

Greenhouse Gas and Noxious Emissions from Dual Fuel Diesel and Natural Gas
 Heavy Goods Vehicles

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13 Abstract

- 14 Dual fuel diesel and natural gas heavy goods vehicles (HGVs) operate on a combination of
- 15 the two fuels simultaneously. By substituting diesel for natural gas, vehicle operators can
- 16 benefit from reduced fuel costs and as natural gas has a lower CO₂ intensity compared to
- 17 diesel, dual fuel HGVs have the potential to reduce greenhouse gas (GHG) emissions from
- 18 the freight sector. In this study, energy consumption, greenhouse gas and noxious emissions
- 19 for five after-market dual fuel configurations of two vehicle platforms are compared relative
- 20 to their diesel-only baseline values over transient and steady state testing. Over a transient
- 21 cycle, CO₂ emissions are reduced by up to 9% however methane (CH₄) emissions due to
- 22 incomplete combustion lead to CO₂e emissions that are 50-127% higher than the equivalent
- 23 diesel vehicle. Oxidation catalysts evaluated on the vehicles at steady state reduced CH₄
- 24 emissions by at most 15% at exhaust gas temperatures representative of transient conditions.
- 25 This study highlights that control of CH₄ emissions and improved control of in-cylinder CH₄
- 26 combustion are required to reduce total GHG emissions of dual fuel HGVs relative to diesel
- 27 vehicles.

28 **1** Introduction

29 1.1 Context

Globally, road freight transport is responsible for around a quarter of transport energy use¹. In 30 31 the UK, heavy goods vehicles (HGVs) involved in freight movements account for 22% and 23% of road transport energy use² and greenhouse gas (GHG) emissions³ respectively. Long 32 haul and regional duty cycles account for approximately 70% of UK HGV CO₂ emissions 33 34 and the substitution of natural gas (NG) for diesel has the potential to reduce HGV CO₂ emissions due to the lower CO₂ intensity of methane compared to diesel⁴. In 2012, the UK 35 36 Technology Strategy Board and the Office for Low Emission Vehicles provided £11.3m 37 (~\$17.5m) to trials of 362 diesel-NG dual fuel trucks, using five different dual fuel systems. 38 Four out of five of these systems are aftermarket conversions, while one is supplied by an 39 original equipment manufacturer (OEM)⁵. In 2013, the European Commission provided €14.3m (~\$15.6m) to the trial of 100 LNG HGVs, including dual fuel vehicles⁶. In the US, 40 41 the Environmental Protection Agency has certified one diesel-NG conversion system for new 42 engines, which grants exemption from the Clean Air Act tampering prohibition, and a 43 number of other dual fuel systems for intermediate age engines⁷.

The use of NG as a transport fuel is at least partially motivated by the potential to reduce CO_2 emissions. The primary component of NG is methane (CH₄). As CH₄ has a higher ratio of hydrogen to carbon atoms (4:1) than diesel (~2:1), less CO₂ is emitted per unit of chemical

47 energy released by combustion. A dual fuel engine is a conventional compression ignition

48 diesel engine in which a significant proportion of the energy released by combustion is

derived from the combustion of a gaseous fuel, such as NG^8 . All of the dual fuel systems

installed on vehicles residing the UK operate by injecting NG into the intake air prior to the
 intake valves so that a NG-air mixture is drawn into the combustion chamber^{5,9–12}. This NG-

52 air charge is then ignited by an injection of diesel at the end of the compression stroke, which

has a shorter ignition delay than the NG^8 . This dual fuel concept differs from systems that

54 employ high-pressure direct injection (HPDI) of NG into the combustion chamber¹³.

55 Dual fuel engines offer an attractive alternative to other engine technologies due to the (i)

56 higher thermal efficiency relative to spark-ignited engines, (ii) flexible fuel capability with

57 the option to operate solely on diesel when gaseous fuel is not available, (iii) reduced fuel

costs, and (iv) reductions in emissions of CO₂ and other engine exhaust components^{4,14}.

59 Previous studies of dual fuel engine emissions have focussed on engine dynamometer testing,

60 rather than full vehicle emissions testing. These studies have shown significant changes to

61 emissions when compared to conventional diesel engines^{8,14-21}. Dual fuel engine emissions

62 depend on a number of parameters including the engine speed, engine load, the composition

- and quantity of primary gaseous fuel, the quantity of pilot diesel, the temperature of the
- 64 intake primary fuel and air mixture, the pilot injection timing and stratification of the gaseous
- fuel and air mixture $^{16,21-23}$. In previous studies, dual fuel combustion with NG as the primary
- fuel led to reduced emissions of oxides of nitrogen (NO_x) but increased emissions of carbon
- 67 monoxide (CO) and unburnt hydrocarbons $(HC)^{14-21}$. The majority of the unburnt
- 68 hydrocarbon emissions from a NG dual fuel engines is CH₄ and the magnitude of these
- 69 emissions depended strongly on the concentration of the gaseous fuel in the cylinder^{14,18}.
- 70 Using a global warming potential (GWP) of 25 for CH₄, Besch et al.¹⁸ showed that while CO₂
- 71 emissions were reduced by 3-8% for three dual fuel engines relative to diesel operation over
- the heavy duty engine Federal Test Procedure, emissions of CH₄ led to 18-129% increases in
- 73 CO₂e emissions.

74 Previous smoke opacity measurements have shown that diesel and NG dual fuel combustion

- has the potential to reduce smoke emissions^{19,20,24}. Graves et al.²⁵ characterised the
- morphology and volatility of particles emitted by an HPDI dual fuel engine, however this is a
- distinctly different dual fuel concept from that employed by the systems currently in use andevaluated in this study.

79 1.2 Overview

80 This paper presents an evaluation of transient and steady state energy use, greenhouse gas, 81 nitrous oxides, carbon monoxide and particulate matter (PM) emissions of five vehicle configurations of two dual fuel aftermarket conversion systems that account for the majority 82 (~60%) of the 362 trialled dual fuel vehicles in the UK⁵. In contrast to previous studies that 83 have tested engines installed on engine dynamometers²¹, this study tests in-use vehicles with 84 85 emissions control devices on a chassis dynamometer to quantify dual fuel HGV emissions 86 factors for subsequent use in environmental impact assessments and provide emissions 87 metrics that are relevant to policy makers and logistics fleet operators. Furthermore, this 88 paper highlights areas for technology development. Due to commercial sensitivities, the 89 vehicle owners and the dual fuel conversion suppliers have not been disclosed.

90 2 Materials and methods

91 2.1 Outline of study

92 The vehicle configurations and specifications of the original vehicle platforms are

- 93 summarised in Table 1. This study's authors did not modify or attempt to control the
- 94 performance of the dual fuel systems or fuelling strategies. On both vehicles where methane

95 oxidation catalysts are present, these were installed upstream of the OEM selective catalytic

- 96 reduction (SCR) unit by the dual fuel conversion suppliers. The dosing of aqueous urea
- 97 solution in the SCR systems on both vehicles was not altered and neither vehicle was fitted
- 98 with a diesel particulate filter. Configurations A0 (4×2 tractor) and B0 (6×2 tractor) are taken
- as baseline tests, while A1 and B1 are the most common diesel-NG dual fuel vehicle
- 100 configurations for suppliers A and B respectively. The configuration in A2 and A4 is unique
- 101 for this study and is not a standard option offered by supplier A; it was tested to evaluate the
- 102 impact of an oxidation catalyst on performance and emissions. This oxidation catalyst was a
- 103 prototype design that was installed immediately before these emissions tests were conducted.
- 104 In contrast, the oxidation catalyst on vehicle B had been installed on the vehicle for
- 105 approximately 500,000 km and is therefore is representative of catalysts in operation.
- 106 Transient cycle and steady state vehicle emissions testing was conducted on a 1.22 m (42 in.)
- 107 single roll chassis dynamometer capable of simulating 20 tonnes of inertia (Millbrook
- 108 Proving Ground Ltd, Bedford, MK45 2JQ, UK). Further experimental details including fuel
- 109 properties and uncertainty analyses are shown in the Supporting Information.

Ref.	After-treatment	Fuel	Hot/cold start
A0	SCR	Diesel	Hot
A1	SCR	Dual fuel	Hot
A2	SCR & Oxi cat	Dual fuel	Hot
A3	SCR	Dual fuel	Cold
A4	SCR & Oxi cat	Dual fuel	Cold
B0	SCR & Oxi cat	Diesel	Hot
B1	SCR & Oxi cat	Dual fuel	Hot
B2	SCR & Oxi cat	Dual fuel	Cold
	Ref. A0 A1 A2 A3 A4 B0 B1 B2	Ref.After-treatmentA0SCRA1SCRA2SCR & Oxi catA3SCRA4SCR & Oxi catB0SCR & Oxi catB1SCR & Oxi catB2SCR & Oxi cat	Ref.After-treatmentFuelA0SCRDieselA1SCRDual fuelA2SCR & Oxi catDual fuelA3SCRDual fuelA4SCR & Oxi catDual fuelB0SCR & Oxi catDieselB1SCR & Oxi catDual fuelB2SCR & Oxi catDual fuel

110 **Table 1: Test vehicle specifications and list of transient test parameters.**

111 2.2 Test cycles

Transient cycle emissions were obtained over the vehicle version of the European Transient
Cycle, developed by the FIGE Institute²⁶. The FIGE cycle has three distinct phases,

114 representing urban (U), rural (R) and motorway (M) driving. Hot start tests followed a

115 consistent engine warm up procedure, while cold start tests were conducted at 0°C after the

116 vehicle had been soaked overnight. Steady state emissions testing on the chassis

117 dynamometer was conducted to develop a map of engine emissions as a function of engine

118 torque and speed (revolutions per minute, rpm) that would also be of further use in vehicle

119 emissions models²⁷. Engine speed and torque were systematically varied in ~200 rpm and

120 ~200 Nm steps respectively. Each test point was held for at least two minutes, consistent with

121 regulatory engine test cycles^{28,29}.

122 2.3 Instrumentation

123 The net flow rate of diesel supplied to the engines was measured using an FMS MK10 fuel

124 flowmeter (JPS Engineering, UK), which recorded flow in 5 mL increments. The mass flow

125 rate of NG delivered to the engines was measured by a Rotamass RCCS34 Coriolis flow

126 meter (Yokogawa Electric Corporation, Japan) placed in line between the low pressure

127 regulator and gas injectors of the dual fuel systems. This Coriolis flow meter was sized to

128 minimise interference with dual fuel system by minimising pressure drop while maintaining

- accuracy; at a nominal flow rate of 22.5 kg/hour at 5 bar and 20°C, the pressure drop and
- 130 accuracy were calculated to be 17 mbar and 1.1% respectively. The energy substitution ratio
- 131 (ESR) is defined as the proportion of total energy supplied to the engine in the form of NG,

$$\text{ESR}\left[\%\right] = \frac{\dot{m}_{\text{NG,in}} \text{LCV}_{\text{NG}}}{\dot{m}_{\text{NG,in}} \text{LCV}_{\text{NG}} + \dot{m}_{\text{diesel,in}} \text{LCV}_{\text{diesel}}} \times 100 \,, \tag{1}$$

132 where $\dot{m}_{\text{NG,in}}$ and $\dot{m}_{\text{diesel,in}}$ are the mass flow rates of NG and diesel supplied to the engine 133 respectively, and LCV is the lower calorific value of the fuel.

134 Engine emissions and exhaust gas temperatures were measured at two locations in the

135 exhaust; (i) post-turbo (PT), equivalent to an engine-out measurement, and (ii) at the tailpipe.

136 A complete list of the emissions analysers and emissions species is included in the SI.

137 Reported emissions factors have been calculated based on the modal 1 Hz data for transient

and steady state testing. For CO_2 , NO_x and CO, these emissions factors were within 1%, 1%

and 7% of emissions factors calculated based on averaged bag measurements over the entire

140 transient test cycle.

141 CH₄ emissions were measured at the post-turbo and tailpipe locations using Fourier

142 Transform Infra-Red (FTIR) spectrometers (Multigas 2030 and 2030 HS respectively, MKS

143 Instruments, MA, USA). CH₄ slip is reported as the ratio of the mass flow rate of exhausted

144 CH₄, $\dot{m}_{CH_4, tailpipe}$, to the mass flow rate of CH₄ supplied to the engine in the NG, $\dot{m}_{CH_4, in}$,

$$CH_4 \operatorname{slip} [\%] = \frac{\dot{m}_{CH_4, \text{tailpipe}}}{\dot{m}_{CH_4, \text{in}}} \times 100\% \,.$$

(2)

145 The effectiveness of oxidation catalysts in the vehicles' exhaust after-treatment systems was 146 evaluated by comparing CH₄ concentrations at the post-turbo and tailpipe sampling points 147 measured by the two FTIR instruments.

148 A DMS500 (Cambustion Ltd, UK) measured real-time particle size distributions at the

tailpipe. PM and black carbon (BC) mass emissions were estimated using the integrated

150 particle size distribution (IPSD) method^{30,31}. The aggregate effective density, ρ_{eff} , as a

151 function of mobility diameter, d_{p} , was calculated as

$$\rho_{\rm eff} = \rho_0 \left(\frac{d_{\rm p}}{d_{\rm pp}}\right)^{D_{\rm m}},\tag{3}$$

152 where ρ_0 is the material density, d_{pp} is the primary particle diameter and D_m is the mass-

153 mobility exponent with assumed values of 2 g/cm³, 16 nm and 2.35 respectively³². For $d_{\rm p}$

154 less than 30 nm, an effective density of 1.46 g/cm³ is assumed³². Total PM mass was

155 calculated by integrating over the entire size distribution (5-1000 nm), while for BC, a

- 156 lognormal distribution was fitted to the accumulation mode. Reported particle number (PN)
- 157 emissions factors are calculated by summing the product of the particle size distribution and a
- 158 detection efficiency curve³³ fitted to data for condensation particle counters with detection
- 159 efficiencies of 50% at 23 nm³⁴. This enables comparison to measurements following the solid
- 160 particle number measurement protocol developed through the Particle Measurement
- 161 Programme (PMP)³⁵.
- 162 Total GHG emissions, as CO₂e were calculated by summing CO₂, CH₄, N₂O, NO_x, CO and
- 163 BC emissions multiplied by their GWPs for a time horizon of 100 years. GWPs for CH₄ (34)
- and N₂O (298) are taken from the IPCC's Fifth Assessment Report (AR5) and include
- 165 climate-carbon feedbacks ³⁶. All GWPs are included in the SI and we note that choosing a
- 166 shorter time horizon (e.g. 20 years) would increase the importance of short-lived species (e.g.
- 167 CH₄).

168 **3 Results**

169 3.1 Transient cycle energy consumption and emissions

170 Distance specific energy consumption and emissions factors over different phases of the 171 FIGE cycle (U = urban, R = rural, M = motorway, C = combined) for the different vehicle 172 configurations are shown in Table 2 and Table 3. This data is shown graphically in the SI. 173 Distance specific energy consumption is generally highest during the urban phase, followed 174 by the motorway and rural phases for all vehicle configurations. In general, dual fuel 175 operation reduces energy efficiency and results in higher total energy consumption compared 176 to the baseline diesel only tests (A0 and B0). For A1 and B1, combined energy consumption 177 is 12% and 10% higher than the equivalent diesel tests. Previous studies have also shown that 178 energy efficiency of dual fuel engines are reduced compared to diesel at low and intermediate 179 engine loads, which is attributed to lower combustion efficiency of the lean NG-air charge mixture and higher rates of heat loss during combustion²¹. Over the four dual fuel tests with 180 181 vehicle A, higher ESR during A1 and A3 compared to A2 and A4, indicate that the dual fuel 182 system supplier may have updated their control software. Comparing A1 to A2, higher ESR 183 is correlated with higher energy consumption yet lower CO₂ emissions. For vehicles A and B, 184 the highest ESR is observed during the motorway phase and during this phase, the CO₂ 185 emissions factor is lowest and the relative reduction compared to diesel is greatest at up to 186 11% and 13% for A1 and B1 respectively. For context, the fuel properties (shown in the SI)

187 indicate that complete combustion of NG would emit approximately 21% less CO₂ per unit of

188 energy than diesel used in this study, assuming no change in energy efficiency. For the cold

189 start tests, (A3, A4 and B2), distance specific energy consumption and emissions of CO₂ are

190 generally greater than the equivalent hot start test. Both dual fuel conversion suppliers

- 191 account for engine temperature in their control of the ESR; during the urban phase, when
- 192 engine temperatures are cold, the ESR is significantly reduced relative to the same phase in
- 193 hot start tests.

Table 2: Energy consumption, energy substitution ratio, CO₂ and CO₂e emissions
 factors from transient testing. U = urban, R = rural, M = motorway, C = combined
 phases of the FIGE cycle.

	E	nergy (MJ/km	ı)		ESR (%)				CO ₂ (g/km)					CO ₂ e (g/km)				
Ref.	U	R	М	С	U	R	М	С	U	R	М	С	τ	ſ	R	М	С		
A0	12.8	10.2	10.5	10.7	0	0	0	0	957	753	747	777	9	54	751	740	772		
A1	13.7	11.1	12.3	12.0	37.6	45.9	58.3	50.9	923	686	662	705	12	64	931	1055	1036		
A2	12.5	10.4	11.6	11.3	31.0	37.1	51.4	43.4	920	708	663	714	13	44	1013	1239	1168		
A3	16.7	11.5	12.3	12.6	16.5	46.8	59.3	49.1	1175	711	676	753	13	05	952	1083	1062		
A4	13.1	12.1	12.6	12.5	5.4	37.3	48.7	40.0	1184	724	673	744	12	69	1089	1352	1242		
B0	14.6	10.1	11.0	11.2	0	0	0	0	1135	754	742	798	11	29	758	748	801		
B1	20.4	9.9	11.9	12.2	32.7	53.6	73.5	60.7	1156	692	649	730	25	22	1408	1945	1817		
B2	15.7	13.3	14.3	14.1	0.9	54.0	71.1	57.4	1443	703	650	751	13	80	1550	2418	1977		

197 CO₂ and CH₄ (shown in Table 3) dominated total CO₂e emissions and capture the first-order 198 climate impacts of dual fuel relative to diesel. The combined cycle relative contribution of 199 CH₄ normalised to the CO₂ contribution was between 40% for A3 and 150% for B2. CH₄ emissions factors are typically highest during the motorway phase, which is also the phase 200 201 with highest ESR. For all dual fuel tests on both vehicles, CH₄ emissions led to increased 202 CO₂e emissions relative to the diesel baseline tests. Comparing A1 to A0 and B1 to B0, dual 203 fuel operation increased CO₂e emissions factors by 32%, 24%, 42% and 123%, 86%, 160% 204 for the urban, rural and motorway phases respectively. Therefore, it is clear that CH₄ 205 emissions outweigh potential reductions in CO₂ emissions that result from substituting diesel 206 for NG. For A1, 8.4%, 6.5% and 8.1%, and for B1, 30.1%, 18.4% and 21.0% of CH₄ that is 207 delivered to the engine is emitted to the atmosphere during the urban, rural and motorway 208 phases respectively. These findings are in agreement with previous engine dynamometer 209 testing of dual fuel retrofit systems, which also measured significant increases in CO₂e emissions due to CH₄ emissions¹⁸. The addition of an oxidation catalyst to A2 compared to 210 211 A1 did not significantly reduce CH₄ emissions; indeed distance specific CH₄ emissions are

- 212 increased, which could be indicative of an altered fuelling strategy or that backpressure due
- to the oxidation catalyst may have had an adverse effect on in-cylinder combustion and
- 214 therefore engine emissions performance. Oxidation catalyst effectiveness is discussed further
- 215 in Section 3.3.

216	Table 3: NO _x , CH ₄ , PM and PN emissions from transient testing. U = urban, R = rural,
217	M = motorway, C = combined phases of the FIGE cycle.

	$NO_x (g/km)$			CH4 (g/km)				PM mass (mg/km)					PN (×10 ¹⁴ #/km)				
Ref.	U	R	М	С	U	R	М	С	U	R	М	С		U	R	М	С
A0	7.8	4.4	4.0	4.6	0.0	0.0	0.0	0.0	54.5	26.8	19.5	26.8		1.66	0.84	0.72	0.89
A1	7.4	3.6	1.9	3.3	8.5	6.5	11.4	9.2	77.8	48.3	13.8	35.1		1.89	1.10	0.43	0.87
A2	10.2	4.4	2.0	4.0	11.5	8.3	16.5	12.8	89.0	54.5	24.6	44.2		2.31	1.47	0.79	1.24
A3	12.3	4.4	2.1	4.2	3.9	6.6	11.8	8.8	29.4	36.3	12.7	23.7		0.61	0.85	0.37	0.58
A4	14.3	5.3	2.6	4.8	2.3	10.5	19.8	14.4	76.2	33.6	13.8	27.7		2.12	0.93	0.51	0.83
B0	7.3	1.0	0.3	1.5	0.0	0.0	0.0	0.0	28.7	10.7	6.9	11.2		0.99	0.28	0.22	0.34
B1	2.4	0.9	2.3	1.8	38.9	18.9	35.6	29.7	40.4	13.9	8.5	14.7		1.38	0.31	0.25	0.42
B2	19.7	2.8	2.6	4.4	0.1	24.7	45.8	33.0	13.0	6.3	5.1	6.3		0.38	0.14	0.15	0.17

218 The combined cycle relative contributions of all species to CO₂e emissions are shown in the 219 SI. For the dual fuel tests on vehicle B, the next largest contributor was N₂O emissions with 220 10% and 16% of the CO₂ contribution for tests B1 and B2 respectively. Measured N₂O 221 emissions for these tests were significantly higher than for B0 or for any of the tests with 222 vehicle A Tailpipe NO_x emissions during the motorway phase of B1 were significantly 223 increased compared to B0. This is despite post-turbo NO_x emissions being approximately 224 halved in B1 compared to B0 over the same phase (shown in the SI). For vehicle A dual fuel 225 tests, post-turbo NO_x emissions were reduced by ~40% and tailpipe NO_x by ~50% during the 226 motorway phase compared to the baseline A0. For A1, post-turbo and tailpipe NO_x emissions 227 were reduced by 30% compared to diesel over the combined cycle. Post-turbo NO_2/NO_x 228 ratios were also different for dual fuel compared to diesel; for A1 this ratio was 0.34 over the 229 combined cycle compared to 0.06 for A0 (shown in the SI). After the SCR, NO₂/NO_x ratios 230 decreased to 0.15 for A1 and increased to 0.22 for A0. Indeed at the tailpipe, dual fuel 231 NO_2/NO_x ratios are generally lower than for the diesel tests for both vehicles A and B. 232 Performance of the SCR systems over the combined cycle reduced from 43% to 34-41% 233 comparing A0 to A1 and A2, and 83% to 67% comparing B0 to B1. These results indicate 234 that duel fuel operation affects the efficacy of the OEM SCR systems. 235 Particle mass and particle number emissions in warm start dual fuel tests were higher than the 236 baseline diesel tests. The distance averaged particle size distributions (i.e. weighted by

237 exhaust volumetric flow and divided by cycle distance) are shown in Figure 1. Bimodal 238 distributions are present for all tests. Comparing A1 to A0, the PM mass emissions factor is 239 increased by $\sim 30\%$ due to a shift in the accumulation mode peak diameter from ~ 65 nm to 240 ~85 nm with total particle number emissions approximately equivalent. The peak of the 241 accumulation mode for vehicle B is not shifted significantly between B0 and B1, however the 242 PN emissions are increased by ~20% leading to a ~30% increase in total PM mass. Cold start 243 tests led to a more significant nucleation mode peaks and reduced accumulation mode peaks 244 relative to equivalent hot start tests. The contribution of BC mass to CO₂e emissions as a 245 percentage of the CO₂ contribution is between 1.8-4.3% for vehicle A and 0.5-1.3% for 246 vehicle B relative to CO₂. BC mass contributed between 60-80% of total PM mass, across all 247 transient tests, as shown in the SI along with uncertainties on the particle size distributions. 248 Steady state PM mass emissions for vehicle A are discussed further in the following section. 249 Engine power specific emissions factors for vehicle A over the combined FIGE/ETC cycle are shown in the SI and compared to EURO V regulatory values for engine dynamometer 250 tests over the equivalent cycle²⁸. The baseline A0 configuration is compliant with CO and 251 252 PM mass standards, however NO_x emissions are ~ 2.5 times higher than the standard (2.0 253 g/kWh). Indeed, NO_x emissions were higher than the EURO V standard for all vehicle A 254 configurations. Dual fuel operation led to increased CO emissions for A1, however the 255 oxidation catalyst on configuration A2 meant CO emissions were equal to those of A0. CH₄ emissions, which are only regulated for NG engines at type-approval²⁸, were 8.9 times higher 256 257 than the regulatory limit value (1.1 g/kWh) for the A1 configuration of this aftermarket dual 258 fuel system.



Figure 1: Distance specific particle size distributions over the combined FIGE cycle.

259 3.2 Steady state emissions

260 Steady state maps of ESR and CH₄ slip for A1, and the ratio of dual fuel to diesel (A1:A0) 261 CO₂ and CO₂e emissions as a function of engine speed and torque are shown in Figure 2. For 262 A1, the ESR reaches ~70% for engine speeds between 1200 and 1600 rpm and relatively low 263 engine torque around ~300 Nm. At higher engine torque output and engine speeds, the ESR is reduced (as controlled by the system supplier's own proprietary software); at 600 Nm, the 264 265 ESR is between 50-60% for the 1200-1600 rpm range. This can be cross referenced to the 266 average ESR over the motorway phase of the transient cycle measured to be 58% for A1, 267 during which the average engine torque and speed were approximately 600 Nm and 1400 rpm 268 respectively. CH₄ slip is greatest at higher engine speeds, indicating that incomplete 269 combustion of CH₄ is most significant when the in-cylinder residence time is lowest. The 270 ratio of CO₂ emissions for A1 versus A0 indicates that the greatest reduction in CO₂ 271 emissions is around 15% and that this occurs for engine speeds between 1000-1600 rpm and 272 engine torque between 300-500 Nm. This corresponds to areas of high ESR (50-70%) and 273 lower engine speeds. However, as shown for the transient cycle emissions, CH₄ emissions for 274 A1 lead to higher total CO_2e emissions over almost the entire map compared to A0; this ratio 275 is highest at low engine torques and higher engine speeds.



Figure 2: (a) Energy substitution ratio (ESR) of A1, (b) CH₄ slip of A1, (c) ratio of CO₂ emissions for A1:A0, and (d) ratio of CO₂ emissions for A1:A0 all as a function of engine speed and torque as measured during steady state testing of vehicle A.

276 Further evidence of incomplete combustion of CH₄ at higher engine speeds is provided in 277 Figure 3, which shows the dual fuel to diesel (A1:A0) ratio of post-turbo NO_x and CO 278 emissions and exhaust temperature. Dual fuel NO_x emissions are lower than diesel over most 279 of the map, however the greatest reductions are observed for engine speeds greater than 280 ~1800 rpm indicating lower average in-cylinder temperatures. This is further supported by 281 the map of post-turbo exhaust temperatures which are generally lower for A1 than A0 for 282 engine speeds greater than ~1800 rpm and engine torque greater than ~400 Nm. Post-turbo 283 CO emissions, a product of incomplete combustion, are greater across the entire map for A1 284 compared to A0, and at high engine speeds they are increased by an order of magnitude. 285 Steady state PM mass emissions are shown in Figure 3 as a ratio of those measured for configurations A1 and A0. In the previous section, we showed that PM mass emissions 286 287 increased for the dual fuel transient cycle tests compared to diesel. In Figure 3, it is evident 288 that PM mass emissions are up to 50% lower for A1 compared to A0 at engine loads below 289 600 Nm and engine speeds below 2000 rpm. However at engine loads greater than 600 Nm 290 and engine speeds less than 1500 rpm, PM mass emissions are increased for A1 compared to 291 A0 by up to a factor of 3.

13



Figure 3: Ratios of (a) tailpipe PM mass emissions, (b) post-turbo (PT) NO_x emissions, (c) PT CO emissions, and (d) PT exhaust temperature as a function of engine speed and torque measured during steady state testing of vehicles A1 and A0.

292 3.3 Oxidation catalyst effectiveness

293 The effectiveness of the oxidation catalysts fitted to vehicles A2 and B1 during steady state 294 testing is shown in Figure 4. The catalysed oxidation of CH₄ is a function of both residence time within the catalyst and exhaust gas temperature³⁷. The measured catalyst effectiveness 295 296 was greatest at high temperatures and residence times, i.e. low exhaust flow rates. For A2, the 297 highest observed catalyst efficiency was 27% for a post-turbo exhaust temperature of 470°C 298 and exhaust volumetric flow rate of 0.07 m^3 /s. For B1, the highest observed catalyst 299 efficiency was 30% for a post-turbo exhaust temperature of 505°C and exhaust volumetric flow rate of 0.10 m³/s. For context, the average post-turbo exhaust temperatures and exhaust 300 301 gas flow rates during the motorway phase of the transient cycle test were 396°C and 0.14 m³/s for A2 and 370°C and 0.16 m³/s for B1. These flow rates correspond to gas hourly space 302 velocities of approximately 190,000 h⁻¹ and 76,000 h⁻¹ for the oxidation catalysts on A and B 303 respectively. Below 400°C, the maximum performance of these catalysts were 15% and 10% 304 305 for A2 and B1 respectively and therefore these results support the transient emissions results 306 presented above that highlighted the significant contribution of tailpipe CH₄ to CO₂e 307 emissions.

308 A benefit of the oxidation catalysts is to oxidise increased CO emissions during dual fuel

309 operation to CO₂. Post-turbo and tailpipe CO emissions factors are shown in the SI. Even

310 without the CH₄ oxidation catalyst, CO emissions are reduced by 72%, 81% and 76% by the

311 SCR system for vehicles A0, A1 and A3 respectively. However, with the CH₄ oxidation

312 catalyst, CO emissions are reduced by 94% and 92% for A2 and A4 respectively. Similarly,

313 CO emissions were reduced by 95%, 93% and 97% for tests B0, B1 and B2 respectively.



Figure 4: CH₄ oxidation efficiency of the catalyst present on (a) vehicle A2 and (b) B1 as a function of exhaust flow rate and post-turbo exhaust temperature as measured during steady state testing.

314 **4 Discussion**

- 315 This study evaluated the emissions performance of two vehicle platforms with five
- 316 aftermarket dual fuel system configurations via chassis dynamometer testing of in-use
- 317 vehicles that are part of trials of low-carbon trucks in the UK. All dual fuel systems evaluated
- 318 in this study increased tailpipe total GHG (CO₂e) emissions compared to their equivalent
- diesel vehicles by 50% and 127% over the combined FIGE cycle for configurations A1 and
- 320 B1 respectively. This is despite CO₂ emissions being reduced by up to ~9% and is primarily a
- 321 result of incomplete combustion of CH₄ and subsequent CH₄ emissions, termed CH₄ slip. The
- 322 three main mechanisms of CH₄ slip are (i) valve overlap which causes a proportion of the
- 323 NG-air charge to be directly exhausted, (ii) incomplete combustion due to crevices and flame

324 quenching at the walls of the cylinder, and (iii) incomplete combustion due to lean NG-air mixtures and in-cylinder temperatures which prevent the flame from propagating throughout 325 the charge.^{21,38}Evidence for incomplete combustion and lower in-cylinder temperatures was 326 327 provided by post-turbo emissions measurements showing higher CO and lower NO_x 328 respectively at steady state conditions, especially at higher engine speeds when in-cylinder 329 residence times are reduced. The results of this in-use vehicle emissions study are consistent 330 with and supplement a large number of engine emissions studies showing that NO_x emissions 331 decrease and CO emissions increase with dual fuel combustion relative to diesel. Thus, 332 refinement of dual fuel systems to reduce CH₄ slip by addressing these three mechanisms and 333 to improve CH₄ combustion efficiency are required. Our results suggests that reducing ESR 334 at high engine speeds could have a beneficial effect on total CO_2e emissions and the 335 magnitude of this effect over a transient drive cycle should be the topic of further modelling 336 or experimental studies. Another example is to increase the diesel pilot quantity and advance the pilot injection timing²³, however trade-offs are increased NO_x emissions and lower ESR 337 338 and therefore a reduced CO₂ benefit compared to diesel combustion. Direct CH₄ injection and 339 stratification of the NG within the cylinder may also have the potential to reduce dual fuel 340 CH₄ emissions.

341 CH₄ slip can also be controlled by exhaust after-treatment, however the oxidation catalysts 342 tested in this study reduced CH₄ emissions by at most 15% at exhaust gas temperatures 343 representative of transient conditions (~400°C). Thus, the commercial development of 344 effective catalysts (e.g. Cargnello et al.³⁹) that successfully oxidise CH₄ below 400°C are 345 critical to the exploitation of NG as a transport fuel to reduce GHG emissions.

346 Both dual fuel platforms had higher PM mass emissions than their equivalent diesel 347 configurations in transient testing. Steady state testing revealed that PM mass emissions were 348 up to three times higher for dual fuel compared to diesel at high engine loads. While the 349 majority of previous studies report decreases in PM mass emissions for dual fuel compared to diesel combustion²¹, Papagiannakis et al.⁴⁰ reported an increase in soot opacity at high engine 350 351 loads (80%) and ESR in the range of 30-50%. It is possible that this phenomena occurs due to 352 the greater likelihood of rich combustion as a result of high diesel and NG flow rates during 353 high engine load conditions. PM mass emissions were up to 50% lower at low engine loads, 354 which is more consistent with existing literature and is attributable to the lower sooting 355 tendency of premixed combustion of CH4 compared to diffusion mode combustion of diesel²¹. 356

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357 Results for tailpipe NO_x emissions suggest that the change in exhaust gas composition in dual fuel operation could interfere with the efficacy of the OEM SCR system, which is optimised 358 359 for diesel operation. Post-turbo and tailpipe exhaust gas temperatures (shown in the SI) are 360 generally higher in the transient dual fuel tests compared to diesel, which may result from heat release via combustion of unburned hydrocarbons over the oxidation catalyst. At higher 361 362 exhaust gas temperatures and higher NO_2/NO_x ratios the conditions in the SCR may be 363 significantly different when the trucks are in dual fuel mode compared to diesel, thus affecting its performance⁴¹. Higher N₂O emissions for the dual fuel tests compared to diesel 364 tests could be due to (i) higher rates of oxidation of ammonia to N₂O by NO₂ potentially 365 caused by higher NO₂/NO_x ratios at temperatures up to $350^{\circ}C^{42}$, (ii) direct oxidation of 366 ammonia by oxygen to N₂O at temperatures above $350^{\circ}C^{43}$ or (iii) by thermal decomposition 367

368 of ammonium nitrate 44,45 .

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374 6 Supporting information

Additional details describing experimental methods, supplemental results and an uncertainty
analysis are included in the Supporting Information (SI). This information is available free of
charge via the Internet at http://pubs.acs.org.

378 7 Nomenclature

$ ho_0$	Material density (g/cm ³)
$ ho_{ m eff}$	Effective density (g/cm ³)
AR5	IPCC's Fourth Assessment Report
AR5	IPCC's Fifth Assessment Report
BC	Black carbon

С	Combined phases of FIGE drive-cycle
CO ₂ e	Carbon dioxide equivalent
D _m	Mass-mobility exponent
d_{p}	Particle mobility diameter (nm)
$d_{ m pp}$	Primary particle diameter (nm)
ESR	Energy substitution ratio (%)
ETC	European transient cycle developed by the FIGE institute
FTIR	Fourier transform infrared spectroscopy
GHG	Greenhouse gas
GWP	Global warming potential
НС	Unburned hydrocarbons
HGV	Heavy goods vehicle
HPDI	High-pressure direct injection
IPSD	Integrated particle size distribution
LCV	Lower (net) calorific value
LNG	Liquefied natural gas
Μ	Motorway (highway) phase of FIGE drive-cycle
$\dot{m}_{ m CH_4,in}$	Mass flow rate of CH ₄ supplied to the engine (kg/s)
$\dot{m}_{ m CH_4,tailpipe}$	Mass flow rate of CH ₄ exhausted at the tailpipe (kg/s)
$\dot{m}_{ m diesel,in}$	Mass flow rate of diesel supplied to the engine (kg/s)
$\dot{m}_{ m NG,in}$	Mass flow rate of natural gas supplied to the engine (kg/s)

NG	Natural gas
OEM	Original equipment manufacturer
PM	Particulate matter
PMP	Particle Measurement Programme
PN	Particle number
РТ	Post-turbo (emissions sampling point)
U	Urban phase of FIGE drive-cycle
R	Rural phase of FIGE drive-cycle
rpm	Revolutions per minute
SCR	Selective catalytic reduction
Т	Temperature (°C)

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