoichiometric iso-contour, a diffusion flame is established with one or two orders of magnitude lower HRR. There is also a rich premixed branch in the inner part of the spray starting from the anchoring point, consistent with the conceptual model proposed by Idicheria and Pickett [73]. For both models the HRR exhibits the same spatial extent as OH species.
Figure 4-14: Spatial distribution of relevant quantities for flame characterization at 5 ms after SOI for the reference case A2 computed with DI (a) and CMC (b). From left to right: mean mixture fraction, temperature, mass fractions of acetylene, oxygen, OH and CH$_2$O, respectively. The black line denotes the location of the stoichiometric mixture fraction $\xi_{st}$. 

Results I: Sandia constant-volume chamber
The thin OH layer obtained when using DI is consistent with former DI studies for diesel spray, e.g. [25, 43]. Comparison of OH profiles using different combustion models was also focus of the ECN2 workshop and the same trends were reported [29]. For the experiment considered in this study there is no OH-PLIF (planar laser induced fluorescence) measurement available. However, under similar conditions LIF measurements have been performed and instantaneous OH profiles were found to be considerably thin (less than one mm), e.g. [168, 169]. At this point it should be recalled however that the RANS solution is representative for an ensemble average over many injection events. Images of OH* chemiluminescence show a considerable flattering of the flame and soot region [29]. Applying ensemble averaging over many injection events, a thicker averaged OH profile is hence to be expected as also discussed in [62], where the thin flame predicted by DI is considered implausible compared to the flame brush structure predicted by the composition PDF method employed.

As a consequence, the peak mean temperature exhibits higher values for DI. The lower peak mean temperature of CMC is, again, caused by the PDF description of the local composition. In this specific case the difference was approximately 100 K.

As acetylene is the unique species responsible for soot formation in the model employed here, it is important to compare its distribution for later soot predictions. For the two models studied, a similar structure and the same concentration values are found. The spread of the soot precursor is, however, larger for the CMC for the same reason of the MF PDF discussed above, hence there is some acetylene escaping in the mean fuel lean region.

The flame lift-off position is clearly visible, due to the strong axial gradient of mean temperature and OH mass fraction. For the reference case A2 DI estimates a slightly higher LOL, as will be discussed next.
4.4.2 Ignition delay and lift-off length

Figure 4-3 compares ID time (left) and flame LOL (right) for the two combustion models against experimental data for ambient oxygen variations at three different ambient conditions: at 1000 K with 14.8 kg/m$^3$ (upper) and 30 kg/m$^3$ (middle) and at 900 K with 14.8 kg/m$^3$ (lower). In general, ID was well reproduced for both methods. The chemical mechanism employed tends to overestimate ignition delay for lower oxidizer reactivities, in particular for low ambient oxygen contents at the low density cases. At the higher ambient density the chemical model showed very good agreement. This trend is consistent with former studies employing the same chemical mechanism, i.e. [25, 26, 132, 158]. Therefore, the influence of TCI on auto-ignition is found to be comparatively small throughout the entire range of conditions considered here.

For what concerns flame LOL, both models capture the correct trend with an increase in LOL by decreasing oxygen content. Here, the influence of the combustion model is seen to be more important. DI predicts a larger LOL for all cases, in particular at the lower ambient density and low oxygen volume fractions where LOL was considerably overestimated.

The above observations are in line with the results reported in [21] showing the comparison of DI with the transported PDF method for the same $n$-heptane spray considered here where it was found that neglecting turbulent fluctuations has a minor influence on ID, whereas considerable increase in LOL has been observed. In the following, reasons for the observed behaviours in ID and LOL are further analysed. There are a few main differences between the models. First, the DI considers the computational cell as well-mixed, whereas CMC assumes the MF distribution within the computational cell to be a $\beta$-function. Differences in average HRR decrease by diminishing MFV. Second, CMC includes the SDR as an important parameter that represents molecular mixing, a phenomenon which is missing from the DI formulation. At high levels of SDR auto-ignition is delayed or even completely inhibited [170]. For the limit case of zero variance and zero SDR, the CMC becomes identical with the well-mixed assumption that implies a delta function for the species PDFs at the mean values and no small-scale mixing effects on the reaction rate.
4.4.2.1 Discussion on ignition delay

Auto-ignition of non-premixed flames is a complex process, a review of which can be found in [170]. Auto-ignition is known to occur at a characteristic MF, the so-called most reactive MF [171], which for higher hydrocarbons such as n-heptane under diesel engine conditions has been seen to be in fuel rich [134, 137]. Recently, DNS results of auto-ignition of n-heptane droplets at high temperature and pressure confirmed that the concept of most reactive MF is still valid for evaporating autoigniting droplets [172].

Here, as a preliminary step to study the influence of SDR on auto-ignition under diesel engine conditions, transient flamelet calculations according to [49] have been carried out with different conditional SDR applying the AMC model [145] for all five ambient dilutions at the higher temperature (1000 K) and the lower ambient density (14.8 kg/m³) conditions.

Results are shown in Figure 4-15 in terms of ID versus SDR (right) at the most reactive MF. The most reactive MF has been determined for every oxidizer dilution assuming zero SDR following common practice, i.e. [137, 170]. The most reactive MF was found to remain in a confined equivalence ratio range of 2-2.5, as illustrated in Figure 4-15 (left), while the stoichiometric MF obeys a linear decrease for increasing dilution. As expected, low values of SDR have minor influence on ignition delay, as can be seen for all states of dilution. The respective, relatively constant delay times are however considerably higher for the diluted cases due to the substantial reduction in reactivity of the oxidiser.

Figure 4-15: Left: Stoichiometric and most reactive mixture fraction at different ambient dilutions. Right: Flamelet calculation results of ignition delay for different values of SDR at 1000 K and 14.8 kg/m³ ambient temperature and density respectively.
For higher SDR, auto-ignition is increasingly inhibited and the critical SDR values, at which ignition can no longer occur, also decrease with oxidizer dilution as the mixture becomes less reactive. For the reference case with 15% O₂, values of SDR at the most reactive MF up to 15 s⁻¹ have a marginal influence on ID and this value is of relevance for later discussion.

The reference case A2 is considered next to study the temporal history of temperature, SDR, and two low-temperature pre-ignition species CH₂O and H₂O₂. The flow field prior to auto-ignition, i.e. at 0.7 ms after SOI (the computed ID is approx. 0.9 ms for both models) is displayed in Figure 4-16. From left to right: mean MF, mean temperature, mean SDR, and mass fraction of CH₂O for both CMC and DI. Note that the MF and temperature distributions are illustrated for the CMC solution since results with DI are very similar prior to ignition as discussed above and have therefore been omitted for brevity. The mean SDR exhibits high values at locations where considerable spatial gradients of the MF are present and very low SDR is increasingly found towards the tip of the spray. Both the concentration and spatial extent of CH₂O were found to be almost equivalent between the models. The highest auto-ignition activity is close to the spray tip where higher temperature in conjunction with low SDR is beneficial for auto-ignition. As a consequence, the predicted location of the ignition spot was found to be virtually identical for both models with a difference of 1 mm.

The first autoigniting fluid particle has been tracked in order to describe the difference in temporal history of chemical activity. The black points in Figure 4-16 corresponds to the trajectory of the autoigniting fluid particle at the given time instants denoted by the numbers in milliseconds. The time instant of 0.7 ms, for which the spatial distributions are shown, corresponds to the CH₂O peak where ignition activity is the highest. The temporal evolution of temperature, conditional SDR at the most reactive MF, and auto-ignition species CH₂O and H₂O₂ along the trajectory are drawn in Figure 4-17. For the early time instants where the fluid particle is still located in the core of the spray the gas temperature is below 800 K due to evaporation and air/fuel mixing. The gradual increase in temperature until high-temperature ignition is mostly due to mixture becoming leaner downstream rather than due to the HRR.
Results I: Sandia constant-volume chamber

Figure 4-16: Field distributions at 0.7 ms after SOI for A2 reference case. Mean mixture fraction, temperature and mean scalar dissipation rate are for CMC. Points represent the position of the autoigniting fluid particle at different time after SOI in ms.

Figure 4-17: Temporal evolution of temperature (blue), CH2O (red) and H2O2 (green) and conditional scalar dissipation rate at most reactive mixture fraction (black) along first autoigniting fluid particle for the reference case A2 for DI (dashed lines) and CMC (solid lines).
Results I: Sandia constant-volume chamber

Of primary importance is the evolution of SDR showing high values in the first phase up to 0.4 ms; at later instants very low values are present, well below the critical value. Therefore, the delaying effect of turbulent straining on auto-ignition is limited to the very early regions of the spray and the overall ignition delay time is hardly affected, as emphasized in Figure 4-17. The evolution of CH$_2$O and H$_2$O$_2$ exhibited similar behaviour with a rapid increase in concentration during low-temperature pre-ignition activity and a period with nearly constant concentration and a rapid consumption during the high-temperature auto-ignition. H$_2$O$_2$ is found to be consumed slightly earlier and the two models showed a very similar behaviour. Generally, for DI higher rate of change of species mass fraction and temperature were observed. Low-temperature species CH$_2$O and H$_2$O$_2$ were formed slightly later with DI, which is probably due to the low mean temperature present there that affects chemical activities. The difference in peak temperature after auto-ignition is due to the thinner flame and complete consumption of oxygen as discussed earlier in the context of Figure 4-14.

Along the same fluid particle, conditional temperature and CH$_2$O are illustrated in Figure 4-18. From the temperature evolution it is clearly visible that the most reactive MF is at around 0.12-0.14, in agreement with the values determined by the stand-alone calculation for this reference condition (cf. Figure 4-15). A subsequent rapid transition of the peak conditional temperature towards stoichiometric conditions can be clearly observed. On the other hand, CH$_2$O is increasingly formed in the fuel rich region prior to auto-ignition and starts to be rapidly consumed at the onset of ignition at 0.85 ms in the presence of high-temperature reactions as is clearly visible from the ‘dent’ at the most reactive MF and continuing towards leaner conditions. The same dynamic is encountered for H$_2$O$_2$ and therefore not presented here.

Overall, both models showed comparable results and confirm that auto-ignition under diesel engine conditions occurs prevalently at low values of SDR, much lower than the critical value, where TCI plays a minor role because the molecular mixing delaying effects are small. Although not shown here, the same argumentation also applies to the other operating conditions considered and explains why good ignition delay predictions can be achieved with DI despite the neglect of the turbulent fluctuations. It is important to note that although dilution substantially decreases the critical SDR (as shown in Figure 4-15, left), the effective SDR a particle experiences decreases as well since the ignition
location is shifted further downstream as a consequence of the lower reactivity, as has also been observed experimentally [173].

Figure 4-18: Evolution of conditional temperature (left) and CH$_2$O mass fraction (right) along first autoigniting fluid particle for the reference case A2. Vertical dashed lines denote stoichiometric mixture fraction $\xi_{ST}$. 
4.4.2.2 Discussion on lift-off length

In the following section, discrepancies in LOL are further discussed by means of conceptual differences between the models. In the literature, there is evidence that the LOL under diesel engine conditions is mainly governed by auto-ignition following considerations put forward in e.g. [173-175] and references therein. Indeed, CH$_2$O as characteristic species for cool-flame has been detected upstream of the LOL indicating that auto-ignition is an important process for the stabilization mechanism. Furthermore, the residence time needed by a fluid particle to travel from the injector tip to the LOL was found to collapse into an Arrhenius type expression, consistent to what one would expect for ignition delay [173]. In this study, DI was found to exhibit a higher LOL compared to CMC for all test cases considered as shown in Figure 4-3. The discussion below attempts to explain this.

Figure 4-19: Mixture fraction PDF at ignition location (blue) at 0.9 ms and at LOL (red) at 5 ms after SOI.

Upstream of the flame stabilization point, high levels of MFV are present, causing a large difference between mean temperature and conditional temperature in MF space. Typical differences in MFV are shown in Figure 4-19 by means of MF PDF at the ignition location at 0.9 ms and at the LOL position at 5 ms after SOI. At the LOL, the mean value is comparable, but the MF PDF spread is considerably larger, caused by the larger MFV present locally. Obviously, high values of variance lead to increased errors in the evaluation of the non-linear chemical source terms by the DI method.
Figure 4-21 compares CH$_2$O and temperature distribution between the models in the upstream region at 5 ms after SOI during the quasi-steady period for the reference case A2. The lift-off position is clearly visible from the temperature at the anchoring point, where DI exhibits a higher distance from the injector tip. Mean MF and mean SDR are from the CMC solution; DI predicts equivalent distributions which are therefore omitted here. Upstream of the LOL high values of SDR are present as well as MFV (not shown here) and downstream a rapid decrease of both quantities is observed, as was the case for the ignition location at the spray tip. As observed in Figure 4-16 (right), both models predict the highest HRR at the LOL, with a higher peak for DI. However, in the region 20-25 mm axially from the injector tip, CMC was found to exhibit a higher chemical activity, which is confirmed by the higher concentration of CH$_2$O in this region (cf. Figure 4-16). DI predicts higher peak values of HRR and CH$_2$O but in a more confined region downstream, possibly caused by the mean value effect that becomes dominant for high values of MFV. For both the CMC as well as the DI approach, low values of formaldehyde are found in regions with high SDR, where chemistry is considerably inhibited, and CH$_2$O is completely consumed in the presence of high-temperature reactions, consistent with experimental observations from [74].

Figure 4-18 shows conditional quantities from the CMC solution at 5 ms after SOI at different locations along the stoichiometric iso-line at the location marked by points in Figure 4-20. Plotted conditional quantities are: temperature, HRR, and CH$_2$O. In the conditional temperature evolution high-temperature ignition is clearly visible between 24 and 26 mm, the latter coinciding with the LOL. There, the spatial gradient of temperature is extremely high with an increase of approximately 1000 K within 2 mm axially. The strong chemical activity at the LOL is confirmed by the high HRR at 26 mm and the coinciding complete consumption of CH$_2$O in the high-temperature region. Upstream, some chemical activity in form of CH$_2$O formation is noticeable, however at a considerably lower HRR and hence a marginal temperature increase. Downstream of the LOL the HRR is again considerably reduced. It is interesting to note that the sudden temperature rise does not occur at the most reactive MF as for conventional auto-ignition, but at around stoichiometric conditions. This is probably due to the fact that at the LOL the flame is already established and therefore lies along the stoichiometric MF; see [170] for a comparison between autoigniting jets of fuel in hot air and lifted flames in cold air.
Results I: Sandia constant-volume chamber

Figure 4-20: Field distribution for the A2 reference case at 5 ms after SOI. From left to right: CMC solution of mean mixture fraction, mean scalar dissipation rate, CH$_2$O and temperature and DI solution of CH$_2$O and temperature.

Figure 4-21: Conditional temperature (left), heat release rate (right) and CH$_2$O mass fraction (lower) for the reference case A2 at 5 ms after SOI at location indicated by point in Figure 4-20. Vertical dashed lines denote stoichiometric mixture fraction $\xi_{ST}$. 
The contact of the incoming unburned mixture with the flame leads to an induced ‘forced’ ignition for MF between 0.02 and 0.1, corresponding to regions with conditional temperatures above around 1200 K. Simulation results revealed that at the anchoring point a convective-reactive balance was found to be the relevant stabilization mechanism as presented in detail in [132] and not repeated here. The chemical source at the LOL is a combination of different combustion modes as visible from the conditional HRR, consisting of a stoichiometric diffusion flame in conjunction with a lean and rich premixed branch travelling in opposite directions in MF space to consume the premixed fluid developed during the time of flight from the nozzle to the flame base. As we move slightly downstream from the LOL, the stoichiometric diffusion flame persists albeit at a lower intensity, whereas both premixed branches are transported towards the extremes of MF and at the same time are diffused, reducing the magnitude of premixed burning. Farther downstream from the LOL, chemical reactions are mainly balanced by molecular diffusion in MF space (not shown here) as expected for diffusion flames.
4.4.3 Soot formation

4.4.3.1 Soot averages

In this section results are presented using three different approaches. First, the conventional CMC method as presented in [26] where TCI is accounted for both flame and soot. In the following, this model is referred to as CMC-CMC. Second is the conventional DI approach that does not account for TCI neither for flame nor for soot, here referred to as DI-DI. The third method is a numerical experiment, a hybrid variant between the first two methods solving the flame with CMC and soot computed directly with unconditional quantities, and is noted as the CMC-DI method. This third method is sometimes used in some CFD codes as a post-processing step to estimate pollutants independently of the combustion model used to get the HRR.

Soot distributions for the reference case A2 at 5 ms after SOI are shown in Figure 4-22. Note that the experimental measurement domain does not extend further than 85 mm axially from the injector. Assuming a quasi-symmetric distribution of measured soot the latter is expected to be present until 95-100 mm downstream. At 5 ms after SOI all three models predict a quasi-steady distribution of soot consistent with the experiment.

![Figure 4-22: Soot volume fraction [ppmv] for the reference case A2 at 5 ms after SOI. From left to right: model results with DI-DI, CMC-DI and CMC-CMC and experimental data. Note that the experimental domain extends only until 85 mm axial distance as denoted by the horizontal red dashed line.](image-url)
The DI-DI and CMC-CMC methods showed practically the same peak soot volume fraction which is a factor of 2.5-3 higher compared to the experimental value. Both models are also able to reproduce the region of high soot concentration. The difference here is, again, the spread of the soot distribution analogous to the species (cf. Figure 4-14). It is interesting to note that the experimental spatial extent of soot lies in the middle between the two model predictions. High-speed LII images show considerable spatial and temporal fluctuations of the soot signal, analogous to OH* chemiluminescence fluctuations. The hybrid CMC-DI approach considerably underestimated both the peak as well as the location of the soot region.

To facilitate the comparison for all nine operating conditions, Figure 4-8 compares the performance of the three approaches by means of maximal soot volume fraction (left) and its axial location (right). The hybrid CMC-DI model shows poor agreement with the experiment in terms of the predicted soot amount as well as its position, which are both strongly underestimated for almost all conditions. The reason for this behaviour will be further discussed in the next section. Both DI-DI and CMC-CMC models were found to reproduce semi-quantitative trends of soot volume fraction and the peak soot axial location was well captured with both approaches. At the lower ambient density the model tends to overestimate soot while at the higher density both CMC-CMC and DI-DI models were able to reproduce very well the peak soot, apart for the 8 percent oxygen case where both methods overpredict soot. Overall, the model performance of DI-DI and CMC-CMC was found to be comparable to each other and models show considerably better predictions than the ‘hybrid’ CMC-DI approach.
The soot inception time is introduced as the period between ID and first soot appearance. The latter has been defined as the first time when a threshold value of 10 percent of peak soot volume fraction during the quasi-steady spray is formed. Predictions of both models are compared to experimental data reported in [176] for the low ambient density cases. For measurements at 8 % O₂ no soot was detected, and hence in this case no experimental inception time is defined. Experiments revealed that characteristic soot inception time augments by increasing oxidizer dilution due to the lower flame temperature and higher fuel dilution. The simulation is capable to reproduce this trend for increasing oxidizer dilution. In general the computed inception time is shorter than measurements. The choice of the arbitrary threshold value of 10 percent of the soot peak was found to have some degree of sensitivity, where a choice of 20 percent increases the simulated inception time by 20 percent at most for the case with the highest dilution. Conceptually a lower inception time is to be expected, as modelled inception is a one step reaction through acetylene and no complex PAH inception path has been attempted. Here, TCI was found to play a minor role.

4.4.3.2 Soot source terms
In order to conduct a fair comparison of the soot source terms and to be able to isolate the effect of TCI (turbulent fluctuations) on soot formation, the flame computed with CMC for the A2 case at 5 ms after SOI is used for the calculation of soot source terms. For the CMC method, soot sources are computed in MF.
space and the unconditional values are obtained by convolution with the presumed PDF at the CFD resolution. The DI methodology employs the species mass fractions and temperature predicted by the CMC and calculates the soot sources directly from these Favre mean values (neglecting any fluctuations). The comparison is presented in Figure 4-24 by means of distributions of the surface growth and oxidation terms since these represent the main sources and sinks: oxidation by O₂ and OH for the DI (left) and CMC (right) methodologies.

The surface growth rate is roughly the same between the models with a slightly narrower distribution with DI in the lean region. On the other hand, the differences in soot oxidation rate are very high, where DI exhibits higher peak values by one order of magnitude for soot oxidation by OH and even two orders of magnitude for the O₂-driven soot oxidation. It is important to note however, that the relative magnitudes are strongly dependent on the dilution level as discussed in [26].

![Figure 4-24: Chemistry related soot sources (surface growth, oxidation by O₂ and OH) for the reference case A2 at 5 ms after SOI using the flame and soot distribution from the CMC solution. Left computed with DI and right computed with CMC.](image-url)
Distributions of soot oxidation by OH have approximately the same spatial extent. Regarding soot oxidation by O₂, DI shows the highest levels at the spray tip in correspondence with the higher oxygen concentration, whereas CMC predict soot oxidation more homogeneously along the entire stoichiometric region of the spray as one may expect.

In the following, the origin of the massive differences in soot oxidation by O₂ is discussed considering conditional quantities in MF space. Characteristic conditional profiles of oxygen and soot surface area in a normalized form in conjunction with the MF PDF with two different MFV are drawn in Figure 4-25a (left).

It is important to note that chemical reactions take place at the molecular level, meaning at the same MF, which describes the state of mixing within the cell. Here, soot and oxygen have a very small MF overlap at around stoichiometry where soot oxidation is supposed to occur. The CMC method accounts for the limited co-existence of soot and oxygen at the molecular level whereas the DI does not consider scalar distribution within MF. The use of unconditional values of soot and oxidiser present in the CFD cell to calculate the oxidation rates in the case of DI neglects the limiting influence of the narrow soot-oxidiser co-existence and consequently dramatically overestimates soot oxidation. The same consideration applies for the OH driven soot oxidation where, OH-soot co-existence is present in a small range of MF. The soot-OH co-existence issue has been formerly noticed by Kronenburg et al. [103] for non-premixed methane-air flames, where the limited soot oxidation was observed. For a given mean composition in the CFD cell the relative discrepancy in soot oxidation rate increases by increasing the MFV, as qualitatively illustrated in Figure 4-25a, left: the larger MFV has a significant impact on mean quantities (indicated by the red and blue hatched areas), however the reaction rate remains almost unaffected as shown by the green ruled surface shown on the right. On the other hand, for a variance approaching zero the models tends predict the same reaction rate as the cell becomes well-mixed and turbulent fluctuations are zero.

In the case of processes related to acetylene, i.e. soot inception and surface growth, the soot-C₂H₂ co-existence at the molecular level is wider because both are present in the fuel rich region at an equivalence ratio of around 1.5-2.5 as illustrated in Figure 4-25b (left). A change in MFV has an influence on formation rate and therefore the well-mixed assumption causes smaller differences.
Figure 4-25: Conceptual visualization in mixture fraction space of soot oxidation by oxygen (upper) and soot surface growth (lower). Left: normalized species profile (blue and red lines) and mixture fraction PDF (dotted line). Right: normalized species profile (blue and red lines), mixture fraction PDF (dotted line) and reaction rates (green lines). Vertical dashed lines denote stoichiometric mixture fraction $\xi_{ST}$. 

(a) Soot oxidation by oxygen

(b) Soot surface growth
It can be generalized that mean reaction rates for processes involving species with a broad MF co-existence range are less sensitive with respect to the neglect of turbulent fluctuations compared to reaction rates for processes involving species with significant curvature, as expected [177].

The same trends of relative differences in surface growth and oxidation by O₂ and OH have been found for all other test cases (not shown here) and the ratios of the maximal soot source terms computed with DI over the ones using CMC have been considered. The soot oxidation by OH showed an almost constant ratio of approximately 10 over all cases, whereas the O₂ supported oxidation showed a considerable increase of the ratio for increasing ambient oxygen concentration for both ambient densities considered. The ratio of the O₂ soot oxidation rates spans between 10 and 1000 for the low and high ambient oxygen contents, respectively. This strong dependency on the oxidizer composition, as well as the associated differences in soot/oxidiser overlap and PDF shapes on soot oxidation behaviour hence present a great challenge for models which use only mean values w.r.t. the derivation of “universal” oxidation rate expressions that are appropriate for the broad range of oxygen concentrations studied here.

Despite the vast differences observed in the soot oxidation rate for the case of a given flame characteristic, the soot distribution for the quasi-steady spray is very similar for the DI-DI and CMC-CMC approaches (cf. Figure 4-9). The main reason for that is twofold: soot formation was not considerably affected by turbulent fluctuations and the complete absence of oxygen and OH within the fuel rich region for the DI case. In this sense the soot oxidation is zero in the rich zone and very high at the stoichiometry and in the lean region, therefore the quasi-steady spatial distribution of soot is almost unaffected.

In typical diesel engine operation the quasi-steady spray period constitutes only a minor portion of the total injection process duration. Only in operating conditions exhibiting negative ignition dwell, i.e. where the ignition delay time is clearly shorter than the injection duration, can a quasi-steady spray flame be established; the duration of which is confined between the end of the premixed burn phase following auto-ignition and the EOI. Cases with significant mixing-controlled combustion of the quasi-steady spray are commonly characterised by high oxidiser reactivity (shortening ignition delay) and long injection durations typical of full load operation (and highly typical also to low-speed large marine diesel engines). Due to the intermittent nature of diesel engines, all operating conditions, i.e. whether a quasi-steady spray can be established or not, are
subject to EOI transient effects which influence the soot oxidation process, and will be discussed next.

4.4.4 Short injection case

Since spray combustion in diesel engines is an intermittent process, and processes occurring after the EOI are very important, especially due to mixture leaning caused by entrainment of oxygen leading to an oxidation of most of the soot previously formed. In diesel engines, exhaust soot is typically about two orders of magnitude lower than the in-cylinder maximal value [178]. As a consequence, the description of soot oxidation, in particular after EOI, is of supreme importance in order to predict engine-out soot. For this purpose, the test case S3 (cf. Table 4-1) with a short injection duration of 1.8 ms at 12 % ambient oxygen has been considered in addition to the ‘quasi-steady’ sprays. In the experiment, phenomena occurring after EOI for the n-heptane spray have been investigated in detail in [176, 179]. In this study, the temporal evolution of soot is of particular interest and processes involved after EOI are emphasized.

Figure 4-26 illustrates iso-contours of the flame structure together with quantities relevant for soot oxidation at 3 ms after SOI (corresponding to 1.2 ms after EOI) for DI (upper) and CMC (bottom). From left to right: mean MF, temperature, mass fraction of oxygen and OH and rates of soot oxidation by O\(_2\) and OH.

Equivalence ratio measurements at 3 ms after SOI for the S3 test case under non-reactive conditions (0% O\(_2\)) performed by Musculus and co-authors [179] revealed a lean non-zero fuel concentration in the upstream region almost until the injector and lean conditions until the end of the measurement domain (55 mm axially from the injector tip). The simulation, although under reactive conditions, reproduces qualitatively well this behavior. As expected, minor differences between computed fields of mean MF were observed that can be caused by the different temperature distributions affecting evaporation behavior (droplet source) and local density. Fuel rich conditions are exclusively present in the spray tip region as depicted from the superimposed black stoichiometric iso-line. Despite the comparable mixing field, the oxygen, OH and, as a consequence, temperature show considerable differences, where the DI flame is thinner for the same reason as discussed in section 4.3.2 describing the flame structure.
Results I: Sandia constant-volume chamber

Figure 4-26: Spatial distribution of relevant quantities for flame characterization at 3 ms after SOI for the S3 case with DI (a) and CMC (b). From left to right: mean mixture fraction, temperature, mass fractions of oxygen, OH and soot oxidation rate by O₂ and OH, respectively. The black line denotes the location of the stoichiometric mixture fraction $\xi_{ST}$. 
In contrast to the quasi-steady spray, after EOI the enhanced oxygen entrainment has a profound impact on soot oxidation due to mixture leaning and thus large differences between the models can be expected.

To elucidate this effect the temporal evolution of the spatial integrated natural luminosity is first compared to experimental data from [176], as shown in Figure 4-27. In the experiment, soot natural luminosity stems mainly from soot radiation due to its much stronger signal intensity compared to chemiluminescence at almost every wavelength as discussed in [179]. Therefore, in the simulation, natural luminosity was assumed to consist of soot radiation only. The latter is computed according to:

\[ S_{rad} = 4\sigma \alpha_{soot} T^4 \]  

(55)

where \( \sigma \) is the Stefan-Boltzmann constant. \( \alpha_{soot} \) is the soot particle mean absorptivity coefficient assumed to be \( \alpha_{soot} = 2370 \frac{1}{mK} \cdot f_{soot} \cdot T \) [148]. Mean values of soot volume fraction and temperature at the CFD resolution have been used. For the CMC method the authors refrained from computing convoluted conditional radiation for this particular data processing since the scope of this consideration is confined to the comparison between combustion models. Results are compared in form of normalized arbitrary units. In the experiment, soot appears with some delay (approximately 1 ms) after high-temperature ignition has occurred and the peak signal of the natural luminosity is reached at around 3.1 ms after SOI. The subsequent decrease due to soot oxidation and cooling can clearly be observed.

During the soot formation phase both simulation approaches were almost identical until peak soot luminosity is achieved. This confirms that the soot formation process is practically unaffected between the two different combustion models. The rate of luminosity increase is in qualitative agreement; however it occurs slightly earlier than experimentally observed, consistent with the shorter predicted inception times discussed in Figure 4-23. The timing of the computed luminosity peak is also slightly earlier for both models. During the net oxidation phase the CMC is capable of well reproducing the luminosity trend, whereas the DI, as expected, considerably overpredicts the oxidation rate. It is important to note that a decrease in luminosity can be caused by a decrease of either soot volume fraction and/or soot temperature. In this case the strong decrease in soot luminosity for the DI is caused predominantly by soot oxidation,
Results I: Sandia constant-volume chamber

Figure 4-27: Temporal evolution of normalized spatial integrated natural luminosity for the S3 case. CMC (red), DI (blue) and experiment (squares).

Figure 4-28: Evolution of spatial natural luminosity for the S3 case. Simulations (DI and CMC) show soot volume fraction with fixed scale range 0-0.6 ppmv. Experimental data modified from [176]. The colour scale is the same as in Figure 4-26. Times after SOI are indicated at the bottom of each figure.

where soot is completely oxidized after approximately 3.8 ms after SOI (not shown here).

Moreover, the evolution of spatial distribution of soot is compared to luminosity measurements in Figure 4-28, from which qualitative comparisons of the sooty region can be drawn: for the simulation the color scale represents the soot volume fraction levels with a fixed scale within the range of 0-0.6 ppmv, for both models with the same color code as employed in Figure 4-22. Regions of soot radiation were found to coincide with those of the soot volume fraction and
therefore soot radiation has been omitted in this diagram. Note that the measurement consists of a line-of-sight signal. The border of the natural luminosity image is considered here to be a qualitative marker for the soot region. This was motivated by recent measurements by Cenker et al. [163] reporting an experimental comparison of natural luminosity with PLII for a similar short injection case (Spray A, see [29]) and the border of the LII signal was found in fair agreement with the border of natural luminosity distribution.

In the following, results are shown for four different time instants during both the formation and oxidation phases. The measurements revealed that soot first appears at the tip of the spray and later in the so-called “roll-up” vortices, where the lowest mixing rates are present [179]. A similar behavior was also reported in [180].

Both DI and CMC predict the first soot around the tip of the spray as can be seen in the left-most group of images. For the DI case soot remains at the tip which is completely oxidized at the later time instants. On the other hand, the soot distributions predicted with CMC remain broader and survive until later stages. The slight shift of the soot cloud towards the roll-up vortices is clearly recognizable in the right-most group of images at 3.7 ms, the time instant at which complete oxidation of soot is reached in the case of DI.

Computed soot oxidation rates by oxygen and OH at 3 ms after SOI are compared in Figure 4-26, nearly corresponding to the instant with peak luminosity. It is clearly visible that DI manifests roughly one order of magnitude higher local oxidation rates due to the presence of O₂ and OH. The sudden presence of oxygen in the sooty region oxidizes the soot very fast as was reported also in previous studies, e.g. [20, 107].
5 Results II: Wärtsilä marine spray combustion chamber

5.1 Experimental test facility

The experimental data used for model validation stems from a large two-stroke marine Diesel engine reference experiment installed at Wärtsilä Switzerland for which documentation is given in [181-184]. The marine Spray Combustion Chamber (SCC) is an optically accessible disk shaped chamber of 500 mm diameter and 150 mm height. Fuel is injected from the periphery into swirling hot air or nitrogen at high pressure reproducing conditions typical of large two-stroke Diesel engines. Figure 5-1 provides a schematic sketch of the SCC working principle.

Figure 5-1: Schematic sketch of the marine spray combustion chamber (adopted from [184]).
In order to achieve realistic conditions at start of injection, a heated and pressurized air (or N₂) flow through inclined inlet ports is provided by a pressure vessel/heat regenerating system. The inner core of this regenerator consists of a tensioned package of electrically heated discs with clusters of plates in between and is insulated against the housing by ceramic rings. Two types of those heat disks are used in order to enforce a labyrinth type flow of the process gas inside the regenerator. The radial passages for the flow between two neighbouring heat discs as well as the heat flux from the disks to the plates are provided by triangular distance plates fitted between the individual plates and disks. Shortly before the desired initial pressure in the chamber is reached, the accumulator valves close and injection starts, followed by combustion in the reactive cases. The swirl level range (15-25 m/s) can be achieved at the same time and has been confirmed by Laser Doppler Velocimetry (LDV) measurements [181, 182]. Finally, the exhaust valve opens; the regenerator is heated up again within a few minutes, while the compressor is refilling the accumulator with air or nitrogen.

The spray has been visualized under reactive and characterized under non-reactive conditions (evaporating and non-evaporating) by means of "Shadow-imaging" [181-183]. The illumination has been performed using a high-speed pulsed diode laser light source (690 nm, 20 kHz) with very short 50 ns laser pulses. In combination with an appropriate narrow band pass filter (CWL 689.1 nm, T 60%, FWHM 10.6 nm), a high signal-to-noise ratio in the recordings of a high-speed CMOS-camera (20 kHz frame rate, 512x512 pixel) with a minimum exposure time of 1 μs is achieved. The flame light is almost completely suppressed and spray visualization becomes feasible even under reactive conditions. As a consequence, considerably "sharper" images are obtained and it is possible to continue observation of sprays even after ignition has taken place. The flame region was detected by means of chemiluminescence imaging of the OH* radical as tracer for high temperature zones [185]. The light from the OH* was directed through a narrow band pass filter (313 nm, FWHM 10 nm) and a UV objective, into a high-speed image intensifier (IRO) connected to a high-speed CMOS-camera. The appropriately configured setup was used to investigate the ignition delay, ignition location and the lift-off during stable combustion. In parallel, the global OH* signal was recorded with an optical probe, allowing to measure the ignition delay with higher sampling rate.

In the experiment, ID and LOL were defined as the time and distance from the injector tip where a minimal OH* chemiluminescence intensity of ten percent of maximal signal intensity was detected as described by [185]. In the simulation, both ID and LOL were defined by an arbitrary maximal temperature of 1600 K
as employed in [186]. The calculated ignition delays and lift-off lengths exhibited a weak dependence on the threshold temperature as all cases considered have 21 percent oxygen and the rate of maximal temperature rise as well as the temperature gradient in axial direction at the lift-off location is very steep.

Test cases considered in this work are subdivided into non-reactive and reactive as summarized in Table 5-1 and Table 5-2, respectively. For a detailed description of the measurements the reader is referred to [183] and [185].

Non-reacting measurements (using nitrogen instead of air) have been conducted for three different ambient densities at 400 and 900 K ambient temperature. The macroscopic spray morphology through shadow images is compared with the liquid phase of the simulation. Reactive test cases have been carried out at the higher ambient density only (33 kg/m\(^3\)) and a wide variation of ambient temperature between 730 and 910 K is studied here. Measurements results are reported in terms of ID time, ignition location and LOL.

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<th>Table 5-1: Test cases considered for non-reacting conditions.</th>
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<td><strong>Gas density [kg/m(^3)]</strong></td>
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<th>Table 5-2: Test cases considered for reacting conditions.</th>
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5.2 Numerical considerations

Numerical simulations have been carried out with the same methodology as described in Section 3 and only relevant test-facility specific items are presented here. The SCC geometry is illustrated in Figure 5-2. The entire CFD grid consists of 1.25 million cells. The red arrow indicates the location and direction of the fuel injector. In the combustion chamber a hexahedral mesh is employed and the region relevant for the spray has been locally refined as illustrated by the green block. The first 300 mm axially and 50 mm radially from the injector, has a perfectly hexahedral grid of 2 mm edge length. Furthermore, the first 150 mm axially and 25 mm radially have been refined to 1 mm cell size. Grid sensitivity has been reported in [187] for non-reactive simulations. Spray penetration length and cone angle have been validated for a broad range of conditions, including variations in ambient density and temperature. For non-reacting conditions a grid resolution of 2 mm was found appropriate, however for reacting conditions a mesh size of 1 mm revealed an improved description of the flame structure as put forward in [186]. Consequently, results for non- and reactive conditions are shown with 2 and 1 mm grid size, respectively.

A two-dimensional CMC grid was employed; collapsing one dimension by means of a PDF weighted averaging in the axial direction of the disk shape representing the cylinder at TDC. The CMC grid is aligned with the spray axis and employs 42 by 43 cells, clustered in the spray vicinity, resulting in a spatial resolution of 4 mm and 1 mm in the nominal spray axial and radial direction respectively. The flow field time step was kept constant at 5.0x10^{-6} s and for the CMC equations a full operator splitting between transport in physical and mixture fraction space and chemistry is employed as described in [134, 137] with 10 sub-steps.

![Figure 5-2: Numerical domain of the Marine Spray Combustion Chamber consisting of inlet (left), regenerator and flange (middle) and combustion chamber (circle). The red arrow indicates the location and direction of fuel injector. The green region represents the refined region of interest.](image-url)
Flow field initialization (temperature, pressure, velocity, turbulent kinetic energy and turbulence dissipation rate) at start of injection (SOI) are obtained from previous simulations [188, 189]. The swirling flow in the combustion chamber is provided by a tilted intake and at SOI it corresponds roughly to a solid body rotation with a circumferential velocity of 20 m/s at 200 mm radius except close to the wall where a no-slip condition is present. Standard wall-function formulation is used to treat heat transfer at the wall in conjunction to a constant-temperature boundary condition \( T_{\text{wall}} = 453 \text{ K} \).

For liquid droplets, thermo-physical properties of \( n \)-dodecane as a function of temperature have been adopted. No time resolved mass flow rates for the fuel injection were available. The injection rate profile has therefore been estimated from the pressure drop across the injector based on experimental data from a pressure sensor installed in the injector body upstream of the needle seat and taking into account the large sac volume typical of marine injector tips.
5.3 Non-reactive conditions

Atomization and secondary break-up model performance are evaluated under non-evaporating conditions in order to suppress droplet mass transfer. Figure 5-3 shows spray penetration length for three different gas pressures. The increase in penetration when reducing the gas density is correctly captured. For the lower ambient pressure the computed initial spray length is underestimated until approximately 0.3 ms and afterwards it slightly over-shoots the measured line. Overall, the good agreement confirms the appropriateness of the mesh resolution and spray model constants.

The upper two sprays in Figure 5-3 compare simulated and measured shadow images under non-evaporating conditions at 5 ms after SOI and 40 bar ambient pressure, respectively whereas the lower two sprays are under evaporating conditions at the same time and ambient density. At the lower temperature the simulation reproduces well the macroscopic spatial extent of the liquid droplets. No information about droplet size distribution is available to date. Interesting to note is the lateral deviation due to the swirling ambient gas. Roughly 200 mm downstream, deviation in the opposite direction occurs, since swirl direction is reversed compared to the spray axis. This effect is well reproduced by the simulation.

![Figure 5-3: Comparison simulation (dotted lines) with experiment (straight lines) of penetration length under non-evaporating conditions for three different gas pressures: 13 bar (blue), 27 bar (green) and 40 bar (red).](image-url)
For the evaporating case (lower two sprays) the measurement reveals a quasi-complete evaporation of the fuel after approximately 130 mm and the liquid length fluctuates in time as reported by von Rotz et al. [183]. The simulation predicts a complete evaporation after roughly 160 mm representing a fair agreement. However, the shape of the computed liquid region is considerably narrower compared to the measured liquid spray spread. This effect is particularly pronounced at 75 mm axially and downstream wards. The deviation of the numerical liquid parcels acted by the swirling flow seems to be overestimated.

Figure 5-4: Spatial evolution of shadow images under non-evaporating (upper two) and evaporating (lower two) conditions with 33 kg/m$^3$ gas density and 5 ms after SOI. Simulation at the top and experiment at the bottom.
5.4 Reactive conditions

5.4.1 Ignition process

As shown in Figure 5-5, the ambient temperature has a profound impact on the ignition delay. This trend is expected and is widely consistent with studies using smaller injectors, e.g. [173]. For the large range of temperatures here the ignition delays vary by one order of magnitude.

The computed ignition delay times are slightly overpredicted for all cases apart from the 910 K case and quantitatively the discrepancy was within 9 and 23 percent, whereas the standard deviation of the experimental values amounts to between 8 and 18 percent.

In addition to ignition delays, the ignition location has also been compared, which is defined as the axial distance from the injector where the temperature first exceeds 1600 K. Analogous to the time, the ignition distance increases substantially by diminishing the ambient temperature as illustrated in Figure 5-6 (lower). The simulation is capable to capture this trend well. The sensitivity of the location is high especially at the lower temperatures as was seen in [185], where it was further reported that once first ignition occurs, the spray ignites in a good portion of the spray within one image frame (62.5 μs).
Results II: Wärtsilä marine spray combustion chamber

Figure 5-5: Ignition delay for different ambient temperatures.

Figure 5-6: Lift-off length (upper) and ignition location (lower) as a function of ambient temperature.
Figure 5-7 presents the computed ignition sequence by means of temperature distribution in a vertical section along the nominal spray axis for the test case at 760 K. The black lines denote the stoichiometry mixture fraction isoline ($\xi_{st}$). In agreement with the experiment, in all test cases ignition takes place on the right side of the spray due to the swirl motion. The effect of increased radial distance of the ignition location from the injector axis with reduced temperature is also captured (not shown here).

Ignition occurs at a slightly fuel lean mixture fraction, which is in contrast with conclusions drawn in [137] reporting the most reactive mixture fraction for sprays in a diesel engine to be on the rich side at a mixture fraction value of around 0.1. A possible explanation for this different behavior could be due to the swirl, which increases the convective transport from the rich spray core toward the fuel lean spray contour, possibly leading to a lean shift of the ignition mixture fraction. However, further investigations are needed to clarify the observed dynamics.

![Figure 5-7: Temperature isosurface evolution at the time of ignition for the 760 K case (times in ms after SOI, $\xi_{st}$ denotes the stoichiometric mixture fraction isoline and the green arrow indicates the swirl direction).](image)
5.4.2 Lift-off length

After ignition the flame quickly develops upstream along the stoichiometric mixture fraction and subsequently stabilizes at a statistically steady distance from the injector, the so-called lift-off length, for which a comparison is shown in Figure 5-6 (upper). The simulation reproduces the correct trend and the quantitative values are within the experimental standard deviation. In the last decades various theories have been proposed to explain the mechanism of flame lift-off stabilization consisting mainly of autoignition, extinction, propagation and triple flame theory as reviewed by [4] for gaseous jets and by [190] for Diesel jets. The most prominent mechanism under diesel engine conditions is auto-ignition following considerations put forward in e.g. [173-175]. For the present setup, the computed scalar dissipation rate at the lift-off location is considerably lower compared to values found for smaller injectors, predominantly due to substantially lower spatial gradients of mean mixture fraction. Simulation results showed values below 1 s\(^{-1}\) for the scalar dissipation rate, compared to several hundred s\(^{-1}\) as reported in [191] for small nozzle orifices.
5.4.3 Flame structure

Since the injection duration is very long (25 ms), the spray reaches a quasi-steady characteristic close to the injector. The flame structure is analyzed in detail for the case at 790 K, which represents a medium-low temperature for Diesel engines. Figure 5-8 shows an instantaneous section plot through the injector axis at 10 ms after SOI for different scalars, from left to right: mean mixture fraction, temperature, species mass fractions of OH, oxygen and acetylene. A typical spray structure is found in the rich region where the oxygen is consumed and soot is formed and peak OH and temperature are around stoichiometry. The lateral deviation of the spray to the right caused by the swirl is clearly visible and the direction is reversed at roughly 220 mm from the injector because of the injector position and orientation. In the computed temperature distribution it can be observed that the spray burns prevalently in the direction of the swirl, as confirmed by observations drawn in [181, 185]. Elevated OH mass fractions are found in high temperature regions in agreement with observations for \( n \)-heptane sprays with an injector size of 0.100 mm (cf. Section 4). The spray structure normalized by the nozzle diameter \( d_0 \) shows close analogies with sprays with considerably smaller injector nozzles as reported i.e. at the Sandia constant-volume vessel, where measurements with orifice diameter within 0.100 and 0.363 mm have been reported in [192]. At the SCC the computed normalized mixture fraction distribution shows similar values and distribution as illustrated in [166] for 0.100 and 0.180 mm.

Figure 5-8: Isocontours of mixture fraction, temperature, OH, \( \text{O}_2 \) and \( \text{C}_2\text{H}_2 \) species mass fractions. Isolines of the stoichiometric mixture fraction \( \xi_{st} \) are superimposed in white or black.
One of the main impacts of a larger injector size is the fact that spray liquid length extends further than the flame lift-off height, since the length of the dense spray core depends linearly on the orifice diameter while the lift-off was seen to scale only to the power of 0.34 as reported in [192]. This effect can be clearly observed in Figure 5-8 in the temperature and oxygen distributions. Due to the presence of burning droplets inside the flame, the conditions present are extremely suitable for soot formation, as indicated by the acetylene concentration which represents a soot precursor and is a marker of where soot is formed and soot particle surface growth occurs. The computed acetylene distribution corresponds, as expected, to the entire fuel rich region from the lift-off location and downstream. This is in conceptual agreement with observations reported in [185] where spectroscopic analysis revealed a significant contribution of thermal soot radiation at the conventional OH* wavelength in the entire flame region. On the contrary, for smaller injectors, a separation between the lift-off location and the soot region has been observed in [173] because of complete evaporation of the spray before the lift-off location is reached. A higher LOL corresponds to enhanced air entrainment and as a consequence a lower equivalence ratio at the LOL position, resulting in less soot.

Figure 5-9: Computed temperature distribution 7 ms after SOI for four different ambient temperatures. The stoichiometric mixture fraction isoline is indicated in gray.
The temperature isocontour at 7 ms after SOI for the four test cases is shown in Figure 5-9. The peak temperatures are comparable for all cases (differences of roughly 50 K) as a consequence of the same stoichiometric mixture fraction due to identical oxygen contents and the low to moderate values of scalar dissipation rate. As the ambient temperature is decreased, the high temperature region at around stoichiometry becomes increasingly broader due to the different mixture fraction distributions caused by altered evaporation behaviour. The simulation showed an increased maximal value of mean mixture fraction for higher air temperatures, covering the value range between 0.25 and 0.4 (not shown here). A lower ambient temperature results in a higher pre-combustion evaporation due to the larger ID and LOL and, as a consequence, a reduced portion of combustion-induced evaporation (burning droplets). The latter tends to increase evaporation rate and consequently the peak mean mixture fraction, reducing the flame volume. Interesting to note is that the simulation revealed a reactive-evaporative balance in the core of the flame as the liquid length is generously larger than the lift-off length. The effect of the droplet terms investigated in [132] are expected to become more pronounced as the droplet-flame interaction is guaranteed. Further work is needed to clarify the importance of such effects.
6 Results III: Sandia heavy-duty diesel engine

6.1 Experimental test facility

Experimental data available from the optically accessible heavy-duty diesel engine installed at Sandia National Laboratories [193] has been used for model validation. The measurements were obtained on a single-cylinder, common-rail, direct-injection heavy-duty diesel engine based on a Cummins N-series with 139.7 mm bore and 2.34 liters displacement. A schematic of the engine and the optical setup for soot measurements is illustrated in Figure 6-1 and the main specifications of the engine and injector are summarized in Table 6-1.

Figure 6-1: Schematic of the heavy-duty diesel engine with optical setup soot for soot measurements (adopted from [193]).
To minimize thermal loading the engine was operated skip fired where every tenth cycle was fired. The injector has eight equally spaced orifices with a nominal diameter of 0.196 mm. An ultra-low sulphur diesel fuel was employed for which the specification is given in [168]. Optical access is provided by an extended piston and a flat piston-crown window for signal collection as well as windows located around the top of the cylinder-wall for laser-based imaging diagnostics. For a detailed description of the engine specifications and diagnostics employed the reader is referred to [168] and references therein. In-cylinder soot mass has been measured with the two-color thermometry as shown in Figure 6-1.

In this study, all five ‘reference’ cases reported in [193] have been considered (cf. Table 6-2). These consist of two high temperature combustion (HTC) cases with different ignition delays, two low temperature (LTC) cases with different start of injection and a LTC case with split injection. HTC and LTC refer to the different adiabatic temperatures as a consequence of the oxygen concentration in the oxidizer caused by dilution. The latter is normally achieved by exhaust gas recirculation; for this engine, pure N₂ is employed due to the skip-fired operation. The two HTC conditions correspond to ‘classical’ diesel combustion, which is characterised by a premixed combustion phase followed by mixing controlled energy conversion; the respective modal split for these two cases is 32 %/68 % and 85 %/15 %.

<table>
<thead>
<tr>
<th>Table 6-1: Engine and injector specifications.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Engine type base</td>
</tr>
<tr>
<td>Swirl ratio</td>
</tr>
<tr>
<td>Bore x Stroke [mm]</td>
</tr>
<tr>
<td>Bowl width, depth [mm]</td>
</tr>
<tr>
<td>Displacement [L]</td>
</tr>
<tr>
<td>Geometric compression ratio</td>
</tr>
<tr>
<td>Fuel injector type</td>
</tr>
<tr>
<td>Fuel</td>
</tr>
<tr>
<td>Number of holes</td>
</tr>
<tr>
<td>Spray included angle</td>
</tr>
<tr>
<td>Nozzle orifice diameter [mm]</td>
</tr>
<tr>
<td>Nozzle orifice L/D</td>
</tr>
</tbody>
</table>
The LTC cases have been chosen to represent three alternative combustion concepts proposed in the literature, all of which attempt to simultaneously reduce soot and NOx, namely the so-called “Premixed lean diesel combustion”, the “Modulated Kinetics” approach as well as the “UNIBUS” concept. For references and further discussion to these combustion strategies the reader is referred to [193].

All cases have been run at a constant engine speed of 1200 RPM. Due to the profound differences in both oxidizer reactivity as well as ignition delays, the operating conditions span between almost fully premixed type combustion to a combination of premixed/diffusion combustion modes. As a consequence, this dataset sets very high requirements for the numerical model.

### Table 6-2: Engine operating conditions considered.

<table>
<thead>
<tr>
<th></th>
<th>HTC-short</th>
<th>HTC-medium</th>
<th>LTC-early</th>
<th>LTC-late</th>
<th>LTC-double</th>
</tr>
</thead>
<tbody>
<tr>
<td>O₂ conc. (vol. %)</td>
<td>21</td>
<td>21</td>
<td>12.6</td>
<td>12.6</td>
<td>12.6</td>
</tr>
<tr>
<td>Speed (rpm)</td>
<td>1200</td>
<td>1200</td>
<td>1200</td>
<td>1200</td>
<td>1200</td>
</tr>
<tr>
<td>IMEP (bar)</td>
<td>4.4</td>
<td>4.5</td>
<td>3.9</td>
<td>4.1</td>
<td>4.5</td>
</tr>
<tr>
<td>TDC motored temp. (K)</td>
<td>905</td>
<td>800</td>
<td>870</td>
<td>840</td>
<td>870</td>
</tr>
<tr>
<td>TDC motored density (kg/m³)</td>
<td>24</td>
<td>22.3</td>
<td>22.9</td>
<td>22.5</td>
<td>22.9</td>
</tr>
<tr>
<td>Rail pressure (bar)</td>
<td>1200</td>
<td>1200</td>
<td>1600</td>
<td>1600</td>
<td>1600</td>
</tr>
<tr>
<td>SOI (°ATDC)</td>
<td>-7</td>
<td>-5</td>
<td>-22</td>
<td>0</td>
<td>-22,+15</td>
</tr>
<tr>
<td>DOI (°ATDC)</td>
<td>10</td>
<td>10</td>
<td>7</td>
<td>7</td>
<td>4, 4</td>
</tr>
<tr>
<td>Fuel mass (mg)</td>
<td>61</td>
<td>61</td>
<td>56</td>
<td>56</td>
<td>31, 33</td>
</tr>
</tbody>
</table>
6.2 Numerical considerations

A sector mesh with an angle of 45 degrees has been considered, as the injector consists of eight equally spaced orifices. Simulation of the intake port and flow past the valves has not been attempted. Instead, a flat cylinder head has been considered as illustrated in Figure 6-2 at TDC and the simulation commences at the time of intake valve closure (IVC) with prescribed values for pressure, temperature, swirl and turbulence quantities. The latter have been estimated based on the experimental data. For this low-swirl engine, the swirl number is 0.5 and is initialized assuming a solid-body rotation, while the turbulent kinetic energy and length scale at IVC were prescribed as one percent of mean piston speed and 10 percent of cylinder bore as common practice [22, 194, 195].

Figure 6-2 depicts exemplarily a section through the cylinder axis with the more resolved CFD grid and the CMC grid overlaid in red at top dead centre (TDC). The CFD resolution in the piston bowl is around 1 x 1 x 1 mm\(^3\) resulting in 50,000 cells at TDC. In the sweep volume a cell size of 1 x 1 x 1.3 mm\(^3\) with 1.3 mm in axial direction is considered resulting in 240,000 cells at Bottom Dead Centre (BDC). This corresponds, to the authors’ best knowledge, to the finest mesh to date presented in the literature for the Sandia heavy-duty optical engine in a RANS context. For the same simulation setup, in most of the works a mesh size of 1.2-2.4 mm is employed, e.g. [22, 46, 107, 194, 196, 197].

![Mesh of engine geometry at top dead centre. In black is the CFD grid with the CMC grid overlaid in red.](image)
The quantities at the lower CMC resolution are obtained by means of PDF weighted averaging of the corresponding quantities at the higher CFD resolution. A two-dimensional formulation denoted 2D-CMC is followed here in which cells in azimuthal direction have been collapsed. I.e. in circumferential direction, all CFD information is reduced into one single CMC cell and the CMC equations are solved in the radial and axial dimensions. In the piston bowl region a 1 mm by 2 mm CMC resolution is considered in cylinder axial and radial direction, resulting in a 35 x 20 CMC mesh at TDC. During the expansion stroke CMC layers are added in analogy to CFD cell layers as proposed in [137, 138]. Cell coordinate interpolation is applied whenever needed during layer addition/deletion.

Note that the modeled crevice length has been reduced, instead the width has been increased in order to maintain the crevice volume the same. The mixture is not expected to enter in the crevice volume as only low-load operating conditions are considered.
6.3 Validation soot model for reference cases

6.3.1 Mixture formation

Non-reactive simulations have been first assessed in order to minimize uncertainties deriving from spray development and evaporation. The fuel vapour distributions have been compared with a non-reactive case (pure N\textsubscript{2} ambient) with the same SOI and fuel pressure as the LTC late injection case, however fuelled with a primary reference fuel blend of 71 % \textit{n}-heptane and 29 % \textit{i}-octane (by volume). Fuel PLIF measurements with toluene as a fuel tracer have been performed in [198], where laser sheets normal to the cylinder axis have been considered at three different distances below the fire deck (7, 12 and 18 mm). Results are presented in terms of equivalence ratio for an equivalent of 12.6 percent oxygen volume fraction as shown in Figure 6-3, where two time instants are illustrated: 7 and 12 degrees ATDC. The first corresponds roughly to the EOI and the second to the peak of premixed burn of the LTC late case. The peak of equivalence ratio in the figure scale drops from 3.7 at 7 °CA down to 1.4 at 12 °CA due to mixing.

![Figure 6-3: Experimental isolines (left column) and computed isocontours (right column) of equivalence ratio in three planes orthogonal to the cylinder axis located 7mm (upper), 12 mm (middle) and 18 mm (lower) below the cylinder head at two time instants 7 °CA (left) and 12 °CA (right).]
At 7 °CA large parts of rich regions are present along the spray axis, predominantly in the upper layer and with a decreasing portion of fuel concentration in the lower region. Later, rich regions are present in the lowest layer and close to the piston bowl in the upper two planes. The simulation reproduces qualitatively these processes with a clear impingement of the spray at the bowl edge and a leaning of the mixture between the time instants. However, the simulation generally predicts a narrower fuel region and richer mixtures within the spray. The mixing of fuel with the ambient is underestimated, resulting in a more stratified mixture. Due to the narrow injector angle (14°), the fuel distribution was observed to be considerably sensitive to the distance from the cylinder head. Small changes in distance have a large influence in the planar fuel distribution.

6.3.2 Pressure traces and AHRR

6.3.2.1 HTC cases
Pressure traces and corresponding apparent heat release rates for the two HTC cases are shown in Figure 6-4 (upper): cylinder pressure evolutions between intake valve closure and SOI are captured accurately with discrepancies within 0.2 bar at SOI the highest. The cases are distinguished by the ambient temperature at TDC (905 and 800 K) and therefore the different ignition delays, resulting in a different partition between premixed and diffusion combustion. The short ID case is the conventional diesel operating condition, resulting in roughly 70 % of fuel conversion occurring in diffusion mode [193]. On the contrary, the long ID burns predominantly in premixed mode.

The simulation was able to reproduce the ID well for both cases. The discrepancy was around 0.5 degrees CA for both cases. For the short ID case the computed portion of premixed combustion is underestimated and as a consequence the diffusion part is larger. The peak pressure resulted higher by approx. 2 bar. For the long ID point the premixed combustion was well captured apart from the last small part of AHRR, where energy was converted in diffusion mode. This is possibly due to the overestimation of the mixture stratification after injection is over, leading to a late conversion of rich zones which take time to mix with oxidizer.
Figure 6-4: Comparison of pressure trace and AHRR with experimental data for all 5 reference operating conditions. High temperature (upper) and low temperature cases (middle) with early (left) and late (right) injection; low temperature split injection case (lower). The dashed lines denote the fuel injection rate in arbitrary units.
6.3.2.2 LTC cases
Both LTC cases are almost pure premixed conditions due to the positive ignition dwell. The model was capable to reproduce ID accurately for both cases. Here, the cylinder pressure increase is underestimated because of the slightly lower computed peak AHRR and a part of the energy is released in a diffusion mode. For the late injection operation the AHRR was well captured with a marginal higher peak AHRR.

6.3.2.3 Double injection
For the double injection cases the injected mass was subdivided into two injections with nearly the same fuel mass. The timing of first injection corresponds to the LTC early (-22° CA) case and the second is considerably delayed (+15° CA). For this reason the spray from the first injection event is expected to be completely burned at the time when the second fuel injection starts as confirmed by the AHRR evolution in Figure 6-4 (lower). As a consequence, the second spray is influenced by the first one only in terms of initial ambient temperature and composition. As expected, the ID of the first injection event is well captured and the predicted AHRR agrees with the experimental findings as was the case for the LTC-early condition. The peak pressure is lower than the experiment. For the second injection the simulation predicts a shorter ID and an almost pure premixed mode energy conversion. As a consequence the pressure increase caused by the second injection is advanced. This is possibly caused by a less stratified mixture formation during the second fuel delivery.
6.3.3 Flame structure

In the following, the flame structure for two different oxidizer dilutions is considered. Modeled OH species distribution in a cut plane along nominal spray axis is compared with OH-PLIF measurements from [169]. Comparisons are presented for a selected time instant for the HTC-short case at 0° CA (upper) and for the LTC-early case at -8° CA ATDC (lower) as shown in Figure 6-5. The timings correspond roughly to the end of premixed combustion for both cases. OH-PLIF was excited at 284.01 nm using a Nd:YAG laser and the fluorescence was observed near 310 nm. In order to quantify interfering fluorescence from other species excited at the same wavelength (e.g. PAH), a slightly different wavelength (283.90 nm) has also been measured, however in a different engine cycle. Therefore, the online (284.01 nm) and offline (283.90 nm) images cannot be subtracted and are hence both drawn in Figure 6-5. The main difference between measured OH-PLIF distributions is the fact that the HTC-short case is characterized by a thin OH layer typical for conventional diesel combustion. It should be noted that in the experiments the OH-PLIF signal is strongly attenuated by soot presence and, as a consequence, only the spray tip is observable. Perhaps, a diffusion flame and OH presence is also supposed to extends upstream as concluded in [159]. On the other hand, in the LTC-early case OH is found throughout the entire spray cross section, mainly due to the long ignition delay causing an over-mixing of the mixture prior to ignition.

![Figure 6-5: Comparison OH-PLIF for HTC-short at 0°CA ATDC and LTC-early at -6°CA ATDC](image)

**Table 6-1:** Comparison OH-PLIF for HTC-short at 0°CA (upper) and LTC-early at -6°CA ATDC (lower). Images correspond to experimental online (left) and offline (middle) and simulation (right).
The thin OH layer is in qualitative agreement with the measurement. For the diluted case the model computes a more homogeneous distribution of OH with a much larger characteristic flame thickness. Nevertheless, the mixture stratification for the simulation is higher than the measurement. This is a possible cause for the lower portion of energy released in premixed mode as observed in the AHRR evolutions (cf. Figure 6-4).

In the following, the flame structure is further analyzed using simulation results only. The influence of oxidizer dilution is studied comparing the HTC-short and the LTC-late test cases at three distinct case-specific characteristic time instants: early soot, peak soot and late soot. Times have been chosen based on model predictions for soot mass evolution as will be presented later in section 6.3.4.3 (cf. Figure 6-12 and Figure 6-15) and correspond to 0, 7 and 15 °CA ATDC for the HTC-short and 15, 24 and 35 °CA ATDC for the LTC-late case.

The influence of EGR is illustrated with the help of three different sorts of diagrams: first, spatial distributions of various scalars relevant for soot formation are displayed separately in a vertical section through the injector axis (Figure 6-6 and Figure 6-8). Quantities shown are from top to bottom: mean mixture fraction, mean temperature, and mean mass fraction of O₂, OH, CH₂O and C₂H₂. The time instants evolve from left to right and the superimposed black lines represent the stoichiometric mixture fraction iso-contour. Note that for the later time instants the cylinder volume is considerably increased due to piston movement during the expansion stroke. The second graph depicts temperature-equivalence ratio (T-Φ) diagrams (Figure 6-7 and Figure 6-9) at the same time instants. The temperature (black) is drawn for every CFD cell within the computational domain, whereas CH₂O (blue), OH (red) and C₂H₂ (green) are displayed only for species mass fraction levels higher than 45 % of their own instantaneous maximum within the domain. Third, the same vertical section as above is considered and CH₂O (blue), OH (red) and C₂H₂ (green) species distributions have been visualized within the same diagram (Figure 6-7 and Figure 6-9). The colour has been averaged in regions where species spatially overlap.
6.3.3.1 HTC case

In general, the main influence of the oxidizer dilution is the reduction of the reactivity resulting in an increase of ID and a reduction of the adiabatic flame temperature. With dilution the peak temperature drops from approximately 2600 to 2000 K. Maximal values of OH, CH$_2$O and C$_2$H$_2$ mass fraction are reduced by a factor of roughly 15, 6 and 3 respectively. The spatial extents are also drastically altered.

At HTC conditions a thin diffusion flame is developed as clearly visible from the OH distribution. After autoignition but still during injection (early soot), CH$_2$O is mostly located in the fuel rich region in the spray core, where the oxygen is not consumed yet. Acetylene, as expected, remains the entire time exclusively in the fuel rich region. Later, at the timings of peak (center) and late (right) soot, the evolution of chemical species and temperature is mainly governed by scalar mixing and energy is released in a diffusion mode as evidenced by the AHRR trace (cf. Figure 6-4). Oxygen as well as CH$_2$O are almost completely consumed for rich mixtures. The mixing is described by the mixture fraction evolution, and at the later time, fuel rich regions persist only at the bowl edge and therefore the soot is progressively oxidized due to oxidizer entrainment into the sooty region.

The gradual transition from partially premixed to non-premixed combustion mode is well visible in the T-Φ diagram. At early soot time, the temperature is more scattered and most of the CH$_2$O is in the fuel rich region. At later stages the temperature is increasingly more ordered as typical for non-premixed flames. There is still a certain temperature scattering, and this is mainly due to the description of the mean temperature with the mixture fraction PDF within the CFD cell. For the fully developed diffusion flame, CH$_2$O is present in a well confined region at an equivalence ratio up to 0.3, corresponding to the outer part of the spray as illustrated in the conceptual spray structure. The effect of scalar mixing is evident from the temporal reduction of the maximal equivalence ratio within the domain varying from 4 to 1.5 between early and late soot time.
Figure 6-6: Spatial distribution of soot relevant quantities in a vertical section through the injector axis for the HTC-short test case at three different times: early, peak and late soot. From top to bottom: mean mixture fraction, mean temperature, mean mass fraction of O$_2$, OH, CH$_2$O and C$_2$H$_2$. The black lines denote stoichiometric iso-contour.

Figure 6-7: Conceptual spray structure (upper) and T-Φ diagram (lower) for the HTC-late case at early soot (left), peak soot (centre) and late soot (late). Quantities shown are: mean temperature (black), C$_2$H$_2$ (green), OH (red) and CH$_2$O (blue).
6.3.3.2 LTC case

Beside the lower combustion temperature and species concentrations, EGR enables a major fuel/oxidizer mixing prior to combustion due to the longer ignition delay time. It results a more homogeneous mixture distribution and at later stages a clear spray structure is not anymore recognizable. Due to the lower stoichiometric mixture fraction, fuel rich regions still persist until the latest time instant. Considering the variation of the stoichiometric iso-line between peak and late soot time, it seems that a larger fuel rich region is retrieved. This is due to swirl motion in conjunction with the recirculation of the spray tip when colliding at the bowl. Note that the LTC-late case reaches the characteristic soot timing later than the HTC-short and therefore the expansion stroke is much more pronounced.

The conceptual differences between the LTC-late and the HTC-short cases are well visible in the T-Φ diagram. The larger time for mixing reduces the peak equivalence ratio. At the early soot time there are vast Φ regions with CH₂O at low temperature. CH₂O is a marker for the low temperature ignition and it is rapidly consumed during high temperature ignition, coinciding with OH formation. Experimental findings revealed CH₂O and OH to exhibit a complementary distribution [74], i.e. CH₂O at low and OH at high temperature respectively. This effect is well visible considering the first two time instants. The early soot time corresponds to a partially premixed mode. There is a large portion of regions where high temperature ignition haven’t taken place yet and when it happen, CH₂O (blue crosses) “jump” upstairs to form OH (red). For the fully developed flame structure at late soot time (right), OH has the same spatial extent in the T-Φ diagram as the HTC case. On the contrary, CH₂O extents up to Φ=0.6 and a much wider area is occupied compared to the undiluted case.

With high EGR levels the amount of unburned hydrocarbons (UHC) is generally increased [199], for which CH₂O is an indicator for incomplete oxidation. The structure of the spray in physical space is wider but conceptually the same as the case without EGR with a stratified arrangement of C₂H₂, OH and CH₂O from the inner to the outer part, respectively.
Figure 6-8: Spatial distribution of soot relevant quantities in a vertical section through the injector axis for the LTC-late test case at three different times: early, peak and late soot. From top to bottom: mean mixture fraction, mean temperature, mean mass fraction of O₂, OH, CH₂O and C₂H₂. The black lines denote stoichiometric iso-contour.

Figure 6-9: Conceptual spray structure (upper) and T-Φ diagram (lower) for the LTC-late case at early soot (left), peak soot (centre) and late soot (late). Quantities shown are: mean temperature (black), C₂H₂ (green), OH (red) and CH₂O (blue).
6.3.4 Soot formation

6.3.4.1 Sensitivity soot model constants

In this study a sensitivity analysis only for constants of the soot model has been considered, as soot is the main focus of this work. The performance of spray and chemical model was assessed in the previous sections separately.

The sensitivity of the soot model constants has been investigated by means of temporal soot mass evolution for the HTC-short case as it is the one with the highest in-cylinder soot formation. Soot related chemical reaction rates obey an Arrhenius-type expression. In the following a systematic variation of reaction rates was applied in order to obtain less soot, i.e. decreased soot inception and surface growth and increased oxidation by O₂ and OH. The pre-exponential factors and activation temperatures (cf. Table 3-2) have been changed by an arbitrary factor of two and by 10 percent respectively and the influences on in-cylinder soot mass are illustrated in Figure 6-10 in a normalized form. Note that only simulated curves are drawn without experimental data as here the relative changes of prediction are of interest.

For what concerns the pre-exponential factors, soot evolution was found to be most sensitive with respect to surface growth, where a decrease of peak soot mass by a factor of 2.5 is observed. The timing of soot maximum and relative soot formation and oxidation rate remained roughly unchanged. For soot surface growth at HTC-short condition an increase in activation temperature by 10 percent, as shown in Figure 6-10, and a decrease of the pre-exponential factor by a factor of 2 were found to have a comparable effect. On the other hand, reduced soot inception rate showed a marginal effect on peak soot mass, which is reduced by less than 6 percent. Since soot inception is the main source of soot particle number, a reduced inception rate increases the mean particle diameter (not shown here) and therefore, given the same soot mass, soot oxidation rate is slightly reduced due to the minor soot surface available for oxidation. With an increase in the oxidation rate by O₂ and OH the peak soot mass is reduced by 4 and 8 percent, respectively. An increase of soot oxidation assisted by OH anticipates the soot reduction by approximately one degree and oxidation by oxygen didn’t have an influence on soot timing.
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Figure 6-10: Influence of pre-exponential factor (a) and activation temperature (b) on soot in-cylinder mass for the HTC-short case.

Although this is a transient spray, these findings are analogous with soot sensitivity results reported for the constant-volume quasi-steady $n$-heptane spray investigated previously in Section 4.3.3.2. For the rest of this study the original reaction rates from Leung et al. [80] are used (cf. Table 3-2), since these values have proved to be very suitable for soot distributions of quasi-steady $n$-heptane sprays using the same methodology.
6.3.4.2 Algorithm for line-of-sight soot natural luminosity

Experimental natural luminosity, which is mainly dominated by thermal radiation of soot particles, is collected through the piston window by means of a high-speed camera, resulting in a line-of-sight integrated signal. Depending on the soot cloud thickness, part of the radiation emitted by soot particles is absorbed by the neighbouring soot. In the literature, simulation work carried out for the same engine as in this study usually compared the measured line-of-sight natural luminosity with the computed soot mass or volume fraction, i.e. [22, 31, 107]. In this study a transformation of the computed 3D soot distribution into a virtual 2D line-of-sight natural luminosity has been attempted in view of more consistent comparison with the experiment.

In this section the comparison methodology is briefly presented, which is analogous to the procedure reported in [194], which provides a 2D normalized natural luminosity signal, where relative changes of signal are of particular interest. Every CA the 3D simulation distribution of soot volume fraction and temperature is used for the data post-processing. For simplicity, luminosity is assumed to derive from soot particles only and radiation is assumed to be emitted exclusively in vertical direction and multiple scattering effects have been neglected. In every vertical column of CFD cells within the computational domain (i.e. analogous to the direction of the collection line-of-sight of the camera through the piston window), soot radiation is vertically integrated accounting for absorption caused by soot presence along the line-of-sight. The normalized natural luminosity CFD signal, $S_{CFD}$, for every vertical cell column is determined as follows:

$$S_{CFD} = \sum_{k=0}^{x} \left[ \sum_{v=1}^{N} \left( \varepsilon_{Soot}(f_v, L_k, \lambda) \cdot I_{\lambda,bb}(T_{Soot,k}, \lambda) \right) \cdot (1 - \varepsilon_{Soot}(f_v, L_K, \lambda)) \cdot \Psi(\lambda) \right]$$

(S56)

Soot emissivity, $\varepsilon_{Soot}$, is estimated in the same way as in [193] and for spectral blackbody radiation intensity, $I_{\lambda,bb}$, Planck’s equation is applied. The index $k$ corresponds to a CFD cell within a vertical column ordered with increasing distance from the piston window. Summation over $k$ results in a spectral luminosity signal, close the correlation presented in [194], where the same methodology was applied for two-color thermometry post-processing. Here, the signal is additionally summed over all wavelengths since the images are compared to natural luminosity measurements and multiplied by the normalized high-speed camera monochrome spectral response, $\Psi(\lambda)$, (IDT XS-4 Camera
Specification Manual). The wavelength domain was summed up from 300 to 1000 μm with steps of 1 μm. Images are shown in gray scale and a linear scaling is applied.

The influence of the quantities to be compared with experimental data is considered using the following three variants: (1) line-of-sight soot natural luminosity accounting for absorption, (2) without accounting for absorption and (3) line-of-sight averaged mean soot volume fraction.

As an illustrative example the soot distribution for the HTC-short case with the highest soot formation is used to emphasize the conceptual differences between the three comparison approaches. Figure 6-11 shows experimental luminosity and the three signals reconstructed from the simulation results at three selected time instants (4, 6 and 10° CA ATDC), which are located during soot formation, at peak soot and during soot oxidation.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a) Luminosity</td>
<td>(b) Luminosity with adsorption</td>
</tr>
<tr>
<td>4° aTDC</td>
<td></td>
</tr>
<tr>
<td>6° aTDC</td>
<td></td>
</tr>
<tr>
<td>10° aTDC</td>
<td></td>
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</tbody>
</table>

Figure 6-11: Comparison of soot natural luminosity distribution for the HTC-short case. From left to right: (a) experimental natural luminosity, (b) soot radiation with adsorption, (c) soot radiation without adsorption and (d) mean soot volume fraction.
It is evident, that the post-processing of the simulation results has a strong influence on the representative distribution: Images from experimental line-of-sight natural luminosity show a low intensity signal at the bowl edge on the nominal spray axis (horizontal direction). However, at this position a high soot concentration in the rich zone of the spray is expected. This is confirmed by the current simulation (see column d) and former studies in the literature for the HTC-short case, e.g. [22, 31, 107, 194]. The low luminosity reaching the high-speed camera is most likely due to optically-thick properties of the soot cloud and is mainly caused by soot radiation adsorption within the spray core. This effect is evidenced by the post-processing of the computed 3D soot distribution. Accounting for soot radiation adsorption along the line-of-sight (column b) the low signal at the bowl is retrieved. This is not the case if adsorption is neglected (column c). The proposed post-processing methodology hence represents a clear improvement of the conceptual comparison procedure. For the rest of this study the measured line-of-sight natural luminosity is compared with the computed luminosity accounting for adsorption.
6.3.4.3 HTC cases

In this section the temporal evolution of soot total mass is presented. Note that plots are labeled with soot volume to be consistent with original measurements quantification from [193]. However, throughout the paper the argumentations are conducted in terms of soot mass, since the soot density is assumed to be constant (2000 kg/m³). Again, results are divided into HTC, LTC and double injection cases. It should be noted that a quantitative soot mass comparison has been presented. This was not the case for most of the studies in the literature at the current diesel engine comparing soot in a normalized form and even omitting the normalization factors, e.g. [22, 31, 107]. As the two-color thermometry is a line-of-sight measurements technique, it is particularly subjected to uncertainties correlated with line-of-sight gradients predominantly in temperature, and to a lesser extent also in soot concentration. As discussed in [194] this results in a bias towards hot soot properties and errors in soot volume fraction may rise up to 50 percent. This should be kept in mind when comparing quantitative in-cylinder soot masses.

Temporal and spatial soot evolutions for the two HTC cases are shown in Figure 6-12 (short ID) and Figure 6-13 (long ID). The increased ID results in a decrease of peak soot by a factor of 6.5, mainly caused by the enhanced leaning of the mixture prior to ignition. The simulation is capable to capture this effect, where the peak soot mass was overestimated by 4 and 23 percent for the short and long ID case, respectively. The timing of the computed soot mass peaks was also well reproduced, being delayed for both cases by approximately one degree compared to experimental data. The first soot appearance was found at the peak AHRR consistent with experimental findings. This is also confirmed by images of natural luminosity. For the short ID case first significant signal is found at -2° CA for both simulation and experiment, whereas for the long ID case the computed first soot is one degree later than the measurement, being the same time difference between the predicted and measured ignition delays. In both cases, the onset of soot coincides with the decrease in the premixed burn as mixing-controlled combustion commences. These findings are in agreement with the observations from [194] and more general findings reported earlier in [159]. For the long ID, the rate of net soot formation is also delayed; however the slope is very well reproduced. For the short ID case, soot formation at the beginning until 2° ATDC is weaker than in the experiment; later the net formation rate is comparable.
Figure 6-12: HTC-short case: In the centre evolution of in-cylinder volume (lines) and AHRR (dashed lines). Upper and lower spatial evolution of soot natural luminosity of experiment (upper row) and simulation (lower row). Note that the soot is not normalized.
Figure 6-13: HTC-long case: In the centre evolution of in-cylinder volume (lines) and AHRR (dashed lines). Upper and lower spatial evolution of soot natural luminosity of experiment (upper row) and simulation (lower row). Note that the soot is not normalized.
On the other hand, soot oxidation is clearly overestimated for both cases: the simulated characteristic net soot decrease rates, averaged during net soot oxidation period, were approximately twice as high as the measured values for both HTC cases.

Despite the quantitative total soot evolution, in the following, a more qualitative comparison is offered by the natural luminosity images. Due to the negative ignition dwell, cf. Figure 6-4 (a), the short ID case consists of a quasi-steady lifted spray until EOI at 3° CA, where downstream of the LOL a conventional diffusion flame is established [159]. During this period soot is present in almost the entire fuel rich cross section of the spray up to the LOL. For the current injector (0.196 mm orifice diameter) no clear spatial separation between LOL and soot is expected because the liquid length is as large as the LOL. For smaller injectors a separation was observed, e.g. in [75] with a 0.100 mm injector. After EOI (3° CA) there is an enhanced oxygen entrainment and over-mixing, in particular in the vicinity of the injector [179]. As a consequence soot in this region is rapidly oxidized and the last soot remaining is closer to the bowl edge where rich mixtures are still present. This effect is qualitatively reproduced by the simulation; soot in the first 30 mm radially between the injector and the bowl edge is progressively oxidized while closer to the bowl edge substantial levels of soot persist. This evolution is clearly visible in the time interval 4-10°CA.

The long ID case manifests a slight negative ignition dwell as can be seen from the AHRR and injection rates given in Figure 6-4 (b), therefore there is a limited period with an injection-soot overlap (only at 5°CA). At this time soot is present close to the injector while later it is found only downstream close to the bowl edge. The simulation correctly captures this phenomenon and predicts small amounts of soot close to the injector, although extending to somewhat later times (10° CA). For the remaining time instants (14-20°CA), the soot region is in qualitative agreement with the measurement where soot is only present close to the bowl edge. In addition, during the period 16-20°CA the strong decrease in signal intensity is qualitatively consistent with the measurement.

6.3.4.4 LTC cases
Figure 6-14 and Figure 6-15 display soot temporal and spatial evolutions for the LTC early and late injection cases. The peak soot mass for the LTC cases were under-predicted by a factor of 2 and 3.8 for the early and late case. These scaling factors have been applied in the diagrams to facilitate the comparison of the evolutions and are transparently provided in the respective figure legends.
Also for the LTC cases the first soot appearance immediately follows the peak AHRR. For the early injection operation the first soot is found at -8° CA in agreement with experiment, whereas for the late injection case simulation anticipates soot formation by two degrees as seen in the soot mass evolution. Despite the underestimation of the total soot mass, the normalized shape is in very good agreement along almost the entire formation and oxidation phases for both cases, revealing an accurate description of global soot formation and oxidation rate.

The inspection of the qualitative spatial luminosity development for the early case reveals that soot is first formed close to the bowl edge inside a circular area, which corresponds approximately to the stoichiometric to rich region. This is due to the relative importance of temperature for soot radiation and its adsorption as discussed in section 6.3.4.2. The simulation captures this behavior very well at -8 and -7° CA. Later, in the experiment, between -3 and +10° CA an isolated luminosity spot with a monotonic decrease in signal intensity is identified. The line-of-sight luminosity intensity calculated from the simulation results reproduces this phenomenon qualitatively well: after a quasi-circular sector dominated by quasi-stoichiometric luminosity between -5 to -3° CA, there is an increasing clustering of luminosity into a concentrated spot. Between 5 and 10° CA this is clearly visible from the mean of its position and the temporal decay of the intensity.

For the late injection case the soot spatial evolution is considerably different. The first luminosity signal is found on the side in correspondence to the so-called roll-up vortices as described in [168] and [180], where the lowest mixing rate is present. There are two main luminosity spots rotating in anti-clockwise direction around the cylinder axis due to swirl. During the formation phase the simulation is advanced by two degrees; the simulated 13° CA hence corresponds to the measured ones at 15° CA. Here it is visible that the first signal is on the side and there is no soot in the first 25 mm radially from the injector in agreement with the experiment. This large signal area persists until the late stages with a mild decrease in intensity. The simulation predicts a similarly large zone with a diffuse luminosity signal exhibiting qualitatively a very comparable decrease in time.
Figure 6-14: LTC-early case: In the centre evolution of in-cylinder volume (lines) and AHRR (dashed lines). Upper and lower spatial evolution of soot natural luminosity of experiment (upper row) and simulation (lower row). Note that the simulated soot is multiplied by a factor of 2.
Figure 6-15: LTC-late case: In the centre evolution of in-cylinder volume (lines) and AHRR (dashed lines). Upper and lower spatial evolution of soot natural luminosity of experiment (upper row) and simulation (lower row). Note that the simulated soot is multiplied by a factor of 3.8.
6.3.4.5 Double injection

The temporal evolution of total soot mass for the split injection case is presented in Figure 6-16 along both injection events. Soot is formed predominantly during the second injection. The simulation predicted a marginal soot formation during the first injection, a factor of approximately 30 smaller than during the second one. This is consistent with the marginal combustion luminosity deriving from the first injection event as reported in [197]. For the second injection, first soot luminosity is again detected at the peak AHRR. The simulation underestimates the ignition delay of the second injection and therefore the soot formation is anticipated by 4 degrees, the same interval between peaks of AHRR. The computed maximal soot mass was 25 percent lower than the experimental value and the peak was located two degrees earlier. This discrepancy is lower than for the other two LTC cases, where peak soot mass was underestimated by a factor of 2 and 3.8. Hence, a similar discrepancy would also be expected for the double injection case. However, as the model underpredicts ID of the second injection event soot is artificially increased due to the lower premixedness of the mixture at the time of ignition. In analogy to the LTC single injection cases, the slope during soot oxidation was very well captured.

Looking at the spatial evolution of natural luminosity, one may notice that the first two images (-8 and -5° CA) refer to the first injection event. Here, a small amount of luminosity close to the bowl edge can be observed. The second image exhibits a slightly higher intensity as is confirmed by the measurement. For the soot generated by the second injection, first luminosity is collected at -23° CA. In general, soot is present in the last 10-20 mm from the bowl edge distributed in azimuthal direction. Apart from a luminosity spot in the middle of the domain between 23 and 29° CA, the simulation predicts luminosity in the correct region with a relatively homogeneous distribution. Between 36 and 50° CA there is a slight convective transport of luminosity region from the bowl edge towards the middle of the domain. The simulation is capable to qualitatively reproduce this convective transport in conjunction with a decrease in signal intensity.
Results III: Sandia heavy-duty diesel engine

Figure 6-16: LTC-double case: In the centre evolution of in-cylinder volume (lines) and AHRR (dashed lines). Upper and lower spatial evolution of soot natural luminosity of experiment (upper row) and simulation (lower row).
6.3.5 Soot structure and discussion

In this section, a detailed analysis of soot related processes for two different cases are presented and analogies to empirical conceptual models for HTC and LTC are identified: for the latter, the suitability of n-heptane as diesel surrogate is questioned and discussed in the context of findings reported in corresponding studies for the same engine. For the HTC cases the observed overestimation of soot oxidation is further analyzed, offering possible explanations for the discovered discrepancies and the related model shortcomings.

6.3.5.1 Soot structure

In the following, two cases with different oxidizer dilutions have been selected to illustrate in more detail the flame structure and corresponding soot sub-processes: the HTC-short ID at peak soot mass (6° CA, cf. Figure 6-12) and the LTC-late injection during soot formation (15° CA, see Figure 6-15). Figure 6-17 displays quantities relevant for soot behavior (upper row, from left to right: mean mixture fraction, mean temperature, mean mass fraction of acetylene, oxygen, OH and soot volume fraction) and the relevant sources for soot mass fraction (lower row, from left to right: surface growth, oxidation by O₂ and OH, diffusion in mixture fraction, convection). The contours are shown for the plane along the nominal injector axis at the given time instants. The unconditional source terms are obtained by convoluting the conditional soot sources with the presumed β-PDF in every CFD cell. In addition the line-of-sight natural luminosity (lower row, right) is drawn. The comparison aims at providing insight with respect to the impact of the combustion mode on the in-cylinder soot-related sub-processes. The LTC case has been selected at 15° CA because later the spray structure is completely over-mixed and a clear spray structure cannot be recognized anymore; this is mainly due to the long ignition delay in conjunction with the swirl of the engine. The main difference between these two cases is the oxidizer dilution, which provokes a decrease in adiabatic flame temperature and a decrease in stoichiometric mixture fraction (\(\xi_{ST,HTC}\approx0.062, \xi_{ST,LTC}\approx0.036\)), which are illustrated by the dotted lines. The time instants considered correspond to 13 (HTC) and 15 (LTC) °CA after SOI. As can be expected, large differences in spray and flame structure are evident. As a consequence of the reduced flame temperature and the corresponding increase in ID due to the oxidizer dilution, the mixture formation is considerably affected: The mean mixture fraction for the LTC case shows lower peak values and a more homogeneous distribution caused by the enlarged time available for mixing prior to ignition.
Figure 6-17: Contours of quantities relevant for soot behaviour in a plane along injector axis for two cases: HTC-short (upper) and LTC-late (bottom). Upper row from left to right: mean mixture fraction [-], mean temperature [K], mass fraction of C2H2 [-], O2 [-] and OH [-], soot volume fraction [ppmv]. Lower row from left to right: soot mass fraction sources by surface growth [1/s], oxidation by O2 [1/s] and OH [1/s], eta-diffusion [1/s], convection [1/s] and soot natural luminosity [a.u.]. Dotted lines represent the stoichiometric mixture fraction. Note that the natural luminosity is integrated over line-of-sight.
For the HTC case the conventional thin diffusion flame along the stoichiometric iso-contour splashing at the bowl edge is retrieved. For the LTC case, two large roll-up vortices slightly rotated in anti-clockwise direction by the low swirl can be identified; this is consistent with experimental observations put forward in [180] for conditions similar to LTC late case.

With dilution the peak OH mass fraction is reduced by one order of magnitude and the flame characteristic thickness in higher as discussed in section 6.3.3. The equivalence ratio range where abundant levels of OH are present however remains unchanged; considerable OH concentration is found at $\Phi=1\pm0.5$, i.e. to both sides of the stoichiometric iso-lines, in agreement with considerations put forward in [168].

As expected, soot and its precursor acetylene are present in the fuel rich region only. Apart from the mean values, the examination of \textit{conditional} soot and acetylene mass fractions (not shown here) revealed peaks at an equivalence ratio of around 2 and 2.5 for both dilutions, i.e. soot is mainly formed at $\Phi=2-2.5$. Similar conclusions were also drawn in 4.3.2.2 for the quasi-steady sprays, where a detailed analysis of conditional budgets of soot formation was presented. Experiments offer additional support for these findings, evidencing that soot \textit{formation} is suppressed for $\Phi<2$, corresponding to the so-called \textit{threshold sooting conditions} [168]. The computed soot volume fraction peaks are approximately 50 and 0.6 ppmv for the HTC and LTC case, differing by almost two orders of magnitude. These values seem to be realistic, when viewed in context with the experimental data: Idicheria and Pickett [75] performed 2D-LII measurements with D2 fuel at HTC conditions for a 0.180 mm injector and a peak soot volume fraction of around 30 ppmv soot was reported [29]. For the LTC case a value of the order of 1 ppmv seems also plausible although for this case the simulation under-predicted peak in-cylinder soot mass by a factor of 3.8. The soot luminosity bias towards the hotter soot properties is well visible for both cases from the line-of-sight natural luminosity (lower row, right) and the effect of radiation re-absorption is also very important for both cases as shown in section 6.3.4.2.

The lower row in Figure 6-17 compares relevant sources for soot mass formation. As expected the surface growth is the main source and the corresponding area roughly coincides with the soot distribution, since it is a function of the soot surface area. At HTC conditions, soot oxidation by OH is found to be more important than the one driven by $O_2$ whereas at LTC conditions the opposite is true. This is mainly due to the substantial decrease by one order of magnitude in OH mass fraction with dilution discussed above.
Oxidation by OH acts more strongly within the fuel rich region compared to the one by O$_2$, because of the presence of the radical also at slightly rich mixtures close to the stoichiometric contour. From the conditional soot mass fraction profiles (not shown) it can be seen, that diffusion in mixture fraction tends to transport soot from rich towards stoichiometric conditions and this effect is balanced by soot oxidation as discussed in detail in Section 4.3.3.4. Soot convection, mainly the component along spray axis, counteracts soot surface growth transporting soot in the downstream direction. The influence of dilution on a variety of soot related quantities has been analysed and has provided helpful insight hereto and the findings are well in agreement with considerations from other studies.

6.3.5.2 n-Heptane hypothesis

n-Heptane has been widely used in the literature as diesel surrogate due to the similar autoignition behaviour and the availability of various reduced mechanisms suitable for CFD applications. Despite the limited size of those mechanisms they have often seen successful applications, namely n-heptane chemistry was used in most of former works at the current engine, e.g. [20, 22, 46, 107, 108, 194, 200], while in [31] an n-heptane/toluene blend was employed. In this study the model was found to perform well in terms of pressure traces and AHRR for all 5 reference cases. The simulation tends to underestimate the premixed burn; this is most likely attributed to an insufficient mixture preparation prior to ignition. In this sense, the simulation showed a comparatively higher stratification in terms of both equivalence ratio (cf. Figure 6-3) as well as OH species (cf. Figure 6-5) compared to experiments.

For what concerns soot behaviour, the most prominent soot inception path is via PAHs [66]. Diesel fuel used in the experiment contains 27 percent fuel bounded aromatics whereas the simulation considers pure n-heptane chemistry without aromatics and the modeled soot inception is assumed to be driven by acetylene only. The simulation results provided evidence that the prediction of soot formation for both HTC cases was excellent while for all LTC cases it was underestimated. The same trend was observed in [20], where a reduced n-heptane chemistry mechanism has been extended to include PAH species up to four fused aromatic rings (pyrene) and the latter is used as the inception species. The authors argued that soot inception at HTC may be dominated by linear unsaturated hydrocarbons such as acetylene due to the fast fuel decomposition. This hypothesis is supported by the importance of fuel decomposition at high temperature for flames in [66]. On the other hand under
LTC conditions PAHs may enhance soot production considerably [20], since the fuel bounded aromatics are expected to contribute to soot formation. Engine experiments at HTC conditions quantifying the survival rate of fuel-bound PAHs have been presented in [201], reporting that only a few percent of the PAHs measured in the exhaust originate from the fuel, while the rest stem from pyro-synthesis. As a consequence, in the first phase of soot formation, the simulation creates less soot compared to measurements at the HTC conditions, as one may expect conceptually, because fuel decomposition and soot precursor formation occurs in a sequential way for a pure paraffinic fuel as \( n \)-heptane.

Unfortunately, no measurements are available in [201] at LTC conditions which could provide quantitative estimates relating to the fate of the fuel-bound PAHs at these considerably lower temperatures. The influence of the fuel bounded aromatics has however been examined in [202] at LTC conditions by means of PAH-LIF and soot-LII techniques (on the same optical engine as investigated in this study). The authors reported marginal differences between diesel fuel and pure \( n \)-heptane for both PAH and soot spatial distribution evolutions, although the characteristic soot inception time was found to be slightly larger with \( n \)-heptane. Large differences were however evident in the camera gain settings indicating substantially different amounts of PAHs for the two fuels (although uncertainties due to attenuation effects vs. changes in species concentrations were reported, obstructing quantitative statements). In [176] the same trend was also experimentally observed in a constant-volume chamber, i.e. a longer inception time for pure \( n \)-heptane as opposed to diesel fuel. For the LTC engine cases examined here, peak soot mass is underpredicted for the PAH-containing diesel fuel due to considerations given above while the current model applied to \( n \)-heptane sprays in [26] overestimated soot at similar low temperature conditions.

In summary, these findings – despite the remaining uncertainties – suggest, that at HTC conditions the use of acetylene as a precursor may suffice, as PAHs are created ‘bottom-up’. Conceptually, the use of pure \( n \)-heptane oxidation chemistry at these conditions is however questionable since pyrolysis and oxidation chemistry of fuel-bound PAHs could in principle play an important role. At LTC, where the conversion of fuel-originating PAH may be substantially reduced (and hence the use of \( n \)-heptane oxidation kinetics may be better justified), the inclusion of alternative inception paths, i.e. via PAH in addition to the acetylene precursor, appears appropriate. Evidently, further research both experimentally as well as numerically for these engine relevant diesel engine conditions is clearly advocated.
6.3.5.3 Soot oxidation

Soot oxidation was found to be in very good agreement for the diluted cases and considerably overestimated for both HTC cases. Here, the computed characteristic oxidation rate was found to be approximately twice as high as the measured one for these two operating conditions. From Figure 6-10 it is evident, that the soot mass evolution for the HTC-short case was only marginally influenced by changes in the reaction rate of soot oxidation by $O_2$ and OH. Furthermore, the discrepancies in the rate of oxidation coincide with the diffusion controlled phase of combustion, which is governed by the oxidizer entrainment. These findings suggest that the overestimation of the soot oxidation rate may be caused by an inaccurate description of the mixture rather than inappropriate soot oxidation kinetics. On the contrary, the LTC cases are characterized by almost purely premixed combustion and soot oxidation agreed very well. For positive ignition dwell cases (LTC) the relative importance of an accurate spray prediction seems to be less crucial because of the time available after EOI for mixing and discrepancies are expected to be reduced.

Overall, results shown in this study represents a very good achievement for soot formation in a real diesel engine for considerably different operating modes. Various considerations concerning soot comparison approaches and detailed soot processes were elucidated. The findings reported contribute towards the improved understanding of in-cylinder processes and constitute a step towards improved predictive modeling of soot in diesel engines.
7 Conclusions

7.1 Conclusions

In this work, numerical simulations of diesel spray combustion with emphasis on soot formation have been performed using multi-dimensional first order conditional moment closure (CMC) coupled with a reduced \( n \)-heptane chemical mechanism, representing the first application of a semi-empirical soot model within the framework of CMC for spray combustion.

Validation and application of the model have been conducted at three different experimental test facilities as follows: (1) model validation at the Sandia constant-volume chamber for quasi-steady fully developed \( n \)-heptane sprays, (2) model application at the Wärtsilä marine spray combustion chamber for diesel sprays representative for large two-stroke marine diesel engines and (3) model application at the Sandia heavy-duty diesel engine with intermittent diesel sprays.

In the following, achievements and conclusions are summarized for each test facility separately.

**Sandia constant-volume chamber:**

A detailed analysis of \( n \)-heptane autoigniting sprays under diesel engine conditions has been conducted, including comparison with experimental data of ignition delay (ID) and flame lift-off length (LOL) for different ambient oxygen volume fractions (8-21\%) and ambient densities (14.8 and 30 kg/m\(^3\)). ID was in general over-estimated, particularly for cases with low oxidizer reactivities. Flame LOL was found to agree well with the experiment with discrepancies within 24\%. During the quasi-steady period (3-6 ms), high soot volume fraction location and semi-quantitative distribution have been well described.

For a reference case A1, a systematic sensitivity analysis was carried out with respect to the soot model constants, which have been varied by a factor of two. A strong sensitivity with respect to the surface growth rate was observed, while the other soot processes showed a weaker influence with changes in soot volume
fraction being at the most 8%, in agreement with findings reported in the literature for flames.

Soot related quantities have been analysed in conserved scalar space for two different ambient oxygen concentrations, illustrating that soot related processes occur almost at the same equivalence ratio. Soot mass fraction is only present in the fuel rich region with a peak at an equivalence ratio of around 2. At this location, equilibrium between surface growth as the dominant source and axial convection as the most important sink was observed. Soot mass fraction diffusion in mixture fraction space towards stoichiometry was mostly compensated by soot oxidation by O$_2$ and OH. The former was important only at high O$_2$ concentrations, where significantly higher OH contents are present in the flame.

The comparison of distributions of formaldehyde, acetylene and soot as well as the location of the diffusion flame agree well with the currently established conceptual models of diesel flame structures.

Furthermore, the influence of the combustion model has been assessed by comparing the CMC model with the direct integration (DI) approach (i.e. neglecting turbulent fluctuations) shedding light upon which conditions turbulence-chemistry interaction (TCI) plays a major role. Neglecting turbulent fluctuations in the evaluation of the chemical source terms played a marginal role for the prediction of the ignition, a success attributed to the fact that for this particular experiment, the scalar dissipation rate is much lower than the critical value above which ignition is inhibited. Trends of LOL were captured with both methods; however DI showed larger discrepancies with experiment than CMC, in particular at the lower oxygen concentrations. This was attributed to the substantial mixture fraction variance. The main difference in the quasi-steady spray flame structure is the oxygen concentration in the fuel-rich region, where DI predicts complete consumption of oxygen, whereas for CMC a certain oxidizer stratification is still present. This results in a much thinner reaction zone prediction by DI. Despite considerable differences in the flame structure, the quasi-steady spatial soot distribution was found to be comparable between the approaches. Given the same flame, soot formation rate with DI was almost the same as with CMC, whereas soot oxidation rate by OH and O$_2$ was found to be one and two orders of magnitude higher, respectively. This was mainly attributed to a small mixture fraction co-existence range of oxidizer and soot, which is not accounted for in the DI formulation.
For some of the simulated experiments, the considerable overprediction of soot oxidation rate by neglecting TCI plays a marginal role for the prediction of quasi-steady soot distribution. The main reason for that is twofold: 1) TCI was found to affect soot formation only little and 2) the complete absence of oxygen and OH within the fuel rich region for the DI case which results in zero soot oxidation in the rich zone. The comparison for an intermittent spray typical of diesel engine operation, however, revealed that following the end of injection as oxygen is re-entrained into the spray, soot oxidation is massively overestimated by DI since segregation of soot and oxidizer in mixture fraction space cannot be accounted for appropriately. In contrast, the CMC model agrees well with the experimental data of the short injection case considered.

Overall, it was shown that for both quasi-steady sprays as well as short injection cases, using CMC shows clear benefits over the ‘direct integration’ approach neglecting turbulence-chemistry interactions. This is especially true for soot oxidation processes after the end of injection of intermittent fuel sprays typical for diesel engines.

**Wärtsilä marine spray combustion chamber:**

Numerical simulations of autoigniting diesel sprays with an injector orifice diameter of 0.875 mm from a two-stroke marine diesel engine reference experiment have been performed. Simulation results are compared with experimental data by means of macroscopic spray development under non-reacting conditions as well as ID time, ignition location and quasi-steady flame LOL for different ambient temperatures ranging between 730 and 910 K.

Good agreement is reported for the ignition delays spanning one order of magnitude at the given conditions, for which the values were over/underpredicted to within 9 and 23 %. Flame LOL were over/underpredicted at most by 13 percent; the trend of the ignition spot shifting to downstream locations for lower temperatures was correctly reproduced with an accuracy of the predictions within 40 percent. The computed ignition sequence on the lee side of the swirl was consistent with experimental data.

The spray flame was analyzed and discussed in the context of the large nozzle orifices employed and analogies as well as discrepancies to sprays with considerably smaller dimensions are highlighted. For large injectors, the main difference lies in the massive overlap of the liquid region with the flame region. Simulation results indicated an evaporative-reactive balance around the liquid region, which poses additional challenges to the already complex task of spray combustion modeling.
**Sandia heavy-duty diesel engine:**

The model has been subsequently applied for the Sandia optically accessible heavy-duty diesel engines for five different operating conditions including two high-temperature cases and two low-temperature cases with two different injection timings and one low-temperature case with a split injection strategy. The spray model was validated against a non-reactive case by means of equivalence ratio distribution. It was found that ignition delays were well predicted with discrepancies of one degree crank angle the highest. Apparent heat release traces were well reproduced across different conditions, although the simulation tends to underestimate the portion of premixed burn. Based on comparisons of equivalence ratio and OH species distributions, this can most likely be attributed to an overpredicted mixture stratification.

A quantitative comparison of temporal in-cylinder soot mass has been presented. The soot model was capable to reproduce semi-quantitative trends of soot mass for all cases considered without any changes in soot model constants. For the high-temperature cases the peak soot mass and time were very well captured, with discrepancies of 4 and 23 percent for the in-cylinder mass and offsets of 1 degree crank angle for the peak location/timing. The characteristic oxidation rate, however, was roughly twice as high as the measured values. Discrepancies were attributed to inaccuracies in mixture formation during diffusion combustion. At low-temperature conditions the shape as well as the timing of soot evolution during formation and oxidation were in excellent agreement with experimental data for all three cases. When compared to the experimentally reported values, the soot peak mass was underpredicted for all cases by a factor of 1.25, 2 and 3.8 for the double, early and late injection cases, respectively. This can possibly be attributed to the importance of soot inception via PAHs of fuel bounded aromatics at low temperatures.

Furthermore, a line-of-sight transformation of the computed results for soot natural luminosity has been applied to allow for a more consistent comparison with experimental data. Apart from the well-known signal bias towards hot soot properties, soot absorption along line-of-sight was found to play an important role on natural luminosity distribution, in particular for the HTC cases. With this methodology additional effects related to radiation adsorption were observed. In general, evolutions of the soot clouds were qualitatively in very good agreement with experiment.
Overall, it was shown that the two-equation soot model implemented in the CMC framework is a highly promising candidate for the prediction of soot emissions in the full complexity of diesel engines.

7.2 Suggestions for future work

In this section, several suggestions for future research that arise from the current work are proposed. This dissertation was mainly focused on the validation and application of the CMC method for studying auto-igniting sprays in diesel engine relevant conditions with emphasis on soot formation. In this sense, suggested work is mostly related to diesel engines applications, and it consists of six different modeling aspects: (1) influence of droplet evaporation on combustion, (2) inclusion of PAH chemistry, (3) improved description of soot dynamics (particle size distribution and agglomeration), (4) influence of soot differential diffusion, (5) inclusion of an optically thick radiation method, and (6) inclusion of multiple injection events.

The influence of evaporating droplets on combustion need further research, in particular for the marine spray combustion chamber where the liquid fuel region abundantly overlaps the flame region and an increased liquid-flame interaction is expected. The influence of evaporation on mixture fraction variance and scalar dissipation rate needs further consideration. This should be done by means of direct numerical simulations (DNS). Recently, Borghesi et al. [172] have reported DNS results of autoignition of evaporating \( n \)-heptane droplets at elevated pressures, providing additional insights on the influence of evaporation on combustion.

A further development of the model would be the usage of a more complex chemical mechanism with the inclusion of a more elaborated soot inception path via PAH. The motivation for this extension is twofold. First, petroleum-based diesel fuel contains generally 20-30 % of fuel-bounded aromatics components that are currently ignored with a pure \( n \)-heptane chemical model. Second, the inclusion of a mechanism for the PAH growth with different governing paths for different temperature regimes may improve soot inception particularly at lower temperatures (with EGR) where a pure acetylene based inception was found to underpredict soot. The size of the chemical mechanism has to be carefully chosen as a compromise between accuracy and computational expense. In this
Conclusions

regard, an exploration of a chemistry accelerator e.g. ISAT may become necessary.

In the diesel engine community the particle number and particle size distribution (PSD) is becoming increasingly of importance. A soot model extension to account for higher moments of the PSD using a moment-based model may be a valuable choice with a modest increase in computational time. In general 4-6 moments are enough for an adequate description of the PSD. Another improvement of the soot model is represented by the inclusion of particle agglomeration which can have an important impact on the soot surface reactions (formation and oxidation). Also in the context of the Engine Combustion Network the particle size has receiving more attention by experimentalists and techniques as laser induced incandescence (LII) or transmission electron microscope (TEM) have seen recent application.

The effect of soot differential diffusion on soot distribution in several atmospheric non-premixed flames was shown to be prominent. Under diesel engines conditions the discussion is controversial. A careful examination of the existing models and a detailed analysis of the performance of the various closures proposed could be a viable procedure.

The presence of soot is generally linked to radiation. For high pressure flames high levels of soot volume fraction (several ppm) are present and an optically thin assumption for radiation does not apply. In this study, soot radiation for the diesel engine simulation has been deliberately neglected in the absence of an optically thick formulation currently available. A possible approach with feasible computational cost would be the discrete ordinate method (DOM), however the influence of differential diffusion should be addressed first.

Considering the inclusion of a model formulation capable to account for fuel multiple injection may be of interest in view of the flexibility of modern injection systems, where operation with multiple injection is routinely implemented. This would considerably extend the applicability of the model for real diesel engines.
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