Study on Pollutants Formation under Knocking Combustion Conditions using an Optical Single Cylinder SI Research Engine

3 Apostolos Karvountzis-Kontakiotis^{1,2,*}, Hassan Vafamehr¹, Alasdair Cairns³, Mark Peckham⁴

4 1. Brunel University London, Department of Mechanical, Aerospace & Civil Engineering, CAPF – Centre of

5 Advanced Powertrain and Fuels, Uxbridge, UB8 3PH, United Kingdom

6 2. City University London, School of Mathematics, Computer Science and Engineering, Northampton Square,

7 London EC1V 0HB, United Kingdom

8 3. Faculty of Engineering, University of Nottingham, University Park, Nottingham NG7 2RD, United Kingdom

9 4. Cambustion Ltd., 347 Cherry Hinton Road, Cambridge CB1 8DH, United Kingdom

10 * Corresponding author email: <u>a.karvountzis@brunel.ac.uk</u>

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12 Abstract

The aim of this experimental study is to investigate the pollutants formation and cyclic 13 emission variability under knocking combustion conditions. A great number of studies 14 extensively describe the phenomenon of knock and its combustion characteristics as well as 15 the effect of knock on engine performance; however the impact of knocking combustion on 16 pollutants formation and how it affects cyclic emission variability has not been previously 17 explored. In this study, an optical single cylinder SI research engine and fast response analyzers 18 were employed to experimentally correlate knocking combustion characteristics with cyclic 19 resolved emissions from cycle to cycle. High-speed natural light photography imaging and 20 simultaneous in-cylinder pressure measurements were obtained from the optical research 21 engine to interpret emissions formation under knocking combustion. The test protocol included 22 the investigation of the effect of various engine parameters such as ignition timing and mixture 23 air/fuel ratio on knocking combustion and pollutant formation. Results showed that at 24 stoichiometric conditions by advancing spark timing from MBT to knock intensity equal to 6 25 26 bar, instantaneous NO and HC emissions are increased by up to 60% compared to the MBT operating conditions. A further increase of knock intensity at the limits of pre-ignition region 27 was found to significantly drop NO emissions. Conversely, it was found that when knocking 28 combustion occurs at lean conditions, NO emissions are enhanced as knock intensity is 29 increased. 30

- 32
- **Keywords:** knocking combustion, cycle resolved emissions, NO formation, optical research
- 34 engine, cyclic emission variability
- 35
- 36

37 Highlights

- Experimental investigation of knocking combustion and cycle resolved emissions.
- Effect of knock index on NO formation.
- Utilization of instantaneous flame images to interpret emissions formation.
- Effect of knock and heavy knock conditions on cyclic emission variability.
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45 Nomenclature

Acronyms			
ATDC	After Top Dead Centre	imep	Indicated mean effective pressure
Bmep	break mean effective pressure	KI	Knock Intensity
BTDC	Before Top Dead Centre	MBT	Maximum Breaking Torque
CCV	Cyclic Combustion Variability	NO	Nitric Oxide
CEV	Cyclic Emission Variability	SI	Spark Ignition
COV	Coefficient of Variation	TDC	Top Dead Centre
HC	Hydrocarbons	λ	Air/fuel equivalence ratio

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- 48

50 1 Introduction

51 Gasoline engine technology has entered one of the most exciting periods in its long history: 52 downsizing, hybridization and stricter emission standards determine the future of SI engines [1]. Advanced engine requirements for high power density and low fuel consumption can be 53 54 achieved through high boost, direct injection, engine start&stop [2] and lean burn operation. Implementation of this technology in SI engines increases the potential possibility that under 55 certain operating conditions auto-ignition or pre-ignition can occur [3]. Knock not only limits 56 engine thermal efficiency but it also affects the formed emissions during combustion. As 57 pollutants formation is directly linked to the combustion process, it is critical to understand the 58 59 effect of abnormal combustion cycles on emissions performance and variability. To this end, meeting fuel economy requirements and future emission standards in high-efficiency SI 60 engines requires a well understanding of knock and its effect on engine emissions. 61

Knock is as old as SI engine itself [4] and takes its name from the metallic 'pinging' noise that 62 63 auto-ignition (spontaneous combustion) creates before the piston reaches TDC. Knock can be divided into two main groups: light to medium knock and super knock. Light to medium knock 64 65 limits compression ratio and as a consequent the engine thermal efficiency due to the end-gas auto-ignition. Auto-ignition is the fast discharge of chemical energy contained in the end-gas, 66 67 which is the final fraction of the air-fuel mixture that enters the cylinder but without inclusion 68 into the flame front reaction [5], [6]. On the other hand, super knock limits raising the boost pressure and the engine power density due to detonation, also known as pre-ignition [7], [8]. 69 Pre-ignition occurs earlier to auto-ignition and leads to a fast and violent combustion that can 70 potentially damage the engine. Apart of the great number of studies on knocking combustion, 71 the correlation between knock intensity, heat transfer, fuel chemistry, pressure oscillations and 72 oil droplets is not well understood [3]. Nowadays, the main research focuses on super knock 73 due to the high boost technology applied in high power density downsized engines, which 74 usually occurs under low speed and high load engine operating conditions. 75

Fuel and cylinder charge properties as well as engine calibration can affect combustion characteristics and simultaneously affect the formed emissions. Methods for suppressing engine knock and controlling emissions involve the optimization of those parameters. Conventional suppression methods of engine knock include control strategies for retarding spark timing [9], raising fuel octane number by using additives [10], [11] and enriching mixture [12] when knock occurs. Recent studies correlated the effect of cooled EGR with the reduction of knocked cycles and fuel consumption. Diana et al. presented a knock free operation of a 83 stoichiometric SI engine at full load conditions with a compression ratio of 12.5 by using cooled EGR [5]. Same trend was also observed in another experimental study where it was 84 shown that the resistance to knock is strongly increased by cooled EGR [13]. It was also found 85 that utilization of EGR can significantly reduce emissions by up to 90% [5], [14], [15], while 86 HC emissions increase across the EGR range [16], [17]. Air-fuel ratio can also affect engine 87 knock limit. Although leaner mixtures benefit fuel consumption, retarded combustion is 88 required to avoid knock [6], while over-fuelling increases the knock limits of the engine [18]. 89 In another study, it is reported that the EGR and the lean burn operation exhibit lower average 90 91 knock indexes. Due to the longer combustion duration by utilizing EGR and lean burn, heat transfer is enhanced and end-gas temperature is decreased [19]. Conversely, another 92 experimental work using a single cylinder research engine that was supplied with ON 75 93 proved that lean mixtures have the earliest onset of knock, and the highest knock intensity [20]. 94 Furthermore it was noted that lean mixtures are particularly sensitive to the charge heating 95 during compression. Overall, this research implies that excess air at high loads does not 96 suppress knock. 97

More than 700 research studies exist in literature on the research area of engine knock [3]. The main research studies that deal with engine knock can be categorized on knock detection, numerical simulation, optical diagnostics, theoretical studies, engine optimization and fuel/oil properties. Although knocking combustion can significantly affect formed emissions, the effect of knocking combustion on pollutants formation is at an even earlier research stage and there is a significant lack of studies in this research field.

104 Pollutants formation during engine combustion is correlated both with mixture properties such 105 as air/fuel ratio, residual gas fraction and fuel properties and with the combustion type. At normal combustion conditions (deflagration), NOx formation is primarily controlled by oxygen 106 107 availability under lean or rich conditions while within the stoichiometric window formed NOx concentration is correlated with the combustion burn rate [21]. Under no knocking combustion, 108 previous studies showed that as in-cylinder peak pressure is increased, NOx emissions are 109 proportionally increased [21]-[23]. However under knocking combustion, the emissions 110 111 formation mechanism can significantly change. As knock intensity is increased, residence time of the residual gases in the burned zone is significantly decreased and peak temperature can be 112 113 enhanced. On the one hand, under super knock conditions combustion duration is decreased to less than 2° CA which means that combustion is almost isochoric, post-combustion residence 114 time is significantly lower but peak temperature and pressure are much higher than in normal 115

SI combustion. Such a type of combustion presents many similarities with HCCI combustion that is known for the ultra-low NOx emissions and the relatively higher CO and HC emissions [24]. On the other hand, under light knock conditions NOx concentration that is formed in the post-flame zone during normal combustion is expected to be increased due to the high temperatures of end gas auto-ignition. However, the above described trends have not been explored in literature and more research is required to well understand the relationship between pollutants formation and knock index.

123 A great number of research studies investigate the main mechanisms of pollutants formation, described in detailed elsewhere [25]. The most well-known NOx formation mechanism is the 124 125 thermal one, co-called as extended Zeldovich mechanism which is responsible for the majority of the formed NO in the post flame gas zone [26], [27]. Apart from the thermal mechanism, 126 127 the prompt NO mechanism [28] can be significant at stoichiometric and richer mixtures [29], [30], the N₂H mechanism becomes important at slightly rich conditions [31] and finally the 128 129 N₂O chemical pathway appears at slightly lean conditions [26], [32]. Regarding carbon monoxide, CO formation is mainly kinetically controlled by water gas shift reaction [33]. 130 Finally, hydrocarbons (HC) are the consequence of incomplete combustion of hydrocarbon 131 132 fuel [34]. In most studies, simplified chemical mechanisms are utilized to predict pollutants formation [29], [35]. Recently, Karvountzis et al. [36], [37] proposed the use of a detailed 133 chemical kinetics model to predict pollutants formation in an SI engine, which operates in the 134 post-combustion zone. The latter is the most accurate state-of-the art emission model existing 135 in literature, as the validation against experimental cycle resolved emission values for both NO 136 and CO emissions under various engine operating conditions showed errors less than 10% and 137 compared to simplified emissions models improvements were higher than 50% [38]. However 138 the accuracy of this emission model under knocking conditions has not been explored, due to 139 140 the lack of experimental data.

Although the relationships between combustion characteristics and pollutant formation under 141 142 normal combustion have been explored in the past, the effect of knocking combustion on 143 pollutants formation is not well understood. Few studies in the past presented an engine model 144 that can predict knock limit and NOx emissions [39], [40], [41]; however those models do not present the pollutant formation under knocking combustion conditions. The only known 145 146 relationship is that knock results to an increase in CO and HC emissions due to incomplete combustion [42]. Furthermore, there is a lack of experimental data at this research field, as 147 there are no experimental studies presenting the correlations between knock index and 148

149 emissions performance on a cycle to cycle base. Simultaneously, the effect of pressure oscillations and hot spots (occurring during knocking combustion) on pollutants formation is 150 also unknown. In the past, fast response analyzers were successfully employed to measure 151 cyclic emission variability under normal combustion conditions [43]. Optical diagnostics have 152 been also utilized to deeper understand the end gas auto-ignition mechanism and explore the 153 detonation limits at pre-ignitive cycles [7]. However, as authors are aware, a study that employs 154 both fast response analyzers to detect emissions from cycle to cycle and optical diagnostics to 155 correlate combustion characteristics with cycle resolved emissions doesn't not exist in 156 157 literature.

158 This experimental work studies the correlation between pollutants formation and knock index, primarily NO and HC, under various air-fuel ratio and spark ignition timings using an optical 159 single cylinder SI engine. Fast response analyzers were employed to measure the cycle resolved 160 NO and HC emissions. Images of the visible light from combustion heat were captured through 161 162 an overhead window using a high speed camera. The aim of this study is to give insights between the type of combustion (deflagration, end gas auto-ignition and detonation) and the 163 formed emissions and determine by which extent emissions are increased under knock. The 164 output of this study can be applicable to new engine design requirements for more efficient and 165 166 less pollute SI engines. To the best knowledge of the authors, this is a unique study of its kind.

167 **2** Experimental Setup

168 2.1 Optical Engine

A customized single cylinder research engine with a distinct optical arrangement was employed 169 170 in this study [18], [44], [45]. The basic engine characteristics of the engine are presented in 171 Table 1. The bottom-end of the engine is based on a commercial Lister-Petter TSI with a combined full-bore overhead access and a semi-traditional poppet-valve valvetrain. Due to the 172 full-bore overhead optical access, the glass of the engine head was designed to withstand peak 173 combustion pressures up to 150bar. The engine incorporates a flat piston crown, two inlet ports, 174 175 and originally was designed with two exhaust ports. As the purpose of this study is focused on knocking combustion, one of the exhaust valves was deactivated in order to assist end-gas 176 autoignition, by increasing residual gas fraction [46]. A schematic presentation of the optical 177 cylinder head is illustrated in Figure 1. The recessed side mounted poppet valves guaranteed 178 valve overlap without piston clash and to maintain a compression ratio of 8.4:1 [45]. Last but 179

- 180 not least, the research engine was coupled to an eddy-current dynamometer with a maximum
- 181 power absorption/supply of 10kW.





Figure 1: Schematic presentation of the optical engine cylinder head [18].

Table 1: Engine characteristics of the optical single cylinder research engine.

Parameter (unit)	Value
Number of Cylinders (-)	1
Compression Ratio (-)	8.4:1
Stroke (mm)	89
Bore (mm)	95
Displacement (cc)	631
Con-rod Length (mm)	165.16
Valve Lift (mm)	5
Inlet Valve Openings/Closing (°BTDC)	375/145
Exhaust Valve opening/Closing (°ATDC)	120/350

185 The engine fuel supply was regulated by a Bosch EV6 PFI fuel injector operating at 3 bar rail pressure. The PFI injector had a 2-hole pattern forming a dual plume spray pattern targeted 186 toward the intake valves. The injection timing was fixed at 400° BTDC firing under all 187 operating conditions. The fuel used in these tests is PRF75, as a result of blending isooctane 188 and n-heptane (provided by a chemical supplier) on a volumetric basis in small batches 189 immediately prior to each testing session. The chemical properties of the reference fuels are 190 provided in Table 2. The ignition system consisted of an NGK ER9EH 8mm spark plug 191 192 supplied by a Bosch P100T ignition coil.

193 Table 2: Fuel properties

Property	Iso-Octane	n-Heptane
Chemical Formula	C8H18	C7H16
Boiling point at 1 atm (°c)	99.2	98.4
Enthalpy of vaporisation at 298.15 K (MJ/Kmol)	35.14	36.63
Density at 25°C (Kg/m ³)	690.4	681.5
Latent Heat of Vaporisation(kJ/kg)	308	318
Reid Vapour Pressure (bar)	0.52	0.12
Oxygen Content by Weight (%)	0	0
Volumetric Energy Content (MJ/l)	30.6	30.48
RON (MON)	100 (100)	0 (0)

194

195 2.2 Experimental Configuration and Measurements

Experiments were conducted using the experimental setup schematically illustrated in Figure 196 2, while the main specifications of the relevant components are presented in Table 3. The test 197 198 protocol included the recording of high frequency data, low frequency data and natural light photography data. The high frequency data were recorded in a National Instruments (NI) data 199 200 acquisition card and included the in-cylinder pressure, encoder crankangle signal, exhaust temperature and exhaust NO and HC emissions recordings from the fast response analyzers. 201 202 The low sampling frequency data comprised ECU derived signals, including air/fuel ratio value $(\lambda \text{ sensor})$, ignition timing, throttle position, inlet air pressure and temperature and coolant 203 204 temperature. Recording also included the high frequency photography data of the combustion

evolution. Finally, the three (3) set of data are stored in a computer as it is illustrated in Figure

206 **2**.



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An AVL GH14DK pressure transducer was implemented on the side of the exhaust valves to 210 measure cylinder pressure (Figure 1). It was manufactured to fit this particular handmade 211 212 engine cylinder head and its position was at the region where autoignition usually takes place. It has to be mentioned that when knock occurs, pressure distribution across the combustion 213 214 chamber is not uniform and the location of the transducer can affect the recordings [47]. The 215 signal from the transducer was fed to an AVL Flexifem charge amplifier. The error in the 216 cylinder pressure measurement was less than $\pm 1\%$ full scale reading (FSO). The exhaust gas temperature was measured with a K-type thermocouple connected to the relevant transducer, 217 218 with maximum error of $\pm 0.75\%$ of reading. The response time of the thermocouple was low (0.1sec); therefore the thermocouple was utilized to measure averaged temperature per cycle 219 rather than monitoring exhaust temperature profile. The total experimental rig is presented in 220 Figure 3. 221

Fable 3: 1	Main	specifications	of	measurement	equipment.
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In-cylinder Pressure Sensor: AVL GH14DK		
Pressure range	0-300 bar	
Natural Frequency	~ 170 kHz	
Linearity	$\leq \pm 0.3\%$ full scale output (FSO)	
Sensitivity Drift	<u>≤±1%</u>	
Pressure Sensor Amplifier: A	VL Flexifem INDI	
Linearity	$\leq \pm 0.5\%$ full scale output (FSO)	
NO fast response analyzer: Cambustion CLD500		
Response time	NO: 2ms, NOx: 8ms	
Linearity	≤±1% full scale output (FSO)	
HC fast response analyzer: Cambustion HFR400		
Response time	HC(C1): 4ms	
Linearity	≤±1% full scale output (FSO)	
Natural Light Photography: Memrecam fx-6000		
Image Resolution	512×384 pixels.	
Frame Speed	6000 (fps)	





229 High frequency cycle resolved exhaust emissions measurements were conducted using two fast response analyzers from Cambustion Ltd. The Cambustion CLD500 analyzer was employed 230 for the cycle resolved NO emissions. From the two available sampling heads of this analyzer, 231 one was used and installed directly downstream of the exhaust valve of the cylinder, while the 232 second probe was installed downstream enough to the exhaust gases to get an average value of 233 234 NO emissions. The sampling response time of NO emissions was in the order of 2 ms (Table 3). Simultaneously, Cambustion HFR400 analyser was employed for HC measurement. The 235 only used sampling head was installed downstream of the exhaust valves and its sampling time 236 was about 4 ms. Signals from both heads (3 in total) were directed to the NI data acquisition 237 card. 238



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Figure 4: Schematic of experimental setup for combustion imaging

Images of the visible light from combustion heat were captured through an overhead window 241 as shown in Figure 4. The camera was capable of imaging at 6000 frames per second (fps) with 242 a resolution of 512 x 384 pixels. All cycles presented in this paper were imaged at 6000fps 243 with an exposure time of 167µs. The internal memory of the camera allowed for a total of 244 10,000 frames to be recorded before the data were stored in a hard drive (laptop). At 1200rpm 245 246 this allowed for 16 cycles to be recorded in a single test. Due to the large variation in intensity of the light emission from combustion between cycles, the gain was adjusted for each cycle to 247 improve the clarity of the images. 248

The reason that in this study an optical engine has been employed is to qualitatively explain
the cyclic resolved emission values under various knocking conditions by utilizing optical data.
Realistic results from commercial engines may quantitatively change, as explained elsewhere
[44].

253 2.3 Engine Test Protocol

254 A fixed reference point was obtained by running the engine under normal non-knocking partload operation until the cylinder head metal temperature reached 88°C. At this temperature 255 knock was induced by resetting the spark timing. After capturing the data, the engine was 256 stopped and allowed to cool down before the process repeated. This process kept the measured 257 wall temperatures within a small range (<5°C peak-to-peak variation), which was considered 258 necessary when considering knock in a qualitative manner with an engine setup lacking water 259 cooling in the head. The thermodynamic results were averaged over three sets of 100 cycles 260 261 for each test condition. The sump oil temperature remained moderately low throughout testing $(\sim 40^{\circ} \text{C})$. Table 4 shows the engine test conditions. 262

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Ta	ble	<u>4</u>	:	Engine	0	perating	conditions.
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Parameter (unit)	Value
Engine Speed (RPM)	1200±5
Relative AFR (λ)	1±0.01
Inlet Pressure (bar)	0.9±0.02
Inlet Air Temperature (°C)	66±2
Exhaust Bridge Temperature (°C)	130±2
Head Temperature (°C)	88±1

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265 **2.4 Data Processing**

An example of processing the in-cylinder pressure of two consecutive cycles with KI index 0.76 bar (light knock cycle) and 15.1 bar (super knock cycle) respectively versus NO concentration is given with reference to **Figure 5**. During the valve-closed part of the current cycle, the analyzer measures the NO level of the gas remained in the exhaust manifold from the previous cycle. When exhaust valve opens (EVO) a portion of the (in-cylinder) gas violently exits the cylinder (blow-down) and clears the exhaust manifold from the previous

cycle gas. However, a delay between EVO and analyzer response (point A in Figure 5) is 272 273 observed, attributed to the distance between the valve outlet and the sampling probe, as well as to the instrument response time [23]. After the exhaust valve closes (EVC), the analyzer signal 274 exhibits a slight fluctuation (area B-C in Figure 5), indicating a variation of NO concentration 275 in the cylinder gas. The NO concentration may vary from cycle to cycle due to the combustion 276 process. During the closed part of the next cycle, the analyzer signal can be considered constant 277 278 (part C-D in Figure 5) and corresponds to the NO level of the current cycle. This average value is taken as the NO emissions amplitude of the current cycle. Last but not least, Figure 5 also 279 shows that the level of NO (C-D area) after a light knock cycle and after a super knock cycle 280 281 (C' point) can significantly vary. The deeper understanding of this observation is within the aim of this study. A similar procedure was followed to detect HC concentration at each 282 283 combustion cycle, taking into account the variations in signal delay, due to the different 284 positioning of the sampling probes for NO and HC species. More details on the exact process per pollutant can be found in open literature [21], [23], [48], [49]. 285



Figure 5: Schematic illustration of the fast NO signal in reference to in-cylinder pressure for two consecutive
cycles with KI=0.76 bar and 15.1 bar respectively.

289 **3 Results and Discussion**

290 3.1 Effect of Spark Ignition Timing

The phenomenon of knock is controllable by the spark advance as advancing or retarding the 291 spark timing can increase or decrease the knock severity or intensity respectively [47]. 292 293 Although knock varies substantially from cycle to cycle and doesn't occur at each combustion cycle, an average combustion cycle can present the 'mean' picture of knock intensity. Various 294 spark timings lead to different heat release rate histories, which can be linked to end-gas 295 pressure and temperature conditions that can cause autoignition. The latter can be observed as 296 297 a rapid increase in the pressure trace accompanied by fluctuations which amplitude varies with time. 298



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Figure 6: In-cylinder pressure traces for six (6) different ignition timings. Advanced ignition leads to heavy
 knock.

Figure 6 shows six (6) different combustion cycles where the range of knock intensity varies. The illustrated pressure traces represent the average imep cycle of the measured 300 cycles at each spark timing condition. It is observed that as ignition timing shifts from TDC to 25° BTDC, the magnitude of peak combustion pressure and the amplitude of pressure fluctuations are impressively increased. In fact, the peak in-cylinder pressure when ignition timing occurs at TDC compared to the ignition timing occurs at 25° BTDC is more than three (3) times higher. Furthermore, it is revealed that knock intensity increases exponentially as knock occurs closer to top center, earlier in the combustion process.



Figure 7: Impact of spark ignition timing on IMEP and KI under stoichiometric conditions and wide open
 throttle conditions.



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Figure 8: Impact of spark timing on NO and HC emissions, considering mean value.

315 Pressure traces substantially vary from cycle-to-cycle; therefore mean values of imep and KI as well as the variability of these indexes can explain the transition from normal combustion to 316 heavy knock by advancing spark timing. Figure 7 schematically presents the relationship 317 between both KI and imep against spark timing. MBT is achieved at spark timing close to 10° 318 BTDC, where the mean value of imep is maximized and the COV of imep is minimized. Further 319 320 spark advance leads to higher imep variability, the mean value of imep drops and KI is exponentially increased. At 15° BTDC spark timing, KI is equal to 2 bar and can be classified 321 322 into a medium knock case while further spark advance leads to heavy knock cycles.



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Figure 9: Relationship between maximum cylinder pressure and NO formed emissions under normal
combustion conditions (spark timing at 5° BTDC).

Cycle to cycle variability is recognized as the main reason of cyclic emission variability; 326 however the impact of knocking combustion on pollutants formation has not been previous 327 investigated extensively. The averaged and variation range of the emission values for 300 328 combustion cycles as well as the relationship between NO and HC emissions against spark 329 330 timing are illustrated in Figure 8. HC are increased against spark timing due to incomplete combustion as KI is increased. However, under heavy knock conditions mean HC are slightly 331 decreased as pre-ignition appears and combustion characteristics change. Regarding NO, it was 332 found that engine-out NO are enhanced versus spark timing which is continued until heavy 333 knock conditions at spark timing 20° BTDC and then average formed NO is decreased. 334 Previous studies on cyclic NO variability presented that MBT is related with the maximum 335 mean NO value and the minimum COV_{NO} value [43], while the effect of knocking combustion 336

in this relationship is not usually considered. The current tested experimental case clearly presents that knock can further increase mean NO exhaust emissions by up to 36%, while considering NO variability, instantaneously this increment can reach up to 60%. A further analysis to better understand the deeper reason of this trend is needed and followed in this study.



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Figure 10: Relationship between maximum cylinder pressure and NO formed emissions under heavy knock
 conditions (spark timing at 25° BTDC).

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In addition to NO emissions, **Figure 8** presents the relationship between HC emissions and spark timing. HC emissions mean value is increased as spark ignition timing is advanced apart from the TDC ignition timing point, as this point seems to represent a very late combustion, very slow heat release rate that declares incomplete combustion takes place. In general, HC emissions present similar trend to NO emissions against spark timing. Under knocking conditions (spark timing exceeds 10° BTDC), HC emissions are enhanced by up to 10% while under heavy knock conditions a slight drop compared to the peak HC emissions is observed.







Nitric oxide is mainly formed at high temperatures, through the well-known extended 356 Zeldovich thermal mechanism [26]. Peak combustion temperature and residence time at high 357 358 temperature conditions are the two primary reasons on nitrogen oxidation. Figure 9 represents the relationship between maximum cylinder pressure and NO emissions for 100 consecutive 359 cycles under normal combustion conditions (ignition timing 5° BTDC). The trend of these 360 points is found close to linear, validating similar observations from previous studies [21], [43], 361 [50]. In fact, deviations from this linear correlation can be related with the cyclic dispersion of 362 mixture parameters such as residual gas fraction, fuel trapped mass or air-fuel ratio at each 363 combustion cycle. This linear trend clearly shows that as maximum cylinder pressure is 364 increased NO formation is enhanced. However a similar analysis of the data under heavy knock 365 conditions (ignition timing 25° BTDC) showed the opposite linear trend; higher maximum 366 cylinder pressure leads to lower NO emissions (Figure 10). Furthermore, the dispersion of the 367 points is at much higher range compared to the case of normal combustion conditions. The 368 latter is resulted from the stochastic behaviour of knocking combustion conversely to the more 369 robust normal combustion. 370





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Figure 12: Comparison of various KI conditions.

NO variability is highly related to the combustion rate, as approved elsewhere [38]. **Figure 11** illustrates the relationship between maximum cylinder pressure and NO emissions for various spark timing conditions. It is observed that the area where points occur is restricted by a 'left' and 'right' limit that both form a triangular region. The third axis of this plot represents the knock intensity which is illustrated as contour lines on the same figure. The 'left' limit of this plot is well-matched with the KI equal to zero contour line. In fact, it describes the observed

trend found at Figure 9; higher maximum cylinder pressure leads to higher NO emissions under 379 380 normal SI combustion conditions and it is the limit of NO formation under deflagration conditions. Moving to the right part of Figure 11, maximum cylinder pressure increases as end-381 gas auto-ignition starts to occur. Under knocking combustion condition, it is observed that the 382 variation between maximum and minimum NO emissions at higher maximum cylinder 383 pressure is decreased and reaches a narrow window area at extreme peak in-cylinder pressures. 384 The latter forms the right region limit which is hard to explain from recorded thermodynamic 385 data. Therefore natural light photography data were employed to better understand this trend. 386

387 Set out in Figure 12 are flame images from three (3) different combustion cycles under various 388 knock intensity conditions; namely light knock, medium knock and heavy knock. It can be seen 389 that for KI=2, auto-ignition is observed at the late phase of the combustion process. Burned 390 zone occurs for long time, enough for the post-flame kinetics to form a substantial amount of NO emissions. On the opposite site, at heavy knock cycles, auto-ignition occurs much earlier 391 392 and respectively burned zone also occurs less, NO residence time is decreased and although peak temperature is higher final formed NO emissions are significantly lower to normal 393 394 combustion case. Summarizing, optical data present that higher knock intensity can be correlated with higher in-cylinder pressure while pressure waves affect the residence time in 395 the post flame region. 396

397 The observation of Figure 12 explains the 'right' limit region on Figure 11. NO formation depends on oxygen availability, peak temperature and the residence time of the mixture at high 398 temperature conditions. In stoichiometric mixtures and under knocking conditions there is a 399 competition between maximum cylinder temperature and residence time of combustion 400 products in the burned region. As KI is increased, knock occurs earlier during the combustion 401 process which means that residence time can potentially decrease although peak temperature 402 and pressure are significantly enhanced. The latter affects the kinetic rate of NO formation and 403 404 lead to lower emissions. In this study was found that combustion cycles with KI higher than 6 405 present a drop on NO emissions.

406 **3.2 Effect of Air/Fuel Equivalence Ratio**

407 Combustion evolution and knock onset are affected by mixture composition and oxygen
408 availability, while fluctuations of air/fuel ratio is an important source of CCV [51]. Pollutants
409 formation is also expected to be affected by the nature of the combustion and the oxygen
410 availability.

The effect of oxygen availability on knock onset and pressure evolution is illustrated on Figure 411 412 13 that shows the in-cylinder pressure traces for three different mixtures: slightly rich, slightly lean and (almost) stoichiometric. The three (3) cycles correspond to identical ignition timing 413 (20° BTDC), engine load (imep~4.4 bar) but different knock intensity conditions. It is observed 414 that slightly rich mixture presents a faster burn rate and reaches end-gas auto-ignition limit 415 closer to TDC, as due to the smaller cylinder volume of the unburned region, the unburned 416 temperature and pressure are relatively high at this crank angle. On the other hand, slightly lean 417 mixture is characterized by a slower burn rate and compared to the stoichiometric combustion 418 cycle, the onset of knock at lean conditions appears with 5° CA delay. 419



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Figure 13: In-cylinder pressure traces for three (3) different air/fuel equivalence ratios and similar ignition
 timings (TDC-20°) and imep. Equivalence ratio significantly affects knock onset and peak in-cylinder pressure.

The knock resistance of lean mixtures has been investigated in the past by Gruden and Hahn [52]. In their study, a production engine was converted to a lean burn engine. Researchers explored the effect of many parameters on engine economy including: high compression ratio, optimization of ignition timing and layout of the combustion chamber shape with intensive charge turbulence. They found that compression ratio can be increased with lean operation, implying that lean mixtures are more knock resistant. However, the operating conditions under which this trend is valid is not clearly stated. Considering the results of **Figure 13**, a possible reason could be the slower burn rate that leads to auto-ignition when piston expands andconsequently end gas temperature and pressure are relatively low.

In order to better understand the effect of air/fuel equivalence ratio on knock intensity and NO 432 433 emissions, the 20° BTDC ignition point was selected from stoichiometric data previously presented and experiments were performed for various air/fuel equivalence ratios. Figure 14 434 435 shows that maximum KI is observed at close to stoichiometric conditions, while under slightly 436 lean conditions KI drops impressively. Conversely to previous studies where it was found that lean mixtures increase the COV of imep and cyclic combustion variability is much higher, 437 knocking combustion lean mixtures reduce cyclic variability as KI drops. The effect of air/fuel 438 equivalence ratio on NO formation under knocking combustion is also explored. It was found 439 that maximum NO emissions are enhanced under lean conditions due to the excess of oxygen 440 and the peak temperature conditions that occur under slightly lean conditions. Compared to 441 normal combustion lean operating (λ ~1.1) conditions, it was found that NO can be up to 3 442 times higher under autoignition conditions. 443



444

445 **Figure 14:** Effect of air/fuel ratio (λ) on average knock intensity and NO emissions for similar ignition 446 timings (TDC-20) and engine load (imep~4.4 bar).

Cyclic combustion variability consists of both stochastic and deterministic phenomena [22].
Fluctuations of the gas mixture motion and turbulence and variation of the quantity and spatial
distribution of the fuel are related with the stochastic aspect of cyclic dispersion. Leaner and
more dilute mixture conditions are related to the deterministic aspect of cyclic variability [53]

- 451 and can also affect the amplitude of knock intensity under knocking conditions. The latter is
- 452 further discussed in this study.





454 **Figure 15:** KI return map for ignition timing TDC-20 and stoichiometric air/fuel ratio conditions.

The return map is identified as an easy and fast way to characterize the nature of the variability of any index and it includes the plot of the current value against the previous or next cycle value of the same index. A boomerang-shaped pattern declares the deterministic aspect of variability while a noisy spot point shows the stochastic feature of the variability.



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Figure 16: KI return map for ignition timing TDC-20 and slightly rich air/fuel ratio conditions.

461 Figure 15 and Figure 16 show the KI return map for stoichiometric and rich operating
462 conditions respectively. It is observed that under stoichiometric conditions KI presents a

boomerang-shaped pattern (Figure 15), while the scatter of data under rich conditions has been 463 464 decreased in an almost noisy spot (Figure 16). This strong indication proves the deterministic character of knock intensity can significantly be reduced under rich conditions. The reason can 465 be the well-known cooling effect of rich mixtures, that decreases the end-gas temperature and 466 pressure and finally the knock intensity is reduced. It has to be noted that the small impact of 467 mixture air/fuel equivalence ratio on average engine load (imep), which differs by 2% between 468 the stoichiometric and rich mixtures, slightly affects the residual gas fraction, which is a strong 469 deterministic combustion parameter. 470



471

472 Figure 17: Relationship between maximum cylinder pressure and NO formation for various KI conditions
473 (contour lines) as a result of various ignition timing conditions under slightly lean conditions (λ~1.1).

474

Nitric oxide formation is related with peak combustion temperature, mixture residence time at
high temperature and oxygen availability [43]. A recent study proved that under normal
combustion conditions, as a result of cyclic emission variability, fast burn rate cycles present
peak formed NO emissions under slightly lean conditions while slow burn rate cycles present
peak NO emissions closer to equivalent air/fuel ratio at unity [21].





481 **Figure 18:** Relationship between maximum cylinder pressure and NO formation for various KI conditions 482 (contour lines) as a result of various ignition timing conditions under slightly rich conditions (λ ~0.9).

483 Under knocking combustion conditions, where peak temperatures are observed, oxygen availability seems to be the most important parameter for NO formation. Figure 17 illustrates 484 the relationship between maximum in-cylinder pressure and NO formation for slight lean 485 486 mixtures and various KI conditions, which are presented as contour lines in the plot. Similar to stoichiometric conditions presented before, the scatter area of the data form a triangular. The 487 488 left limit matches with the KI=0 isoline; under normal combustion conditions NO is 489 proportional to maximum in-cylinder pressure. Compared to stoichiometric conditions, the rate 490 is much faster due to the excess of oxygen.

491 Although the 'left' limit at **Figure 17** is similar to the trend found at **Figure 11**, the 'right' limit line is significantly different to stoichiometric conditions. The only parameter that alters 492 between those figures is oxygen availability. The competitive relationship between peak 493 temperature and residence time that was found under stoichiometric conditions is not appeared 494 under slightly lean conditions. The excess oxygen enhances post flame NO kinetics which are 495 496 further benefit from higher peak temperatures in high knock intensity combustion cycles. The general trend shows that under high KI values, NO emissions are clearly increased with the 497 498 scatter of the data being also decreased, which means that the average value of NO follows an 499 important increment.

500 On the opposite site, **Figure 18** presents the relationship between maximum in-cylinder 501 pressure and NO formation for slight rich mixtures and various KI conditions. The trends are 502 identical to the previous discussed under stoichiometric conditions at **Figure 11**, presenting the 503 competition between residence time and peak temperature for various KI conditions.

504 **4** Conclusions

505 This study explores pollutants formation under knocking combustion conditions using an 506 optical single cylinder SI engine. A novel experimental setup has been utilized including fast 507 response analyzers for NO and HC emissions, natural light photography and in-cylinder 508 pressure recordings. The effect of two engine operating parameters where explored such as 509 spark ignition timing and air/fuel equivalence ratio.

The effect of ignition timing on pollutants formation under various knock index conditions has 510 been explored. It was found that as spark timing is advanced and knock intensity is increased, 511 both NO and HC emissions are enhanced. Up to knock intensity equal to 6 bar and under 512 stoichiometric conditions, it was found that instantaneous NO emissions are increased by up to 513 60% compared to the MBT operating conditions. A further increase of knock intensity at the 514 limits of pre-ignition region was found to significantly drop NO emissions. Simultaneously HC 515 emissions were found to increase at the region of light and medium knock where the peak of 516 HC emissions was observed, while at heavier knock region HC emissions drop. 517

The effect of air/fuel equivalence ratio on pollutants formation under knocking combustion has been also investigated in this study. It was found that leaner mixtures present the onset of knock later compared to richer mixtures due to the slower deflagration rate. When knock occurs, the formed NO in lean mixtures are higher than in stoichiometric or rich mixtures due to the oxygen availability. Last but not least, it was found that under lean conditions NO emissions are enhanced as knock intensity is increased while under stoichiometric or slightly rich operating conditions it was found that NO are decreased for knock intensity higher than 6 bar.

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