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Part-Load Particulate Matter from a GDI Engine and the Connection with Combustion Characteristics

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ABSTRACT

The Gasoline Direct Injection engines are an important source of ultra-fine particulate matter. Significant research effort is still required as improved understanding of soot formation is critical in considering further development or adoption of new technologies. Experimental measurements of engine-out soot emissions have been taken from a modern Euro IV GDI engine at part-load operating conditions. The engine speed and torque were varied in the range 1600 to 3700 rev/min, and 30 to 120 Nm, respectively. The engine was invariably operated in stoichiometric and homogeneous combustion mode, with fuel injection early in the intake stroke. The results indicate that for engine load in excess of 3 bar Brake Mean Effective Pressure, due to incomplete gas-phase mixture preparation, a consistent linear correlation establishes between combustion duration and soot particle number. On average, a six-fold increase in number concentration between 1.0 and 6.0×10^6 particle per cc, arises from shortening the rapid duration of 4 crank angle degrees. For engine speed in excess of 3000 rev/min and load in excess of 7 bar BMEP, this correlation appears to be superseded by the effects of spray-to-piston impingement and consequent pool-fire. Three main areas of concern have been identified within the part-load running envelope: 1. the higher load-lower speed range and 2. the mid load-mid speed range, where high nucleation rates induce copious increases of engine-out soot mass; 3. the upper part-load range where, most likely as a result of spray impingement, high levels of soot concentration (up to 10 million particles per cc) are emitted with very small size (23-40 nm).

1. Introduction

Compared to more conventional Port-Fuel Injection (PFI) engines, Gasoline Direct Injection (GDI) engines show a significant 5 to 15% improvement in fuel economy [1], especially because of higher volumetric efficiency and higher knock resistance, which allow the use of generally higher compression ratios with sizeable benefits in thermal efficiency and specific power output. In spite of this, GDI engines are an important source of environmental pollution because of their fine and ultrafine Particulate Matter (PM) emissions. Sizeable research effort has been devoted in the last two decades to investigate the causes of soot emission from this comparatively young technology. Recent medical research work, showing that aerosol particles in the ultrafine size range (diameters of less than 100 nm) cause adverse health effects [2-6], continue to give relevance and impetus to GDI research. Pulmonary inflammation, asthma and cardiovascular conditions are some of problems associated with the deposition of soot in the respiratory tracts. Health risks generally increase with decreasing particle size and increasing concentration. A recent, large, European study associate an 18% increased risk of lung cancer to a 5×10^{-6} $\mu\text{g}/\text{cc}$ increase of $\text{PM}_{2.5}$ in atmospheric air [7]. Compared to diesel engines without particulate filter (non-DPF), GDI engines produce smaller particles in greater number [8] and hence represent an increased level of threat. Gaining an improved understanding of the soot formation process, and its connection with combustion, is of paramount importance as this would enable further developments in engine design and combustion control strategies to reduce PM emissions [9, 10]. Limiting the exhaust gas soot number concentration in GDI engines is one of the main objectives of new proposed emission regulations across the globe [9].

In GDI engines, the process of fuel vaporisation and gas-phase mixing remains essentially incomplete [8], even when early fuel injections are used to enable an homogeneous combustion

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58 mode [11, 12]. This leads to the establishment of sub-stoichiometric mixture-pockets, which are an
59 important source of soot formation. A second source is from fuel-spray impingement into cylinder
60 liner and piston crown, and resulting pool-fire [9, 13, 14]. As suggested by Maricq et al. [15],
61 another source of PM formation, especially when GDI engines operate in stratified mode, is
62 attributable to incompletely volatilized fuel droplets, as the unburned gas is swept across by the
63 incoming flame front. The particle number concentration emitted by GDI engines are generally
64 higher than conventional PFI engines and Diesel engines equipped with Particulate Filter (DPF).
65 This trend was shown in a study by Braisher et al. [16], which compares four types of modern
66 engines. The reported total soot particle numbers over the length of the New European Drive Cycle
67 (NEDC) are as follows: 6.7×10^{14} for Non-DPF Diesel engine; 6.8×10^{13} for GDI engine, 6.0×10^{12} for
68 PFI engine and 1.3×10^{12} for DPF Diesel engine. A recent paper by Choi et al. [10] compares wall-
69 guided to more recent GDI combustion systems, such as spray-guided and air-guided. The paper
70 reviews the technologies and the potential mechanisms of soot formation in each of them, on the
71 basis of experimental measurements of exhaust soot. Remarkably, "*none of these [technologies]*
72 *were able to satisfy the proposed particle number regulations for Euro 6 standards*" [10]. Improved
73 combustion chamber design and injection strategies appear highly necessary at present to reduce
74 particulate matter emissions.
75

76 1.1 Soot Formation Mechanism and Exhaust Measurements

77 The phenomenon of soot formation is typically described in terms of three steps: nucleation,
78 growth and oxidation [17]. The process occurs under fuel-rich conditions, in both premixed and
79 non-premixed flames, where the local equivalent ratio is more than one. The nucleation process
80 takes place under high temperature conditions, between 1000 and 2800 K, with unburned
81 hydrocarbons, in particular acetylene and polycyclic aromatics hydrocarbons (PAH), being
82 pyrolyzed and oxidized. The condensation reactions of these gas-phase species lead to a large
83 number of *primary soot particles* with diameter lower than 2 nm and insignificant soot loading.
84 Surface growth, coagulation and aggregation represent the particles growth. During surface
85 growth, concentric shells on nuclei and spherules are formed by deposition of hydrocarbon
86 intermediate gas-phase species on particles surface. By means of coagulation the particles collide
87 and merge reducing their total number. After formation, the collision between particles leads to
88 clusters or chain-like aggregates (*secondary particles*), with a consequent size increase
89 (equivalent diameter of 100 to 900 nm). In the overall soot formations process, the precursors, the
90 nuclei and particles can be oxidized if in contact with oxidizing species as O_2 , O, OH, CO_2 , H_2O at
91 the right conditions. The exhaust soot emissions depend on the balance between formation and
92 burnout processes [18]. The vast majority of formed soot is completely oxidised within the
93 combustion chamber [9, 13].
94

95 One of the most common instruments to measure size-resolved soot number concentration
96 distribution in exhaust gas samples is the Differential Mobility Spectrometer (DMS). The DMS-500
97 system by Cambustion, used in this work, is a real-time instrument which classifies particles on the
98 basis of their electrical mobility equivalent diameter [19-22]. Electrical mobility is measured within
99 the classifier column in which a central high-voltage rod deflects the charged particles towards 22
100 grounded electrometer rings via electrical repulsion. Size discrimination is obtained by separating
101 the particles on the basis of their aerodynamic drag-charge ratio, ultimately dependent on the
102 particle diameter. The range of detection is between 5 and 1000 nm, and the response time 200
103 ms. Typically, the particles are said to be in nucleation mode when their size is smaller than 50 nm;
104 and in accumulation mode when size ranges between 50 and 1000 nm [21]. Another system based
105 on electrical mobility theory is the Differential Mobility Analyser (DMA), usually coupled with a
106 Condensation Particle Counter (CPC) that measures particles number concentration by optical
107 detection [23]. The combination of DMA and CPC is configured as a Scanning Mobility Particle
108 Sizer (SMPS) [24]. The Electrostatic Low Pressure Impactor (ELPI) is based on inertia principles
109 [25, 26]. Price et al. [20] found a qualitative agreement between all instruments by measuring PM
110 emissions from a passenger vehicle equipped with a GDI engine. Importantly, the agreement was
111 stronger for PM mass measurements than for PM number concentration.
112

113 Maricq et al. [15] thoroughly examine the connection between soot formation and engine variables,
114 which are changed in isolation enabling the identification of potential pathways to reducing

115 particulate emissions. At fixed speed and load, stratified combustion mode produces in general up
116 to two orders of magnitude more particles than homogeneous mode. In both modes, PM number
117 density increases as the ignition timing is advanced, while the peak of the size-resolved distribution
118 shifts towards bigger particle diameters. In homogeneous mode, an ignition timing of 21 CA deg
119 BTDC produced peak number density of 6.0×10^5 N/cc, located at 70 nm. Advancing the ignition
120 timing to 40 CA deg BTDC raises the peak number density to 2.0×10^6 N/cc, and shifts the peak
121 location to 85 nm. Increased nucleation rates are associated with greater peak combustion
122 temperature, as the ignition timing is advanced earlier in the compression stroke. On one hand,
123 higher particle concentration in the burned gases leads to higher coagulation rate; on the other
124 hand, greater ignition advance causes lower exhaust gas temperature, hence lower post-flame
125 oxidation. Both mechanisms would enable the emission of generally bigger particles when the
126 ignition timing is advanced [27].

127
128 Graskow et al. [28] investigate the exhaust soot characterisation using a vehicle equipped with GDI
129 engine. In theoretically-homogeneous mode, at a vehicle speed of 90 km/h, the total soot number
130 density was 9.3×10^7 N/cc and the Geometrical Mean Diameter (GMD) was 88 nm. In stratified
131 mode, an increase in vehicle speed was associated to increased soot nucleation rate. The total
132 number density increased in the range 1.14 to 2.8×10^8 N/cc, and the GMD increased between 35
133 to 81 nm.

134
135 Price et al. [29] investigate soot exhaust emissions from a spray-guided GDI engine using toluene
136 as fuel. In line with previous results, they show that increasing ignition advance between 30 and 45
137 CA degrees BTDC produces an order of magnitude increase in peak soot number density,
138 between 3×10^5 and 3.1×10^6 N/cc, with a slight downwards trend in particle size at peak values,
139 which changed between 25 and 18 nm. Soot particles size distribution shows a peak at about 25
140 nm independently of Start Of Injection (SOI), whereas the peak number density increases from
141 2.5×10^5 to 2.4×10^6 N/cc when SOI is shifted from 300 to 200 CA deg BTDC. Spray-guided
142 combustion forms soot mostly in the nucleation mode range, with typical GMD of 30 nm. Over the
143 same operating range, a second experimental GDI engine using wall-guide combustion system
144 emitted on average bigger soot particles, in much greater quantity (up to two orders of magnitude).

145
146 Farron et al. [30] report a comprehensive investigation on the influence of engine operating
147 variables on PM exhaust emissions, using a GDI engine and SMPS measurements. At low engine
148 load, they found the lowest particle number density (1.6×10^5 N/cc distribution peak) for End of
149 Injection (EOI) of 280° BTDC and an increase for both earlier EOI of 330° BTDC (1.2×10^7 N/cc)
150 and later EOI of 100° BTDC (1.15×10^7 N/cc). The corresponding particle sizes at peak number
151 density were 30 nm, 150 nm and 80 nm, respectively. Increasing engine load led to higher PM
152 mass due to increased fuelling quantity. Doubling the mass of fuel injected per cylinder from 11 to
153 21 mg, produced almost an order of magnitude increase in peak number density, between 1.6×10^5
154 and 1.2×10^6 N/cc. As the engine load was increased, the particle size of peak N/cc increased
155 between 30 and 60 nm. At low engine load, increased ignition retard induced a reduction of PM,
156 both in size and number density. A similar effect was obtained by increasing the fuel pressure, on
157 account of smaller fuel droplets size.

158
159 Whitaker et al. [31] investigate various gasoline direct injection strategies to reduce soot particle
160 number. In cold start conditions, a 4-stage split, high-pressure, reduced-duration injection reduces
161 the peak soot number density from 1.8×10^8 to 3.0×10^7 N/cc. Similar strategies are used to reduce
162 soot emissions at steady-state, fully-warm engine conditions.

163 Barone et al. [24] use SMPS to characterise exhaust soot from a 2.0 litre GDI engine operated in
164 theoretically-homogeneous mode, and Transmission Electron Microscopy (TEM) to investigate the
165 morphology of particles and aggregates. An early fuel injection strategy (Start Of Injection, SOI of
166 320 CA deg BTDC) produced total particle number density N/cc of 1.7×10^7 , with a Geometric Mean
167 Diameter (GMD) of 66 nm. Morphology studies showed single solid spheres with a sub-25 nm
168 diameter, nano-particle aggregates ranging between 8 and 52 nm and liquid droplets with size
169 between 40 and 400 nm. Most aggregates had fractal-like morphology similar to diesel soot. A
170 more retarded injection strategy (SOI of 280 CA deg BTDC) lowered the total number density of

171 about one order of magnitude to $1,2 \times 10^6$ N/cc, with a GMD of 48 nm. The particle morphology was
172 similar to that of early injection, but the presence of liquid droplets was negligible. Similar results on
173 PM characterisation, and its variation with injection timing, have been reported by Sabathil et al
174 [32]. The reduction in PM number density experienced at later injection timings may be explained
175 with the reduced incidence of the fuel-spray impingement, hence reduced wall-wetting.
176

177 Pei et al. [33], in their recent GDI engine investigation, emphasise the reliance of PM emissions on
178 engine operating parameters, at part-load conditions. Owing to the inherent incomplete gas-phase
179 mixing, the measured soot number density increases with decreasing Air Fuel Ratio (AFR),
180 retarded SOI and advanced ignition timing. When AFR varies between 18 and 11, the total range
181 of variation of soot number concentration is between 5.1×10^7 and 5.6×10^8 N/cc. Correspondingly,
182 the soot GMD increases from 9 to 30 nm. For all parametric studies, Pei et al. [33] attempt
183 identifying a relationship between PM emissions and combustion characteristics such as
184 combustion temperature and heat release rate.

185 This paper analyses experimental measurements of engine-out soot emissions and combustion
186 characteristics from a wall-guided GDI engine operated in stoichiometric and homogeneous mode,
187 at part-load running conditions. A survey of relevant literature shows compelling evidence that soot
188 formation in GDI engines is triggered by incomplete air-fuel mixture preparation [9, 10, 13, 15, 24,
189 29, 30, 32, 34]. As variable amounts of soot are emitted as a result of different engine operating
190 variables, this evidence suggests the existence of a relationship between engine burning
191 characteristics and the process of soot formation. The primary aim of the present work has been to
192 explore the correlation between soot number density and combustion characteristics, with specific
193 emphasis given to flame propagation duration and unburned gas temperature.
194

195

196 **2. METHODOLOGY**

197

198 **2.1 Engine Testing and Data Acquisition**

199 Engine testing was carried out using a 1.6 litre, turbo-charged and intercooled, Euro IV, Direct
200 Injection Spark Ignition (DISI), gasoline engine with no modifications. The engine technical
201 specifications are given in Table 1. The engine was installed on a laboratory test bed and
202 controlled by a Schenck W150 eddy-current dynamometer, via a CP Engineering CADET engine
203 control and data acquisition system. Fuel consumption was measured via a CP Engineering FMS-
204 400 gravimetric system. Engine testing was carried out under steady-state, fully-warm operating
205 conditions crossing a large portion of the part-load running envelope. Engine speed was varied in
206 the range 1600 to 3700 rev/min, in steps of 700 rev/min; torque was varied in the range 30 to 120
207 Nm, in steps of 30 Nm. A total of 16 operating conditions were investigated and the tests were
208 repeated three times to ensure consistency and repeatability. The spark timing was controlled via
209 the manufacturer ECU, optimised for best thermal efficiency and minimal fuel consumption. The
210 fuel injection strategy was also controlled via the ECU, ensuring a theoretically homogeneous
211 air/fuel mixture with an equivalent ratio of 1.0 at the conditions tested. In the wall-guided DISI
212 combustion system used here, the fuel is injected into the combustion chamber by a side-mounted
213 injector. The mixture is then 'guided' towards the spark-plug region by the charge motion (reverse
214 tumble) and a spoon-shaped piston crown. The delivery of fuel is via a common-rail direct injection
215 system; injection pressure is regulated via the ECU as a function of load and speed, with a
216 maximum value of 120 bar. The fuel used for testing was pump-grade, 95 RON unleaded gasoline
217 with a stoichiometric AFR of 14.4. A Kistler piezo-electric, spark-plug mounted, pressure
218 transducer was installed in one cylinder to acquire in-cylinder pressure variation. Batches of 100
219 consecutive pressure cycles were acquired via an AVL Indicom acquisition system to evaluate the
220 ensemble-averaged pressure trace, and the combustion evolution in terms of Mass Fraction
221 Burned (MFB) profile. A schematic of the engine rig experimental setup is presented in figure 1.
222

223 **Figure 1**

224

225 **Table 1**

226

2.2 Engine-Out Soot Measurements

Parallel measurements of exhaust gas soot content and size distribution were performed using the Cambustion DMS-500 system. The sampling point for engine-out gas stream was fitted into the exhaust manifold, close to the exhaust port of the cylinder where pressure measurements were taken. The gas sample was passed through a capillary tube into a heated switch valve at a constant temperature of 190° C. A heated sample line maintained at 58° C connected the switch unit to the PM measurement unit. A scroll vacuum pump was used to draw the sample at a constant mass flow rate. PM measurements were consistently carried out using a primary dilution ratio of 5 and a secondary dilution ratio of 25, respectively. At each steady-state condition, the exhaust stream was sampled for 10 minutes. The data was time-averaged via the manufacturer software to yield the size-resolved soot number density distribution. The particle mass was calculated by taking a limiting assumption that the particles are spherical and have a density of 1800 kg/m³. As for the in-cylinder pressure measurements and cycle-resolved engine data, also the engine-out soot measurements were repeated three times. Following Farron [30], the repetitions were taken not only during the same testing day, but also during different days. Typical repetitions of time-averaged, size-resolved number density distributions are presented in figure 2. Although some low variability is expected as the running conditions can never be exactly reproduced [30], the low combustion variability at the conditions investigated enable PM measurements with acceptable repeatability. Similarly to Chen et al. [34], the data referring to particle size below 10 nm were excluded from the analysis due to increased variance in that range.

Figure 2

2.3 Mass Fraction Burned Calculation

The following correlation, derived from the original methodology by Rassweiler and Withrow [35, 36], has been used to evaluate the Mass Fraction Burned, x_{MFB} , as a function of crank angle:

$$[x_{MFB}]_{\vartheta} = \frac{[m_b]_{\vartheta}}{m_{fc}} \approx \frac{\sum_{\vartheta_{ST}}^{\vartheta} \Delta P_c}{\sum_{\vartheta_{ST}}^{EOC} \Delta P_c}$$

Here, $[m_b]_{\vartheta}$ is the mass actually burned at any location ϑ after combustion initiates (ignition timing) and m_{fc} is the mass of fresh charge, including air and fuel, trapped inside the engine cylinder at IVC. ΔP_c is the pressure rise due to combustion in one CA step, calculated as difference between the total experimental pressure rise, and the one due only to volume variation. EOC indicates the CA location of End Of Combustion, corresponding to 100% MFB. In the present work, two different values of the polytropic index are used to model the pressure variation due to volume change; one for the compression and one for the power stroke. The polytropic compression index is calculated as the negative of the slope of the experimental $[\log V, \log P]$ diagram over 30 consecutive points before spark timing, and maintained unvaried up to TDC. While the sensitivity of the MFB profile to the compression index is relatively low, the selection of the expansion index is more important. The expansion index is estimated with an iterative procedure where, starting from a reference value (e.g. 1.3), the index is progressively adjusted together with the EOC. The iterations continue until the MFB curve acquires a *reasonable S-shape*, which meets the requirement of zero combustion-pressure condition, i.e. the curve remains steadily at 100% after EOC. In this work the combustion process is supposed to terminate when ΔP_c becomes a negligible fraction (within 0.2%) of the total pressure increment, for 4 CA-steps consecutively. The uncertainties and accuracy associated to this methodology are more comprehensively discussed in [35, 36].

Two common combustion duration indicators are calculated from the MFB curve, Flame Development Angle (FDA) and Rapid Burning Angle (RBA). FDA is calculated as the interval between spark timing and 10% MFB. RBA is calculated as the interval between 10% and 90% MFB. The selection of 90% MFB as the end of rapid combustion provides a safe reference point,

279 as this location is not overly affected by parallel heat release processes taking place in the
280 chamber during the so-called combustion termination. In the present work, further combustion
281 indicators are used to aid the examination of soot formation. These are presented in the results
282 and discussion section.

283
284

285 3. RESULTS AND DISCUSSION

286

287 3.1 Analysis of Engine-Out Soot Measurements – Number Density

288 The experimental results for engine-out total particle number density (N/cc), at part-load running
289 conditions, are presented in this section. N/cc values, averaged upon three consistent steady-state
290 test repetitions, are shown in figure 3 as a function of engine speed and load (BMEP). A contour-
291 plot type map has been used for the analysis. The average experimental records of N/cc are
292 interpolated using a triangle-based cubic model over a uniform grid of speed and load points. The
293 same approach has been used to presents other results in this paper. Data regarding spark ignition
294 advance (CA degrees BTDC) and mixing time, calculated as time in ms between EOI and TDC of
295 combustion, are presented in figures 4 and 5. The process of “mixture preparation” includes fuel
296 droplets vaporisation and mixing of air and combustible vapours to create a homogeneous and
297 uniform mixture throughout the chamber. Mixture preparation may continue after the inception of
298 combustion. Ignition timing and mixing timing constrain mixture preparation, hence are of
299 significance in the process of soot formation. Three high-soot regions can be identified in the N/cc
300 map of figure 3. The first is located in the top-left corner of the map, at low speed and high load.
301 The second region is shaped as an island of high number density in the mid-speed, mid-load
302 domain. The last one, showing a steep increase in soot particle number density, corresponds to
303 top-right corner, the upper part-load domain.

304

305 **Figure 3**

306

307 **Figure 4**

308

309 **Figure 5**

310

311 Mid to low load (4.5 bar BMEP and below).

312 At the lowest engine speed investigated (1600 rev/min) and a fixed load of 4.5bar, the
313 comparatively long mixing time (about 28 ms) allows the moderate amount of fuel injected (11.5
314 mg per cycle per cylinder) to vaporise and mix with the intake air, minimising the rate of soot
315 nucleation. As speed increases, the available mixing time shortens, causing poorer mixture
316 preparation. Although greater engine speed tends to mitigate these effects by stretching the
317 combustion process over wider CA intervals, the increasingly advanced spark timing also leaves
318 less time for mixing, as well as inducing higher temperature during combustion [15, 29, 30, 37].
319 When engine speed grows between 1600 and 3000 rev/min at 4.5 bar BMEP, the net effect of
320 these factors is a six-fold increase in soot N/cc (to 6×10^6 particles/cc). As engine speed exceeds
321 3000 rev/min, N/cc decreases approaching levels as low as 2×10^6 at 3700 rev/min. In this region,
322 the mixing time is approximately constant (see figure 5) and the greater in-cylinder turbulence
323 would enhance vaporisation and mixing of the moderate amount of fuel injected [37], reducing the
324 rate of soot nucleation. At lower, fixed engine load (e.g. 2.5 bar BMEP) the small amount of fuel
325 injected (7 mg per cycle per cylinder) inhibits the formation of particulate matter, especially at the
326 lower engine speeds when longer air-to-fuel mixing intervals are available. The distribution of soot
327 N/cc is remarkably similar to that at 4.5 bar BMEP, only scaled down as expected to lower
328 concentrations.

329

330 Higher load (7 bar BMEP and above)

331 At a fixed engine load of 7 bar BMEP, the speed-dependent soot number density shows a
332 characteristic U-shape distribution, common to all higher load cases. As the engine speed is
333 reduced below 2600 rev/min, the total number density rises towards a maximum level of 6.5×10^6
334 N/cc at 1600 rev/min. In spite of the longer time available for mixing, the large mass of fuel injected
335 (between 17 and 22 mg) may cause a less effective process of mixture preparation. The greatly

336 retarded spark timing in this region would also contribute to the recorded sharp number density
337 increase. As the engine speed is increased above 2600 rev/min, the major influence on N/cc is
338 likely to be fuel spray-to-piston/liner impingement [10], whose effects start appearing at 7 bar
339 BMEP, becoming much more significant as the injection mass and pressure grow (i.e. for greater
340 engine load). As the load is raised to 9 bar in the high-speed end of the map (upper part-load
341 domain), an early SOI (between 318 and 321 CA deg BTDC) has to be used for mixture
342 preparation. The experimental data indicates a very sharp, ten-fold increase in soot number
343 density (10 million particles per cc) at these conditions. A possible explanation is that wall wetting
344 becomes an increasingly important source of particulate matter.

346 **3.2 Combustion Characteristics and Correlation with Soot Number Density**

347 The correlation between soot formation and engine operating variables such as spark timing and
348 injection strategy is evident and well-acknowledged [10, 15, 24, 28, 29, 30]. The following section
349 focuses on exploring correlations between relevant combustion indicators and engine-out soot
350 number density.

352 Flame Development Angle

353 The contour-plot map of FDA as a function of engine speed and load is presented in figure 6. The
354 development of combustion and its duration depend mostly upon the thermo-chemical state of the
355 charge at the start of combustion. At fixed engine speed, an earlier start of combustion leads to
356 smaller initial in-cylinder temperature and pressure, and ultimately to a longer development
357 process. Higher engine speed at fixed load leads to greater chamber turbulence, which shows
358 some beneficial effects on the rapidity of flame development; at the same time, greater speed
359 tends to stretch the process over wider crank angle windows [36]. As clear from figures 4 and 6,
360 the net outcome is that the FDA shows a strong degree of correlation with ignition advance; the
361 two quantities are linearly correlated and the FDA grows with a gradient of 0.7 CA degrees per 1
362 degree variation in spark advance. As engine speed and load are varied in the part-load envelope,
363 the spark timing strategy is such that changes to the location where fast burning commence are
364 minimised.

365
366 Despite being distorted by the bulk motion and corrugated by turbulence, during flame
367 development the growing burned volume V_b would be located approximately centrally, in a region
368 of high turbulence. In homogeneous charge mode, this region would be conceivably well-mixed,
369 and only a small proportion of soot is likely to form during the flame development stage. A cross-
370 check of figures 3 and 6 reveals that soot N/cc and FDA are, as expected, virtually independent of
371 each other.

373 **Figure 6**

375 Rapid Burning Angle and Unburned Gas Temperature

376 In a gasoline spark ignition engine most of the charge burns at a fast pace during the so-called
377 Rapid Burning Angle. It is during this phase, when the expanding flame front travels between the
378 spark plug area and the far corners of the combustion chamber, that most soot particles would be
379 nucleated. The duration of rapid combustion and the temperature of the unburned gases
380 approaching (and being entrained into) the flame front, would exert an influence on soot nucleation
381 rates if the process of mixture preparation remains unfinished when combustion commences. This
382 is more likely to happen at medium to high engine load, i.e. with greater fuelling rates. The
383 existence of sub-stoichiometric regions, segregated away from the central combustion space,
384 would promote the nucleation of carbonaceous particles at the arrival of the flame front. A longer
385 rapid burning duration leaves more time for mixture preparation, reducing soot formation. In this
386 case, an increasing proportion of the process takes place along the expansion stroke at reduced
387 unburned gas temperature. Because of smaller laminar flame velocity [36], the flame front travels
388 at reduced speed, reaching the remote areas of the combustion chamber in a longer time interval.
389 Reduced unburned gas temperature would also cause lower flame-front temperature, reducing the
390 soot nucleation rate via a second mechanism [15, 27, 30]. On the contrary, a shorter rapid
391 combustion interval would be located closer to TDC, animated by hotter unburned gases. Both
392 greater flame velocity and temperature would promote the nucleation of soot particles.

393

394 Two separate quantities, RBA and maximum unburned gas temperature, $(Tu)_{max}$ (corresponding to
395 the location of maximum in-cylinder pressure), are considered to further the analysis of soot
396 formation. The contour-plot maps of RBA and $(Tu)_{max}$ as a function of engine speed and load are
397 presented in figures 7 and 8, respectively. The unburned gas temperature is modelled from spark
398 timing, assuming polytropic compression during combustion. Figure 7 depicts a fast burning
399 engine, with RBA varying only between 14 and 21 CA degrees. As expected, RBA increases with
400 increasing engine speed, whereas it appears relatively constant as the BMEP is varied in the range
401 2.5 to 9 bar. The expected decrease in duration, due to greater charge density as engine load
402 rises, vanishes in the present case as an effect of decreasing spark advance, which typically would
403 cause a lengthier burning process [38]. As speed and load are varied, different ignition setting is
404 used so that peak pressure locates consistently in a narrow crank angle window after TDC (8 to 18
405 CA deg ATDC), ensuring high thermal efficiency [36].

406

407 **Figure 7**

408

409 **Figure 8**

410

411 Parallel examination of figures 3, 7 and 8 reveals a significant relationship between soot number
412 density and combustion characteristics. The results suggest that incomplete mixture preparation
413 enables increased nucleation at those conditions where the rapid burning phase is shorter and,
414 correspondingly, the unburned gas temperature is greater. This is particularly evident in the mid-
415 speed, mid-load region where a shorter and hotter rapid combustion causes the establishment of
416 an island of high soot N/cc. Remarkably, as the engine speed increases over or decreases below
417 3000 rev/min at medium load (between 4 and 5 bar bmep), rapid combustion slows down, enabling
418 lower $(Tu)_{max}$. As anticipated above, these two separate mechanisms discourage the nucleation of
419 soot particles. Consistent phenomena determine the characterisation of soot number density at low
420 speed (1600 to 2000 rev/min) and higher load (6.5 bar and above), i.e. in the top left corner of the
421 N/cc map. Here, retarded spark timing and quick development process cause short rapid
422 combustion and greater unburned gas temperature; as expected, on account of a high fuelling rate
423 at high load, an incomplete mixture preparation process enables more soot particles to be formed
424 during the RBA. As the engine speed is increased at high load (in excess of 7 bar), the RBA
425 extends into the expansion stroke towards regions of lower temperature. Correspondingly, soot
426 N/cc decreases as expected until a very different phenomenon, namely spray impingement, is
427 likely to occur. The experimental soot measurements suggest that the detrimental influence of
428 spray impingement supersedes the beneficial effects of high in-cylinder turbulence, lengthened
429 combustion duration and comparatively lower combustion temperatures.

430

431 Rapid Combustion up to Peak Pressure (RBP_{max})

432 Reference to RBA has been used so far as this represents a commonly used combustion duration
433 indicator. However, the data suggests that a separate parameter may be more relevant. Figure 9
434 shows the contour-plot map of RBP_{max} , calculated as the crank angle interval between 10% MFB
435 and the location of peak pressure. Peak pressure, compared to the location where 90% of the fuel
436 mass has been consumed, should provide a safer indication of the flame front reaching the furthest
437 walls of the combustion chamber. Past this point, the rate of combustion is forced to reduce,
438 alongside with the flame temperature. A comparative examination of figure 3 and 9 indicates that,
439 in spite of its narrow range of variation, limited to 4 CA degrees, RBP_{max} shows a remarkable
440 degree of correlation with soot number density. Bar the top-right corner of the N/cc map (potential
441 spray impingement) and the region where BMEP < 3 bar (small mass of fuel injected, enabling a
442 fuller mixture preparation), RBP_{max} and soot N/cc are inversely correlated and the regions of
443 greater N/cc are almost invariably identified by shorter RBP_{max} . If a relatively coarse (20 x 20) grid
444 is used for the cubic interpolation of the experimental data for soot N/cc and the calculated data for
445 RBP_{max} , a speculative distribution of N/cc as a function of RBP_{max} can be created, as presented in
446 figure 10. In spite of the high scatter, this linear relationship suggests that the bulk of the soot
447 formation process is restricted to, and would depend on the length of, the rapid burning process up
448 to the point where the bulk of the combustion chamber has been swiped by the expanding flame
449 front.

450

451 **Figure 9**

452

453 **Figure 10**

454

455 **3.3 Analysis of Engine-Out Soot Measurements – Particle Size and Mass**

456 The measurements of size-resolved soot number density distribution have been summarised in
457 figure 11, which shows the percent distribution of number density upon five relevant particle
458 diameter bins. In line with the relevant literature [10, 15, 24, 28, 29], the vast majority of soot
459 emitted by the wall-guided GDI test engine can be considered as primary particles in the 10 to 100
460 nm diameter range, whereas in excess of 55% is emitted in nucleation mode (below 50 nm).
461 Although the distribution of small particles (size between 10 and 20 nm) is highly fragmented, a
462 clear overall trend can be seen and, at all operating conditions except one, an average of 42% of
463 particles is emitted in the 20 to 50 nm diameter bin. Interestingly, a drop in number density of
464 particles greater than 100 nm is consistent for all the 16 cases. The particle size distribution shows
465 a slightly asymmetric shape, which peaks in the 20-50 nm size bin; similar trends have been
466 reported by Maricq et al. [15]. For the 3700 rev/min cases, less than 10% of the soot agglomerates
467 reach a size greater than 100 nm. The higher late-cycle oxidation, due to the higher exhaust gas
468 temperature, can explain the decrease in particle size as the speed is increased. The light load
469 cases of 30 Nm shift the distribution towards small particle size, i.e. below 50 nm, probably due to
470 the reduced locally rich regions of the fuel mixture and the burning of completely volatilized fuel
471 droplets. Nevertheless, the only engine condition to produce measurable particle number in the
472 size range above 300 nm (proper cluster or chain-like aggregation mode) was the lowest engine
473 speed/load point, i.e. 1600 rev/min and 30 Nm. Differences in nanoparticle size may reflect a
474 combination of different operating conditions, combustion temperatures and chemical species
475 contributing to particle growth at various stages. It is interesting to note that the peak values of
476 number density are much lower than typical 130 nm soot agglomerates from diesel engines [39,
477 40]; possibly because the hydrocarbons involved are more volatile compared to diesel.

478

479 **Figure 11**

480

481 The contour-plot map of exhaust soot Geometrical Mean Diameter, as a function of engine speed
482 and load, is given in figure 12. The analysis of GMD at part-load operating conditions is complex as
483 the influence of engine operating variables is quite subtle. The literature surveyed above suggests
484 two variables, namely ignition timing and exhaust gas temperature, are most significant in
485 determining soot particle size [15, 30]. The extent of their influence would depend on the rate of
486 nucleation, as high concentration of soot should enhance the coagulation and/or aggregation of
487 particles [15]. Contrarily to this premise, the data collected in the present work contains no strong
488 evidence of a positive association between soot concentration and size, as indicated by the GMD.
489 The correlation between engine speed, load and exhaust gas temperature, measured close to the
490 point of access for soot emissions sampling, is presented in figure 13. Spark timing affects how the
491 combustion process is phased about TDC, determining the resulting post-combustion gas
492 temperature. The influence of ignition timing on exhaust temperature is, in the present case,
493 mitigated by the changes in engine speed and load. Nevertheless, the exhaust gas temperature is
494 seen to increase with increasing engine speed and load. As explained below, this has the effect of
495 promoting post-flame oxidation [15, 30].

496

497 **Figure 12**

498

499 **Figure 13**

500

501 In figure 12 three GMD regions have been identified. Region 1, at low engine speed and mid to
502 high load, is characterised by greater GMD in the range 50 to 60 nm. Region 2 is the upper part-
503 load domain, where spray impingement is likely to be dominant; here, the GMD sharply decreases
504 reaching size as low as 23 nm. Region 3, at medium to low load and increasing speed, sees a
505 relatively constant average particle size, between 35 and 40 nm. In region 1, as the engine load is
506 increased at constant speed, the GMD firstly grows reaching a peak at 6.5 bar BMEP, and then

507 decreases. Remarkably, a strong resemblance is found between the distribution of GMD and that
508 of unburned gas temperature, $(Tu)_{max}$ (see figure 8). As suggested by Pei et al. [33], greater
509 combustion temperature (of which $(Tu)_{max}$ gives an indication) would favour the formation of
510 particles in the accumulation mode. The inherent stratification of fuel at low speed and mid-to-high
511 load leads to greater particle inception, while simultaneously the high combustion temperature
512 enhances coagulation and/or agglomeration [41]. This is also suggested by the results presented
513 in figure 11, where for an engine speed of 1600 rev/min and a load of 90 Nm, 95% of particles are
514 emitted with size above 20 nm. The relatively low incidence of post-flame oxidation at low engine
515 speed (see figure 13) allows the particles to conserve an increased size when emitted. As the
516 GMD map is swept to the right of region 1, temperature $(Tu)_{max}$ decreases, and the GMD also
517 decrease, but with a smaller gradient of descent, possibly mitigated by the effects of increasing
518 speed as well as earlier fuel injection [15, 24, 30]. As the engine speed is further increased, the
519 influence of greater exhaust temperature on post-flame surface oxidation becomes more important,
520 reaching its apex in region 2, for temperature in excess of 1000 K. Comparative examination of
521 figures 3, 11 and 12 reveals that, at high speed and load, high concentrations of exhaust soot (up
522 to 10 million particles per cc) are emitted, mostly with very small size in the nucleation mode range
523 (between 23 and 40 nm). High fuel stratification due to extended spray-wall interaction, along with
524 high exhaust gas temperature, are thought to be responsible for the “upper part-load” soot particle
525 characterisation. As shown in figure 11, at the highest engine speed and load investigated (3700
526 rev/min and 120 Nm), only 10% of particles are emitted with size above 50 nm. As the “upper part-
527 load” operating range is often swept across as a result of typical stop-and-run driving pattern in
528 urban areas, region 2 must be identified as an area of concern. The characterisation of GMD in
529 region 3 is less clear. The experimental results suggest that, as the engine speed is increased at
530 light-to-mid load, the influence of worsened mixing, which would normally lead to greater average
531 size [41], is mitigated by that of higher oxidation, as a result of higher exhaust gas temperature. In
532 agreement with Pei et al. [33], advanced ignition timing and greater combustion temperature bear
533 no visible influence on the average size of the engine-out soot in this region. The net result of such
534 multi-faceted phenomena is that in region 3 the GMD remains approximately constant.
535

536 Figure 14 shows the contour-plot map of soot mass concentration in $\mu\text{g}/\text{cc}$ as a function of engine
537 operating conditions. Bar the high speed/high load impingement area, the mass concentration
538 closely follows the distribution of particles number density and, to a lesser extent, that of average
539 particle size. In terms of soot mass, two regions of concern can be identified. The first one, at high
540 engine load and low speed, assumes a distinctive triangular pattern where, relatively to the lowest
541 baseline value, soot mass concentration shows a sharp four-fold increase. The second one, at
542 moderate speed and load, corresponds to the island of high number density shown in figure 3.
543 Here, soot mass concentration shows a three-fold increase. Finally, due to very low GMD, low soot
544 mass is emitted in the upper part-load corner, in spite of very high particle number.
545

546 **Figure 14**

547
548 Further work, already being undertaken by the Authors, aims at a more comprehensive
549 assessment of the connection between mixture preparation, combustion characteristics and soot
550 formation. Further work is carried out on two fronts. On one hand modelling work, using CFD for a
551 deeper insight into mixture preparation and combustion; on the other hand further experimental
552 work, aimed at measuring engine-out soot over a more refined part-load grid.
553

554 **4. Conclusions**

555
556
557 Experimental measurements of size-resolved soot number density distribution have been taken
558 from a modern Euro IV, wall-guided GDI engine, using 95 RON gasoline fuel. Engine tests covered
559 a large portion of the part-load running envelope, with engine speed in the range 1600 to 3700
560 rev/min and torque in the range 30 to 120 Nm (BMEP between 2.36 and 9.44 bar). All tests were
561 carried out at steady-state, fully-warm conditions, and the engine was operated in stoichiometric
562 and homogeneous mode, with fuel injection early in the intake stroke. The primary aim of this study

563 has been to investigate the correlation between engine-out soot measurements and combustion
564 characteristics.

565

566 The results indicate the existence of a linear correlation between rapid combustion duration and
567 total soot particle number concentration for engine load in excess of 3 bar BMEP. This is enabled
568 by incomplete gas-phase mixture preparation at the start of combustion. Higher total particle
569 number density is invariably correlated to faster rapid combustion. On average, a six-fold increase
570 in number density between 1.0 and 6.0×10^6 particles per cc arises from shortening the rapid
571 duration of 4 CA degrees. A clear association is also found between greater unburned gas
572 temperature feeding the flame-front and greater rate of soot formation.

573

574 The correlation between combustion characteristics and soot number density disappears in the
575 upper part-load range, for engine speed in excess of 3000 rev/min and load in excess of 7 bar
576 BMEP. Very early SOI (315 CA deg BTDC or earlier), used to enable better mixing at high speed
577 and load, causes fuel spray impingement, possibly wall wetting and much higher soot nucleation
578 rates, likely as a result of pool-fire.

579

580 The total soot number density measured at part-load was between 1.0×10^6 and 1.0×10^7 particles
581 per cc, with the greatest levels measured in the upper part-load range. The Geometrical Mean
582 Diameter varied between 23 and 60 nm. The greatest average particle size, between 50 and 60
583 nm, is measured at low engine speed and mid to high engine load. Particles emitted in the upper
584 part-load domain were on average very small (GMD between 40 and 23 nm).

585

586 In terms of soot emissions, three main regions of concern have been identified within the part-load
587 running envelope: 1. the higher load-lower speed range and 2. the mid load-mid speed range,
588 where high nucleation rates induce copious increases of engine-out soot mass; 3. the upper part-
589 load corner, where the highest levels of particle numbers were recorded.

590

591 Beside the necessity of improved combustion systems design, the results suggest that for given
592 part-load engine conditions, alterations of combustion duration may be induced to conceivably
593 obtain sizeable reduction of exhaust soot. As the more recent spray-guided GDI technology
594 appears to suffer, though to a lesser extent, from similar soot formation mechanisms (imperfect
595 mixing, pool-fire), the analysis presented here carries wider potential relevance.

596

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734 **Nomenclature**

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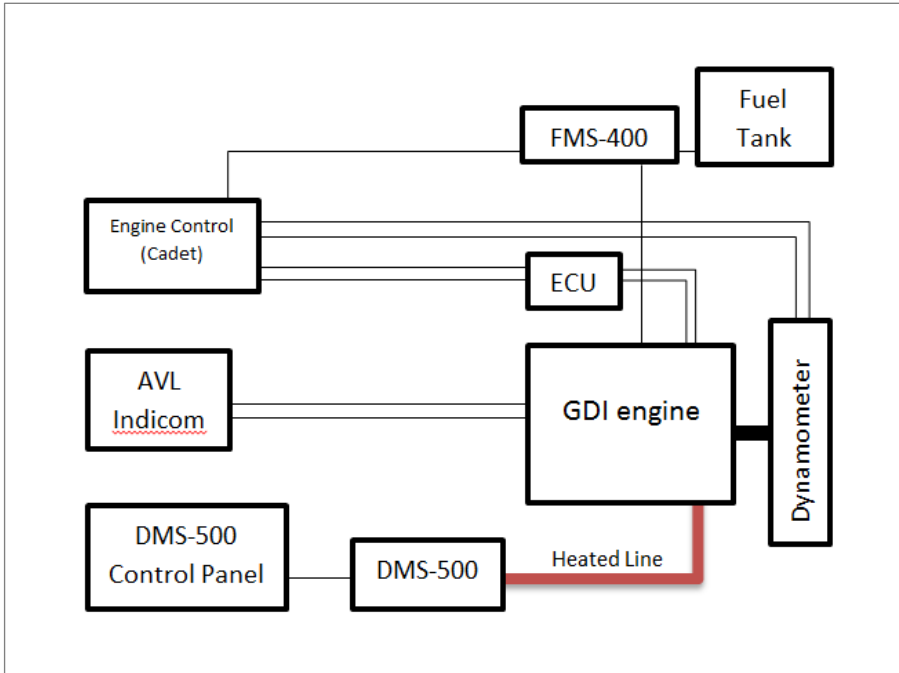
AFR	Air Fuel Ratio
ATDC	After Top Dead Centre
BMEP	Brake Mean Effective Pressure
BTDC	Before Top Dead Centre
CA	Crank Angle
CPC	Condensation Particle Counter
DISI	Direct Injection Spark Ignition
DMA	Differential Mobility Analyser
DMS	Differential Mobility Spectrometer
DPF	Diesel Particulate Filter
ECU	Engine Control Unit
ELPI	Electrostatic Low Pressure Impactor
EOC	End of Combustion
EOI	End of Injection
FDA	Flame Development Angle
GDI	Gasoline Direct Injection
GMD	Geometric Mean Diameter
IT	Injection Timing
IVC	Intake Valve Closing
MFB	Mass Fraction Burned
NEDC	New European Drive Cycle
PAH	Polycyclic Aromatic Hydrocarbons
PFI	Port Fuel Injection
PM	Particulate Matter
RBA	Rapid Burning Angle
RBP_{max}	Rapid Burning Angle at P _{max}
SMPS	Scanning Mobility Particle Sizer
SOC	Start of Combustion
SOI	Start of Injection

- ST** Spark Timing
- TDC** Top Dead Centre
- TEM** Transmission Electron Microscopy

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737 **Figures**

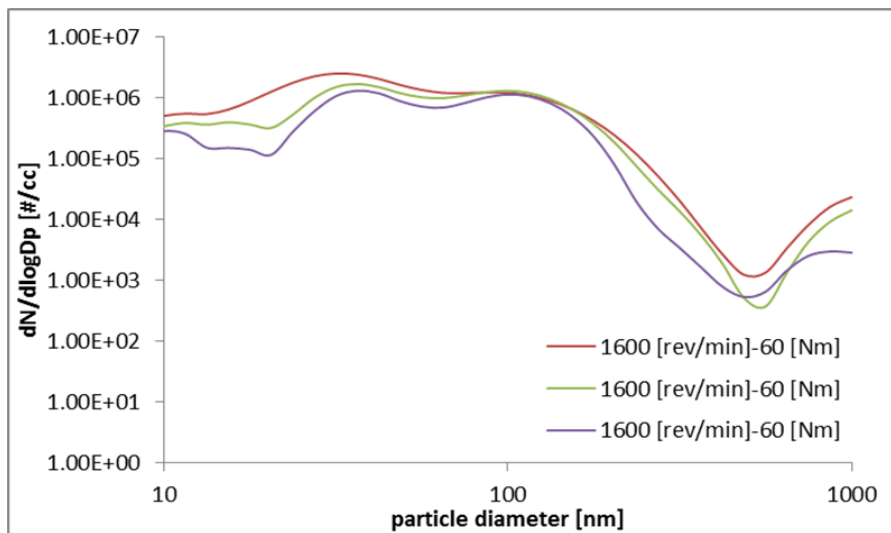
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740 Figure 1. Schematic of engine rig experimental setup

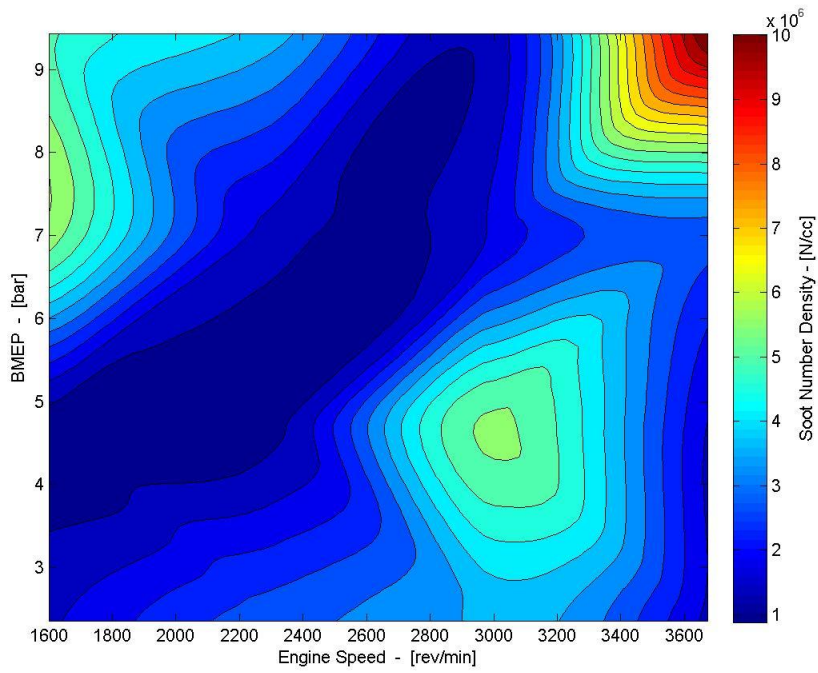
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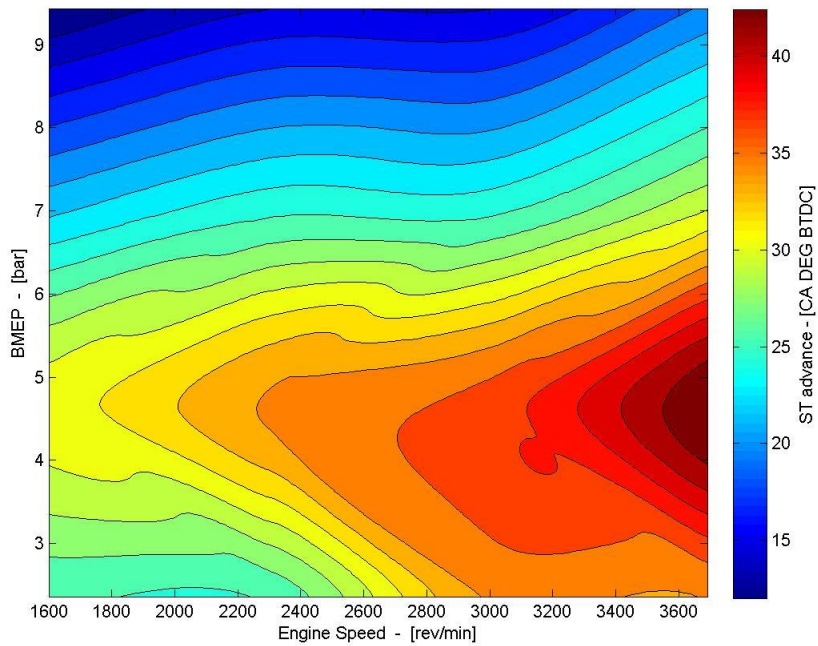
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743 Figure 2. Repetitions of time-averaged, size-resolved engine-out soot number density
 744 distributions, at engine speed of 1600 rev/min and load of 60 Nm. On the horizontal axis is the
 745 electrical mobility equivalent diameter in nm; on the vertical axis, the number density N/cc,
 746 normalized with the differential interval of the logarithm of particle size, $dN/d\log D_p$.

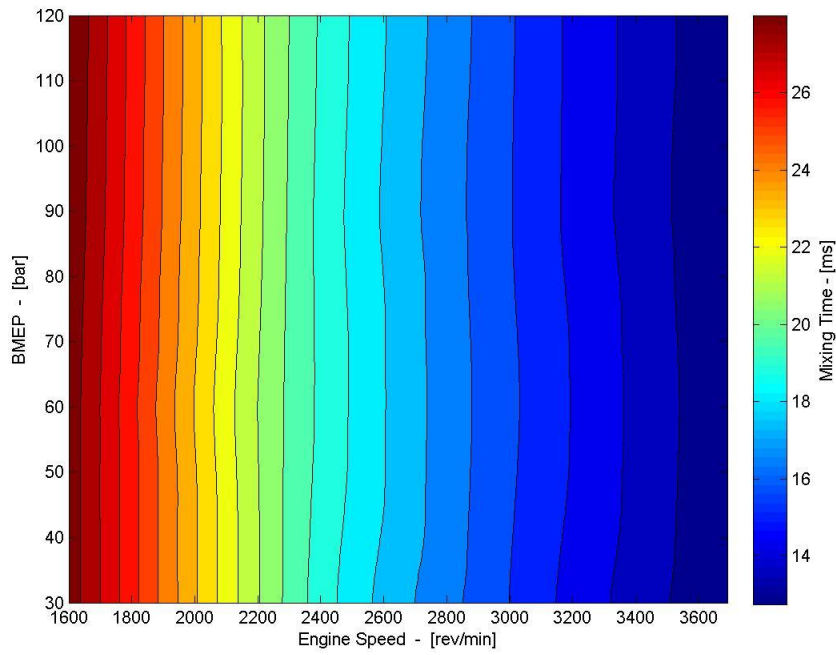
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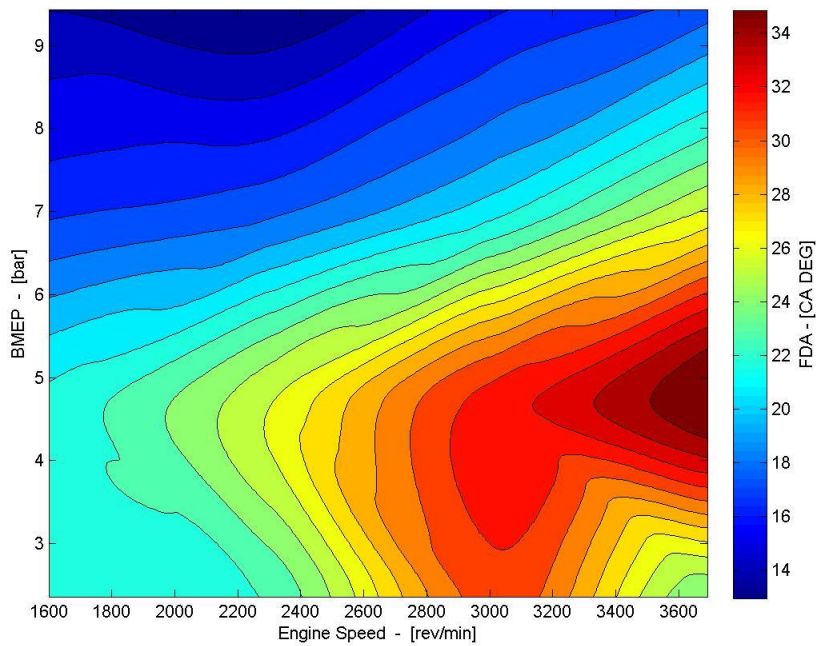
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 749 Figure 3. Experimental soot number density as a function of engine speed and load (BMEP).
 750 Measured particles with diameter smaller than 10 nm are excluded from the analysis.
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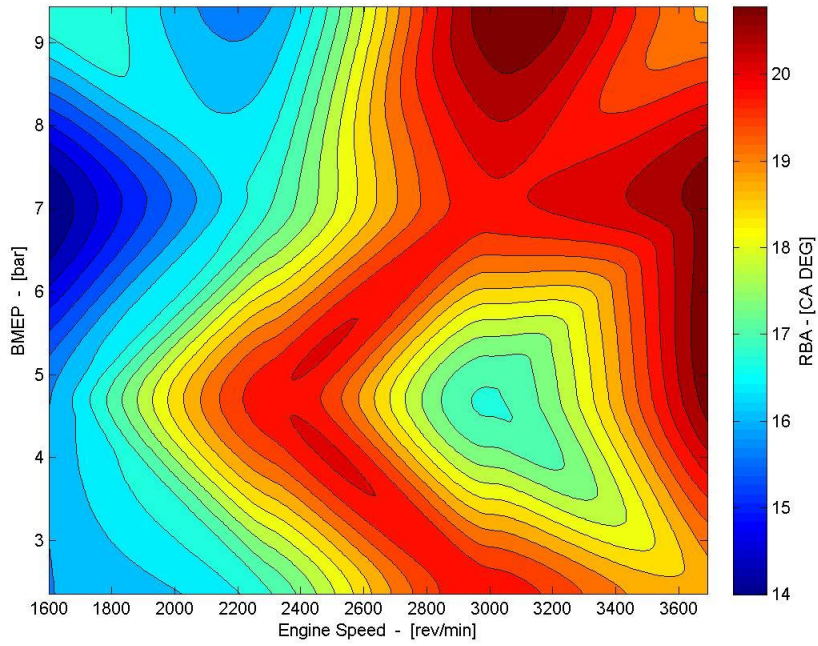
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 753 Figure 4. Spark Timing (ST) advance as a function of engine speed and load (BMEP).
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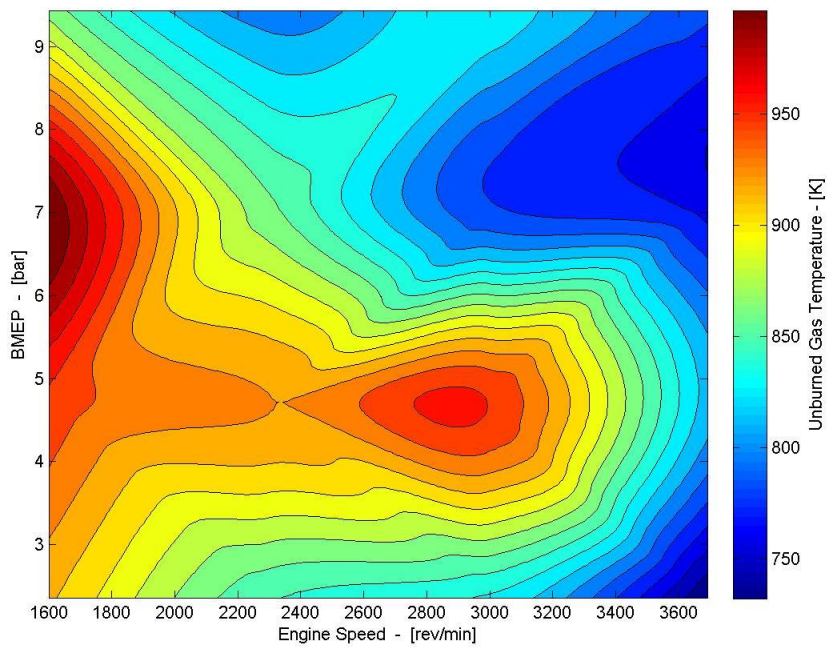
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 756 Figure 5. Mixing time (time between EOI and TDC of combustion) as a function of engine speed
 757 and load (BMEP).
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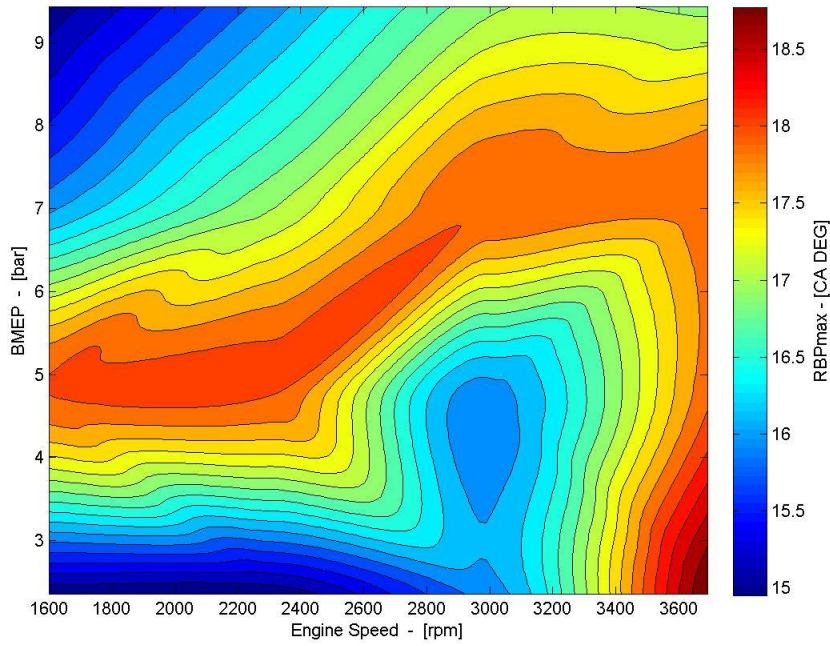
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 760 Figure 6. Flame Development Angle (FDA), as a function of engine speed and load (BMEP).
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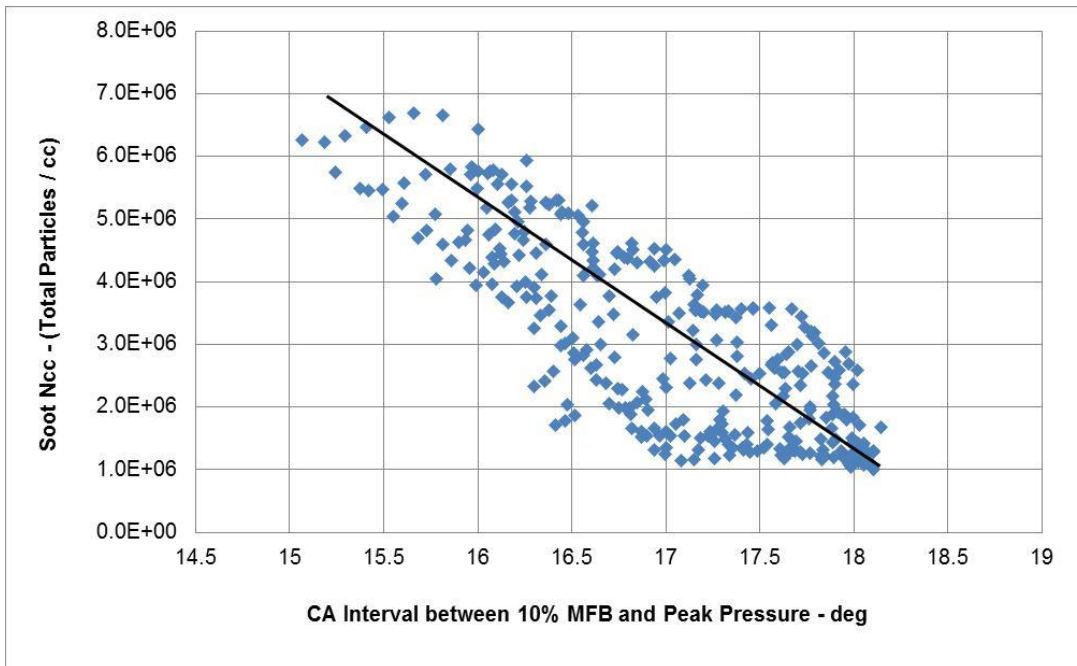
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763 Figure 7. Rapid Burning Angle (RBA), as a function of engine speed and load (BMEP).
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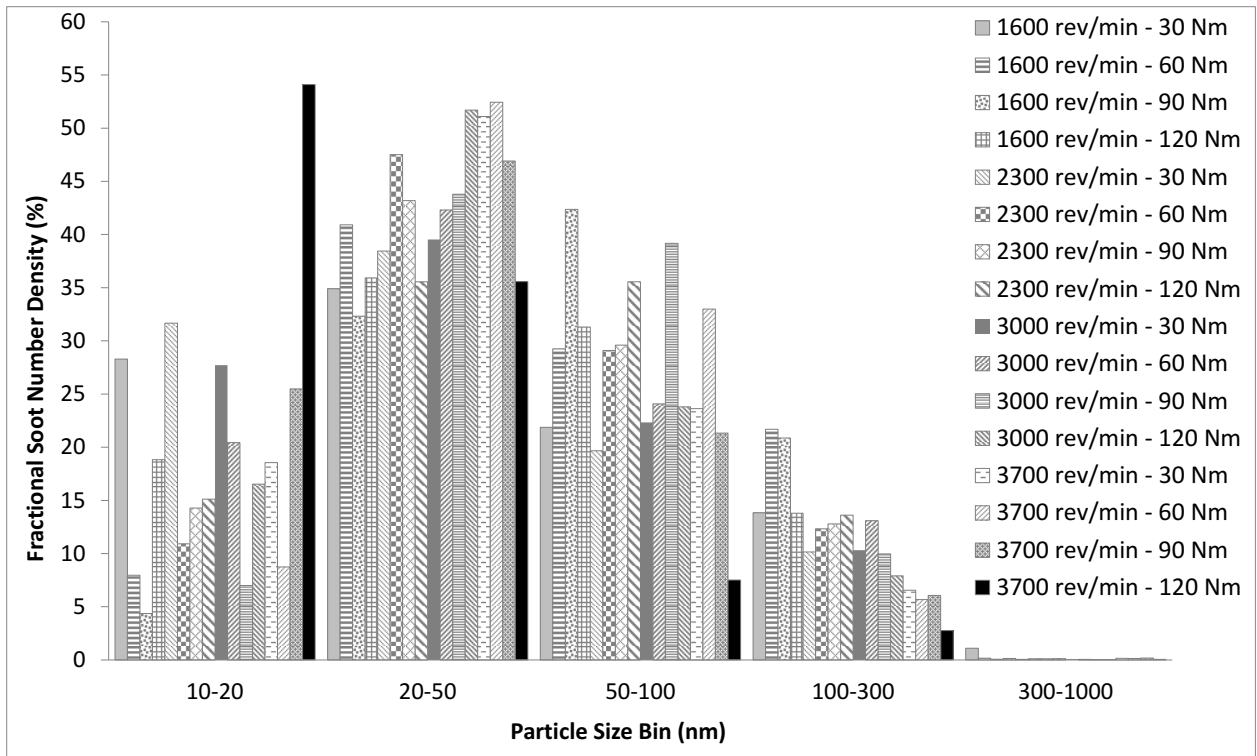
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766 Figure 8. Modelled unburned gas temperature at the end of the rapid combustion phase, as a
767 function of engine speed and load (BMEP).
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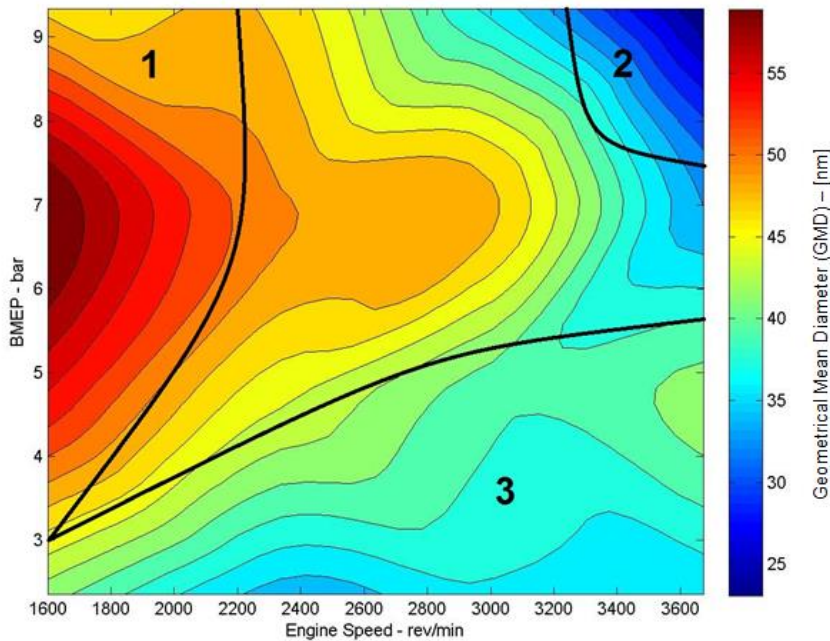
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 770 Figure 9. Crank angle interval between 10% MFB and location of peak pressure (RBP_{max}), as a
 771 function of engine speed and load (BMEP).
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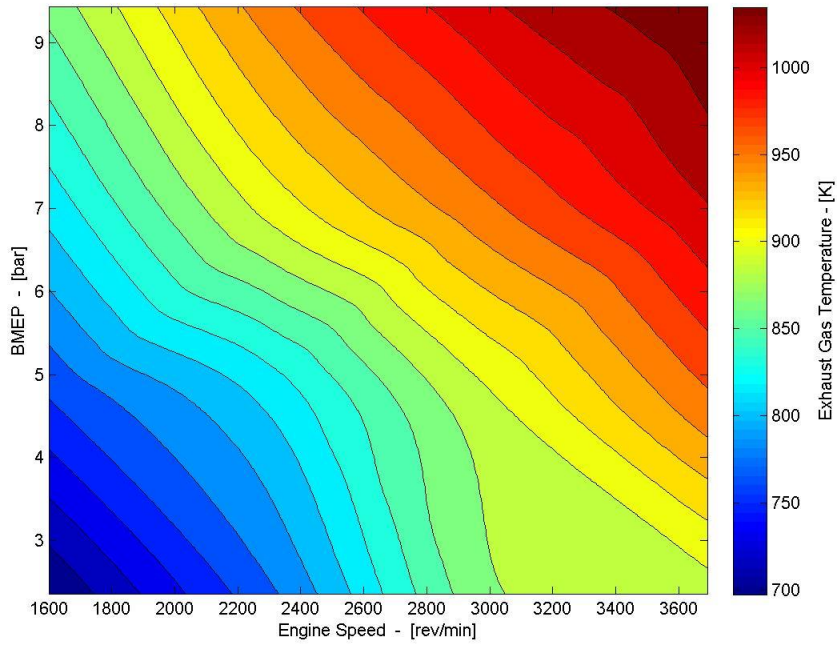
774
 775 Figure 10. Correlation between exhaust soot number density and RBP_{max}. Points are derived by
 776 applying a classic (20 x 20) grids for the cubic interpolation of the two experimental quantities.
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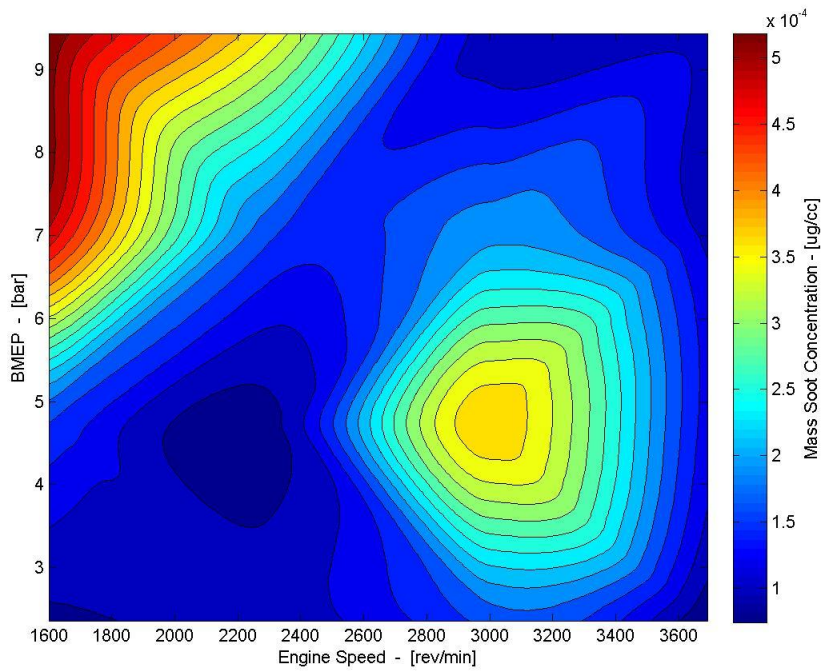
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 780 Figure 11. Fractional soot number density (%), as a function of 5 particle size bins (particle
 781 diameter in nm), for 16 engine running conditions. Measured particles with diameter smaller than
 782 10 nm are excluded from the analysis.
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784
 785 Figure 12. Experimental Soot Geometrical Mean Diameter (GMD), as a function of engine speed
 786 and load (BMEP). Measured particles with diameter smaller than 10 nm are excluded from the
 787 analysis.
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790 Figure 13. Measured exhaust gas temperature, as a function of engine speed and load (BMEP).
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792
793 Figure 14. Computed soot mass concentration, as a function of engine speed and load (BMEP).
794 Measured particles with diameter smaller than 10 nm are excluded from the analysis.
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805 **Tables**

806

Displacement (cm ³)	1598
Bore (mm)	77
Compression ratio	10.5:1
Connecting Rod Length (mm)	138.4
Combustion Chamber	4-Valve, Central Spark Plug, Pent-Roof Design
Engine type	In-line 4-cylinder
Cycle	4-stroke Spark Ignition
Fuel Injection system	Direct Injection Common Rail
Fuel Injectors	Side-mounted, wall-guided spray
Maximum Injection Pressure (bar)	120
Maximum Engine Speed (rev/min)	6000
Maximum Rated Torque (Nm)	240

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Table 1. Test engine technical specifications