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26 **Keywords:** Flash boiling; Tomographic reconstruction; Plume direction; Plume-to-plume interaction; Projected liquid  
27 volume; Spray cone angle

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#### 34 **Abbreviations**

AMR	Adaptive mesh refinement	PDI	phase Doppler interferometry
CFD	Computational fluid dynamics	PFI	Port fuel injection
ECN	Engine Combustion Network	PLV	Projected liquid volume
EGR	Exhaust gas recirculation	PN	Particulate number
GDI	Gasoline direct injection	RANS	Reynolds averaged Navier-Stokes
KH	Kelvin-Helmholtz	RT	Rayleigh-Taylor
LED	Light emitting diodes	SOI	Start of injection
LES	Large eddy simulation	SPCCI	Spark controlled compression ignition
LTGC	Low temperature gasoline combustion	TDC	Top dead center
ON	Octane number		

#### 35 **Nomenclature**

$\alpha_p$	volume fraction of parcel in a cell [a.u.]	$P_a$	Ambient pressure [Pa]
$C_{ext}$	Extinction coefficient [mm <sup>2</sup> ]	$P_v$	Vapor pressure [Pa]
$d$	Droplet diameter [mm]	$\varphi$	Equivalence ratio [a.u.]
$F/A$	Fuel to air ratio [a.u.]	$Re$	Reynolds number [a.u.]
$I$	Image frame with spray [a.u.]	$\rho_f$	Fuel density [kg/m <sup>3</sup> ]

$I_o$	Averaged background image [a.u.]	$\tau$	Optical thickness [a.u.]
$m_{cell}$	Total mass in a cell [kg]	$V_{cell}$	Volume of a cell [m <sup>3</sup> ]
$m_{f_l}$	Liquid fuel mass in a cell [kg]	$We$	Weber number [a.u.]
$m_{f_v}$	Vapor fuel mass in a cell [kg]	$x_{N_2}$	Nitrogen mass fraction in a cell [a.u.]
$m_{N_2}$	Nitrogen mass in a cell [kg]	$y$	Projection direction [a.u.]

## 36 1. Introduction

37 Gasoline direct injection (GDI) engines have been adopted by the automotive industry over the past few years.  
38 Different from conventional port fuel injection (PFI) engines, fuel is injected directly into the combustion chamber,  
39 creating charge cooling and higher turbulence. These improvements mitigate knocking tendency and improve engine  
40 efficiency. A combination of direct fuel injection with **additional engine-operation** technological **enhancements** such  
41 as high-pressure injection, multiple injection strategies, exhaust gas recirculation (EGR) and enhanced air utilization  
42 by swirl or tumble motion enable GDI engines to be operated under fuel-lean conditions with even higher efficiency  
43 [1-4]. Furthermore, advanced combustion concepts such as low-temperature gasoline combustion (LTGC) and spark-  
44 controlled compression ignition (SPCCI) have been considered as next-generation GDI engine technologies [5-6].  
45 These concepts utilize early fuel injection during the intake stroke to form a homogeneous air-fuel mixture, followed  
46 by injection during compression to create a stratified charge near the spark plug and stable flame kernel. For **such**  
47 **technologies** to succeed, spray control during all **stages of the operating** cycle is of utmost importance. Inappropriate  
48 injection parameters and fuel properties cause fuel impingement on the cylinder wall and result in unacceptable  
49 particulate matter (PM) emissions [7]. Thus, understanding the direction and ultimate penetration of each liquid plume  
50 of the injector under various ambient conditions and fuels is crucial to improve combustion and emission  
51 characteristics.

52 **Gasoline sprays exhibit a distinct topology evolution** under flash-boiling conditions. Flash boiling occurs at throttled  
53 or early-injection conditions when ambient pressure is lower than the saturation pressure. Rapid changes in  
54 temperature and the velocity field are accompanied by a bulk conversion of fuel from liquid to vapor during injection.  
55 Common observations under flash-boiling conditions are shorter liquid penetration length, enhanced atomization,  
56 lower mixture temperature, and large recirculating vortex closer to the injector tip [8-10]. The ratio of ambient pressure  
57 to vapor pressure ( $P_a/P_v$ ) has a dominant impact on spray process under flash-boiling conditions, unlike non-flashing

58 sprays where relationship such as Reynolds number ( $Re$ ) and Weber number ( $We$ ) are typically used to summarize  
59 spray characteristics [11-13]. A key feature of flash-boiling operation is that vapor formation commences when  
60 pressure decreases as fuel flows through the nozzle, even without any fuel-air mixing or heat transfer, thereby creating  
61 a wider radial dispersion of individual injector plumes. Plume dispersion increases with higher fuel temperature or  
62 lower ambient pressure (i.e. lower  $P_a/P_v$ ). The larger plume dispersion enhances plume-to-plume interaction and alters  
63 velocity and pressure field, which may lead to deflection of the plume direction or complete combinations of plumes  
64 and collapse at certain conditions of injection timings. In the previous study of Zeng et al., it was found that sprays  
65 from an eight-hole injector with a nominal total spray angle of  $60^\circ$  had a linear increase in liquid spray width as the  
66 pressure ratio decreased from 1.0 to 0.3, and finally spray collapse occurred at pressure ratios under 0.3, showing one  
67 integrated large plume [14]. Flow analysis using particle image velocimetry (PIV) showed the vortex around spray  
68 plumes strengthening under lower pressure cases due to enhanced momentum transfer by interaction between thicker  
69 spray plumes and ambient air [15]. Plumes also move towards each other during time, accelerated by a larger pressure  
70 difference between the center of the spray and the ambient air [8], and complete collapse occurs under flare flash-  
71 boiling conditions.

72 Fuel properties affect spray mixing over a range of conditions, including flash-boiling operation. As the use of  
73 renewable fuels increases, it is important to understand how varying properties affect fuel delivery in either neat fuels  
74 or when blended with refinery stock [16]. Properties considered important for limiting liquid impingement upon in-  
75 cylinder surfaces include distillation characteristics, heat of vaporization, vapor pressure, surface tension, viscosity  
76 and so forth. Over the past decades, many researchers have investigated the effects of fuel properties on spray  
77 characteristics summarized by penetration length, spray width, and spray residual time [17-20]. The majority of these  
78 studies rely on two-dimensional measurements utilizing Mie-scattering or extinction imaging for spray  
79 characterization. The diagnostic approaches are relatively simple and able to provide information about global  
80 characteristics of the spray, however, they cannot describe how individual plume motion and growth is affected by  
81 fuel properties. Thus, spray analysis in three-dimensional space is essential for a more complete understanding of fuel  
82 property influence on plume direction and interaction. Moreover, there is lack of understanding how vapor pressure  
83 or shape of distillation curve affect to plume direction and spray collapse. Therefore, in-depth evaluation on the effects  
84 of vapor pressure and distillation curve on plume direction and plume collapse is needed.

85 Numerous non-intrusive optical diagnostics have been applied to investigate the internal motion of plumes and the  
86 mixing process, rather than being limited to line-of-sight information. Among the techniques, laser diagnostics were  
87 widely employed. In one example, the peak velocity magnitude in a spray plane measured by phase-doppler  
88 interferometry (PDI) was analyzed to represent plume direction movement throughout injection [21]. Planar laser  
89 imaging with corrections for multiple-scattering has been performed, however, light attenuation issues still inhibit  
90 quantification [22]. On the other hand, x-ray radiography with tomographic reconstruction has been applied to identify  
91 individual plume direction [23-24]. X-rays have high penetrating power and negligible scattering to enable data  
92 acquisition even in the near-nozzle region, but measurements have been performed only under low-temperature  
93 atmospheric ambient conditions at a few planes. A commercial optical patternator (En'Urga Inc, SETscan AP400)  
94 using line-of-sight measurements along a plane has been applied to windowed spray chambers at engine-relevant  
95 pressures and temperatures including intake-injection as well as late-injection conditions [25]. By rotating the injector  
96 in 22.5° increments for a total of eight views over 180° rotation, sufficient data was collected to enable computed  
97 tomography along the plane. The technique showed success in identifying major changes in plume position and growth  
98 with variation in chamber conditions, including flash-boiling conditions. However, the technique was limited to only  
99 1 kHz temporal resolution, which is essentially an average over the entire injection event [25]. Further work is needed  
100 to identify the plume direction and growth under engine-like conditions with improved temporal and spatial resolution.  
101 The techniques described above have drawbacks in terms of complexity in measurement, high-cost in measuring  
102 equipment, laborious measurement procedures, or lower spatial and temporal resolution.

103 In this study a new three-dimensional tomographic reconstruction method using ensemble averaged extinction images  
104 was developed to overcome technical issues that conventional diagnostic approaches have. This new method can be  
105 applied in the entire spray domain and injection period with a much simpler setup compared to conventional laser or  
106 x-ray facilities. High-speed Mie-scattering, diffused back illumination extinction, and schlieren imaging were  
107 performed in a constant flow vessel using the Engine Combustion Network (ECN) Spray G injector. The primary  
108 objective of this work is to understand plume direction and spatial mixing quality based on tomographically  
109 reconstructed spray in three-dimensional space. Four different fuels including single component fuel iso-octane, the  
110 multi-component fuel di-isobutylene, olefinic, and e30 blend were applied under flash-boiling and early injection  
111 ambient conditions. The tested fuels are considered as promising alternative fuels for conventional gasoline because  
112 of high research octane number (RON, 98) and production from renewable sources. Furthermore, large differences in

113 vapor pressure and distillation characteristic between fuels also provide opportunity to investigate their effects on  
114 plume direction and collapse process. Augmenting interpretation of the new experimental data, in this study  
115 computational fluid dynamics (CFD) simulations using CONVERGE code were also performed to estimate mixture  
116 equivalence ratio effects on vaporization, as these data have not yet been acquired in the experiment.

## 117 **2. Test method and condition**

### 118 **2.1. Injector and fuels**

119 An ECN-designated Spray G injector (primary injector serial number: AV67-028) was utilized for fuel injection. The  
120 Spray G injector is a solenoid driven gasoline direct injector that has nominally axisymmetric eight-holes. The detailed  
121 dimensions and specifications are presented in Fig. 1 [26]. The nozzles were designed to have a counter bore shape,  
122 which has an inner orifice diameter of 165 $\mu\text{m}$  and a step diameter of 388 $\mu\text{m}$ . The nozzles were drilled with an angle  
123 of 37° from the center axis of the nozzle tip. A standard ECN Spray G electronic injector driver was used to hold an  
124 injection profile consistent with previous studies. The electronic command was generated by an external data  
125 acquisition system and the signal was delivered to the injector driver.

126 Four different test fuels, single component iso-octane, multi-component surrogate di-isobutylene, multi-component  
127 fuel with olefin molecular structure, and e30 blend fuel were tested and their properties are shown in Table 1. Among  
128 the fuels, e30 blend has the highest density, viscosity, and heat of vaporization amongst test fuels. The e30 blend has  
129 the lowest energy content due to oxygenated species (30% ethanol). The vapor pressures of olefinic and e30 blends at  
130 90°C are approximately 2.5 and 4 times larger than iso-octane, respectively. The effect of compositional difference  
131 between fuels is illustrated in distillation and vapor pressure curves presented in Fig. 2 [27]. Ambient pressure of test  
132 conditions are also presented in Fig. 2. The distillation curve of iso-octane is shown as a straight line at the boiling  
133 point. A pseudo distillation curve for the di-isobutylene blend is provided, based on the boiling point temperatures of  
134 five different species: 1-hexene (4% v/v), n-heptane (12.1% v/v), iso-octane (44.2% v/v), toluene (20.1% v/v), and di-  
135 isobutylene (19.6% v/v). The curve remains relatively flat because these species have a relatively narrow range of  
136 boiling point temperatures. It is noted that even though the di-isobutylene blend contains species heavier than iso-  
137 octane, the vapor pressure is higher because of the existence of light species (1-hexene). On the other hand, olefinic  
138 and e30 blends have light and heavy components contributing to a wide range of distillation temperature from 50°C

139 to 200°C. The large amount of light species in olefinic and e30 blends creates higher vapor pressure, and this trend is  
140 maintained throughout the temperature range.

## 141 **2.2. Vessel system and optical setup**

142 Spray test were conducted in a spray vessel at constant pressure and gas flow rate at controlled temperature. The  
143 constant-flow spray vessel system has many advantages compared to optical engines or closed (constant-volume)  
144 spray chambers: 1) Thermodynamic parameters can be controlled independently in the vessel system. Analysis of  
145 spray process at known boundary conditions promotes understanding the effects of each thermodynamic parameter  
146 on spray characteristics. 2) The flow-vessel system has a much higher repetition rate than a closed chamber, which  
147 must be vented and prepared to a target condition before each injection. 3) The vessel system has much larger optical  
148 access than in an optical engine, where the viewing area is limited to a portion of the piston or cylinder liner. This  
149 feature facilitates simultaneous imaging for different views and multiple diagnostics. 4) There is less liquid  
150 impingement upon surfaces in the spray vessel than in optical engines. With limited space, the spray in optical engines  
151 can create wetting on the cylinder wall or piston depending on the ambient condition. On the contrary, the spray vessel  
152 has much more spacious volume so the entire spray process can be recorded without wall impingement. Ultimately,  
153 the goal of the spray vessel dataset is to provide reliable experimental data with tightly controlled boundary conditions  
154 that can be utilized as a validation source for modelling work.

155 A schematic of the constant-flow spray vessel is presented in Fig. 3. The vessel has an injector port and three optical  
156 windows. Quartz windows which have a diameter of 130mm are installed in the three ports for optical access. Internal  
157 heaters and an internal optical section are designed to be thermally isolated (with insulation) from the outer pressure  
158 vessel. The designed maximum operating pressure and temperature are 150 bar and 1100 K, respectively. Water  
159 cooling jackets in the wall, exhaust pipe, and the outer window ports are utilized to cool the pressure vessel, especially  
160 under high ambient temperature conditions. Sprays are injected into a continuous nitrogen flow (~0.1m/s) controlled  
161 by an electro-pneumatic flow controller. The rate of nitrogen flow is measured by flow meters at the upstream of the  
162 vessel and at the exhaust pipe. A customized vacuum pump is used to reach sub-atmospheric conditions in the vessel.  
163 The vessel pressure is measured by three different pressure transducers at the vessel inlet line, used for control and  
164 appropriate pressure range. The nitrogen flow enters a heating coil surrounded by an insulator piece at the bottom part  
165 of the vessel and then moves through a diffuser to enhance uniformity in the velocity and temperature field. To monitor

166 temperature distribution prior to spray events, 24 thermocouples were installed above the diffuser, under the spray,  
167 and above the spray region. The vessel region corresponding to gases that mix with the spray was controlled to a given  
168 target temperature. The mixture of injected fuel and nitrogen gas scavenges through the exhaust pipe at the top of the  
169 vessel, replaced by a fresh charge of ambient gas before the next injection. Relevant temperature, oxygen  
170 concentration, and vessel pressure are digitally recorded.

171 Two different optical setups were employed in this study. The first setup is for liquid fuel visualization by Mie-scatter  
172 extinction and scattering imaging at the side and front views, respectively. The liquid-phase fuel was identified by the  
173 diffused back illumination extinction imaging technique shown in Fig. 3. A high-speed green light-emitting diode  
174 (LED), Fresnel lens (150mm, f=150mm), engineered diffuser (20°), and band pass filter (center wavelength: 527nm,  
175 bandwidth: 20nm, full width-half max: 22nm) were utilized. The engineered diffuser (and collection optics) supplied  
176 a homogeneous light field and suppressed beam steering by evaporation or temperature field in the vessel [29]. This  
177 imaging technique is designed to collect extinction only by liquid phase fuel, but not from fuel vapor. The recording  
178 was extended up to 5ms after the start of injection (SOI) to capture the entire spray development and evaporation  
179 process. Normalized by incident light intensity (and other optical parameters), the side-view extinction imaging is  
180 quantitative for projected liquid volume (PLV) as explained below. Mie-scatter imaging from the front view was used  
181 primarily for visualization of plume-to-plume interaction effects. Here in the first optical setup, extinction and Mie-  
182 scattering imaging were carried out simultaneously using two different cameras. The second optical setup is Z-type  
183 schlieren imaging to capture the extent of fuel vapor penetration along a line of sight [30]. An effective point source  
184 was created using light from a red LED (500ns duration) collected by lens (Nikkor, 50mm f/1.8) and then focused  
185 through an aperture (0.6mm diameter). The expanding light collimated by a concave mirror (GSO, 152mm f/6.0) and  
186 the parallel rays of light passed through spray region. The parallel light after the spray was then collected by an  
187 identical concave mirror, with knife-edge (~50% cut-off) was located at the focal point to enhance sensitivity. Table  
188 2 lists detailed values for optical imaging setup.

## 189 **2.3. Image processing method**

### 190 **2.3.1. Extinction image and tomographic reconstruction**

191 [Liquid characterization using projected liquid volume \(PLV\) through](#) extinction imaging is highly recommended by  
192 the ECN community. [Thresholding with a certain PLV provides a great potential to resolve experimental issues.](#) In

193 the past, liquid penetration was usually measured by Mie-scattering imaging and arbitrary thresholding, associated  
194 with high uncertainty [31]. At the same time, an arbitrary threshold value based on pixel intensity was utilized so  
195 liquid length can be varied according to optical setup and research institute even if the spray test was performed under  
196 identical target condition. Therefore, extinction imaging with diffused backlit was suggested to measure optical  
197 thickness, not scattered light intensity as in Mie-scattering imaging. Using optical thickness rather than Mie image is  
198 much more universal and it enables direct comparison with CFD simulation result. At first to acquire PLV, the optical  
199 thickness in a spray region can be calculated based on Beer-Lambert law as equation (eq-1).

$$200 \quad \tau = -\ln(I/I_o) \quad (\text{eq-1})$$

201 Where  $I$  is transmitted light intensity with extinction (from droplets) and  $I_o$  is incident light intensity without  
202 extinction. This level of transmission is reasonable for detection of spray “edge” above the noise floor of the camera,  
203 but considerations such as vapor-phase beam steering need consideration [29].

204 The measured optical thickness  $\tau$  may be related to the PLV, which is the integral of liquid volume fraction (LVF)  
205 along the cross-stream direction  $y$ , as

$$206 \quad \text{PLV} = \tau \frac{\pi d^3/6}{C_{ext}} = \int_{-y_\infty}^{y_\infty} (\text{LVF}) \cdot dy \quad (\text{eq-2}).$$

207 Mie scattering and extinction theory is applied in eq-2, along with the simplifying assumption that droplet diameter  
208  $d$  and extinction coefficient  $C_{ext}$  (which depends upon  $d$ ) do not vary along the line of sight. The PLV has units of  
209 liquid volume per projected area, or  $\text{mm}^3(\text{liquid})/\text{mm}^2$ , and may be easily calculated from CFD simulations for  
210 comparison to experiment. However, the experimentalists must have access to information about  $d$  and  $C_{ext}$  to  
211 estimate PLV. In particular,  $C_{ext}$  depends upon droplet size, wavelength of light, and collection angle of the receiving  
212 optics (e.g. some forward-scattered light is collected). Fortunately, droplet diameter measurements have been  
213 performed for Spray G by ECN participants at General Motors and Shanghai Jiao Tong University using phase-doppler  
214 interferometry (PDI) [26]. The measurements show an SMD near  $7\mu\text{m}$  with fair uniformity across the plume during  
215 injection, which justifies the assumption to use single values for  $d$  ( $7\mu\text{m}$ ) and  $C_{ext}$  ( $72.70 \cdot 6 \text{ mm}^2$ , from Mie-theory  
216 relations using experimental collection angle of  $69.3\text{mrad}$ , light wavelength of  $527\text{nm}$ , refractive index of  $1.391$  and  
217  $7\mu\text{m}$  droplet diameter, MiePlot available at [32] was used). Note that droplet size measurements were performed only  
218 upstream at one axial position (at  $z = 15 \text{ mm}$ ). We make use of these droplet size measurements only during injection

219 because droplets produced upstream during injection constitute the majority of droplets in the domain—they are  
220 transported downstream and reside after injection has ended. [We will discuss uncertainties associated with the droplet  
221 size assumptions in the Appendix section.](#)

222 For the macroscopic spray characterization, a threshold for PLV may be chosen to indicate the extent of liquid  
223 penetration and width. The ECN recommends thresholds of  $0.2 \cdot 10^{-3}$  or  $2 \cdot 10^{-3}$  mm<sup>3</sup>(liquid)/mm<sup>2</sup> [33]. [This is correlated  
224 to past definitions of liquid length \(3% of maximum intensity in Mie-scattering image, optical thickness of 0.374 by  
225 laser-derived measurement \(Please refer to \[34-35\]\)](#) In this study, the lower threshold value of  $0.2 \cdot 10^{-3}$   
226 mm<sup>3</sup>(liquid)/mm<sup>2</sup> was applied to process and binarize PLV images according to this threshold value. In the binarized  
227 image, liquid penetration length was defined as the farthest axial distance from the nozzle at the primary view point  
228 ( $0^\circ$  rotation angle) and the spray liquid width was measured at axial distances of 15mm and 25mm away from the  
229 nozzle tip.

230 Ensemble-averaged PLV data from 300 injections at [three different viewing angles](#) were utilized for tomographic  
231 reconstruction. Fig. 4 indicates extinction images at three different views and corresponding PLV images in the  
232 middle. The built-in ‘iradon’ function in Matlab was used for tomographic reconstruction, using a Hamming filter  
233 value of 0.7. It is noted that reconstruction routine creates artifacts in optically thick region so each individual plumes  
234 could not be identified for near nozzle spray ( $\sim z=5$ mm), however, comparison with x-ray data revealed that it predicts  
235 plume center correctly at  $z=12$ mm. For the first step, PLV data at a certain location (presented as line in the PLV  
236 images) need to be extracted and placed in a map shown at the bottom of the figure. Because of the limited injector  
237 turns, the data within the viewing angle was interpolated and then the data out of viewing angle was extrapolated by  
238 using the map from  $0^\circ$  to  $22.5^\circ$ . The interpolation and extrapolation were performed by linearly-weighted averaging.  
239 Through this method, a projection map from  $0^\circ$  to  $180^\circ$  is generated for each axial position. After tomographic  
240 reconstruction, a three-dimensional liquid volume fraction distribution is generated at each time after the start of  
241 injection. The detailed description and validation of reconstruction method can be found in [36], including  
242 confirmation of the process using synthetic model data for liquid volume fraction.

### 243 **2.3.2. Schlieren image**

244 Schlieren images under high-pressure and high-temperature conditions are sensitive to fuel vapor (with a different  
245 refractive index than the ambient), however, non-uniformities in refractive index created by window thermal boundary

246 layers must also be considered [37]. The problem is especially severe if the window thermal boundary layer moves  
247 during injection, making image normalization at the beginning of injection unsuitable for processing. However, in the  
248 present study an image correction before the start of injection was sufficient to identify the jet vapor boundary. The  
249 image processing routine for schlieren results is shown in Fig. 5(x). First (a), the spray was isolated from the initial  
250 background  $I_o$  by subtracting spray image  $I$  from  $I_o$ . (b) The vapor envelope was identified by using the Matlab  
251 ‘imgradient’ function, returns the spatial gradient magnitude. (c) The gradient magnitude was binarized with a certain  
252 threshold value and (d) the area inside the vapor boundary (where gradient magnitude is low because of liquid spray)  
253 was filled to create a single, connected vapor boundary (e), produced using the ‘bwperim’ Matlab function and  
254 presented as red line in Fig. 5(e) as a visual check of the process. Vapor penetration length is defined at each time and  
255 injection as the distance between the nozzle tip and the farthest axial length on the boundary. (f) A vapor probability  
256 map was also derived using results from multiple injections. For 100 injections, the binarized image (d) from each  
257 injection and time step was summed to represent the probability percent. The region between 0 to 100 represents a  
258 zone of intermittency for low fuel mixture fraction, which can be evaluated with some rigor when compared to CFD.

#### 259 **2.4. Computational setup**

260 Lagrangian parcel spray simulations using CONVERGE (v2.4) were performed to estimate mixing effects  
261 (equivalence ratio) on evaporation and overall spray development. The simulation work in this study mainly used  
262 default CONVERGE settings for an example Spray G case, so only a brief review of the setup is provided here. The  
263 injector specifications for the 8-hole nozzle are given in Fig. 1. The modified settings from the example case were  
264 plume cone angle ( $25^\circ$ ), plume direction angle ( $34^\circ$ ), domain size (to enlarge without wall impingement), and  
265 turbulence model. [Initial values for the plume cone angle and plume direction angle eventually used for the simulations](#)  
266 [were determined by comparing the liquid-penetration predicted values with the experimentally derived PLV values.](#)  
267 Simulations were performed for Spray G conditions using iso-octane as fuel at ambient conditions matching the  
268 experiment. Following the Spray G work of Saha et al. [38], Reynolds averaged Navier-Stokes (RANS) standard k- $\epsilon$   
269 model was used instead of the large-eddy simulations (LES) example case. Beginning with blob injection at the size  
270 of the nozzle, O’Rourke dispersion, Kelvin-Helmholtz (KH) and Rayleigh-Taylor (RT) models were used to capture  
271 turbulent dispersion, primary and secondary breakup, respectively, while the Frossling model is used for evaporation.  
272 An injection rate profile provided by ECN was used for the simulation [26]. The simulation time was extended up to

273 5ms after start of injection (SOI) to match with experimental data. The computational domain and detailed setting for  
274 simulation are presented in Fig. 6 and Table 3, respectively.

## 275 **2.5. Operating conditions**

276 The spray experiments were carried out under ECN Spray G2 (flash-boiling), G3 (early injection), and G3HT (G3  
277 with elevated temperature conditions). As per ECN Spray G standard, an electronic injection command with a duration  
278 of 680 $\mu$ s (780 $\mu$ s actual hydraulic duration) was delivered to inject 10mg of fuel with injection pressure of 200 bar.  
279 The temperature of injector tip was maintained by a water circulator at 90°C during entire experiments. The ambient  
280 temperature was set to 60°C for G2 and G3 condition, but increased to 120°C under G3HT condition. The ambient  
281 pressures were 0.5 bar, 1 bar, and 1.18 bar for G2, G3, and G3 HT, respectively. Corresponding ambient densities  
282 were 0.5 kg/m<sup>3</sup> for G2, and 1.01 kg/m<sup>3</sup> for G3 and G3HT conditions. Three different injector viewing angles were  
283 applied and the spray was repeated 300 times per case for extinction imaging. The schlieren imaging was repeated for  
284 100 injections at the primary viewing angle. The experimental conditions are summarized in Table 4.

## 285 **3. Results and discussion**

### 286 **3.1. Effects of ambient condition on macroscopic spray characteristics**

287 Providing a general overview of the baseline iso-octane sprays, the liquid and vapor envelopes under G2, G3, and  
288 G3HT conditions are presented in Fig. 7 ([Data available at \[39\]](#)). Recall that the liquid and vapor results here are  
289 derived from ensemble averaging many different injection events. The liquid boundaries determined with PLV  
290 threshold of 0.2·10<sup>-3</sup> mm<sup>3</sup>(liquid)/mm<sup>2</sup> are compared according to ambient conditions with vertical lines indicating  
291 liquid penetration length in Fig 7(a). Fig. 7(b) shows vapor boundary (dotted) with 20% intermittency and liquid  
292 boundary (solid) as defined above. The quantitative comparison of liquid penetration length and liquid width is given  
293 in Figs 7(c) and 7(d). [In the plots, standard error \(-2 \$\sigma\$ / \$\sqrt{n}\$  to +2 \$\sigma\$ / \$\sqrt{n}\$ \) of the data is shown as a shade area. Error](#)  
294 [analysis showed that shot-to-shot variations in spray penetration and width converged after 100 injections.](#) Results  
295 from the CFD simulation were also analyzed by applying the identical PLV criteria as the experiment, and results for  
296 the G2 condition are shown for simplicity.

297 Analysis of macroscopic spray characteristics shows that the spray under G3HT condition evaporates much faster than  
298 G2 and G3 conditions, which is expected because of the higher ambient temperature. Higher ambient density also

299 tends to slow vapor penetration, and results are indistinguishable for G3 versus G3HT. Higher ambient temperature  
300 confines liquid to a smaller radial width, which also eventually applies to axial penetration. However, the more  
301 remarkable effect is that after liquid penetrates to approximately the same maximum axial position, [the time of](#)  
302 [complete evaporation](#) to the PLV criteria is much earlier for G3HT (about 2 ms aSOI) compared to that of G3 (about  
303 3.5 ms aSOI). The vapor envelope is similar for either G3 or G3HT conditions. The G2-condition spray shows the  
304 longest vapor penetration length, which is expected because of the lower ambient density. Liquid penetration is also  
305 the longest, despite the enhancement in vaporization expected for the G2 flash-boiling condition. The spray boundary  
306 images and liquid width measurements indicate a transition in spray mixing at some point. While the near nozzle spray  
307 outline (refer Fig. 7(a)) under G2 and G3 conditions is similar at 0.5 and 0.8ms, contraction of the spray in radial  
308 direction begins under flash-boiling G2 conditions, leading to smaller liquid width than the G3 condition. Factors such  
309 as potential for a larger individual plume growth for G2 condition and overall 3D liquid plume movement deserve  
310 consideration, and we will discuss these issues after 3D data are presented.

311 [One of the most interesting](#) results from the macroscopic liquid spray parameters is the extended persistence of liquid  
312 in both space and time, particularly for the flash-boiling G2 condition with liquid persistence exceeding 3 ms. The  
313 long persistence of liquid time under G2 and G3 conditions, even with heated fuel and somewhat elevated gas  
314 temperature, is considered significant time scale in an engine system. There is clearly enough penetration distance and  
315 time for liquid to impinge upon in-cylinder surfaces during intake injection in an engine. To gain some physical insight  
316 into factors affecting vaporization, we perform a thermodynamic analysis for different mixtures (equivalence ratio),  
317 guided by CFD simulations for prediction of these mixtures.

318 Equilibrium-state calculations are performed with liquid and vapor fraction predictions for different mixtures, using  
319 standard energy and fugacity balances for liquid fuel and gas initially at the conditions of the experiment. [This](#)  
320 [calculation predicts the mixture composition \(solution\) and temperature according to equivalence ratio. For example,](#)  
321 [a small amount \(low equivalence ratio\) of liquid fuel at fuel temperature of 90°C under G2 condition \(0.5bar, 60°C\)](#)  
322 [will be evaporated. In this process, heat is absorbed due to phase change, thus mixture temperature decreases.](#)  
323 [However, achieving complete evaporation under high equivalence ratio is not possible, thus, a certain amount of liquid](#)  
324 [will persist. In other words, the term ‘solution’ refers to vapor only or liquid/vapor mixture under complete and](#)  
325 [incomplete evaporation conditions, respectively.](#) The calculation result shown in Fig. 8 present the equilibrium

326 mixture temperature according to a given equivalence ratio for G2, G3, and G3HT conditions. If the solution is for  
 327 entirely vapor, a dashed line is indicated. If the solution shows a mixture with both liquid/vapor present, a solid line  
 328 is used. The transition point is a saturated vapor-liquid equilibrium mixture. If vaporization is driven entirely by  
 329 mixing, and perfect equilibrium mixtures are formed, no liquid will exist at equivalence ratios less than this transition.  
 330 While this assumed state is unlikely in a dynamic spray, this very assumption has been applied with encouraging  
 331 results at diesel conditions at higher temperature and pressure [40]. Fig. 8 shows that the rank ordering for saturated  
 332 equivalence ratio is in agreement with the experiment in terms of vaporization time. But it also shows that substantial  
 333 vaporization cooling occurs under all conditions. Fuel initially at 363 K and ambient initially at 333 K cools below  
 334 300 K at saturated conditions, and the cooling is more substantial for the flash-boiling condition. Moreover, an  
 335 equilibrium condition with liquid exists even for flash-boiling conditions. Therefore, it is not surprising to observe  
 336 liquid for an extended period of time as even equilibrium calculations show a liquid state provided that the mixture  
 337 equivalence ratio is rich enough.

338 CFD simulations are utilized to estimate the actual equivalence ratio, in comparison to equilibrium expectations. Fig.  
 339 9 shows mixture total equivalence ratio (considering both liquid and vapor) predictions both during and after injection  
 340 at positions where liquid is observed in the experiment. The images present equivalence ratio at two different planes  
 341 and timings after start of injection (aSOI) timings. The Lagrangian parcel simulation does not provide total (liquid and  
 342 vapor) fuel-air ratio by default, so it was calculated using cell mass, mass fraction, volume fraction of parcel in a cell,  
 343 cell volume, and fuel density, as shown by equation (3) to equation (5) below

$$344 \quad m_{f_v} = m_{cell} - m_{N_2} = m_{cell} - m_{cell} \cdot x_{N_2} \quad (\text{eq-3})$$

$$345 \quad m_{f_l} = V_{cell} \cdot \alpha_p \cdot \rho_f \quad (\text{eq-4})$$

$$346 \quad \varphi = \frac{(F/A)_{actual}}{(F/A)_s} = \frac{(m_{f_v} + m_{f_l})/m_{N_2}}{(F/A)_s} \quad (\text{eq-5})$$

347 where  $m_{f_v}$  is vapor mass of iso-octane in a cell,  $m_{N_2}$  is mass of nitrogen in a cell,  $m_{cell}$  is a total mass in a cell,  $x_{N_2}$   
 348 is a mass fraction of nitrogen in a cell,  $m_{f_l}$  is a liquid mass of iso-octane in a cell,  $V_{cell}$  is a volume of a cell,  $\alpha_p$  is a  
 349 volume fraction of parcel in a cell, and  $\rho_f$  is a liquid fuel density. Even though the simulations and experiments are  
 350 for mixing of fuel with nitrogen, we convert to equivalence ratio as if the mixture were pure air to provide a more  
 351 intuitive sense of the mixture using equation (5) where  $\varphi$  is an equivalence ratio,  $F$  is a fuel mass in a cell,  $A$  is a

352 nitrogen mass in a cell and  $(F/A)_s$  is stoichiometric fuel/air ratio by mass for iso-octane and pure air (value of 0.0661  
353 was used). In fact, the equilibrium calculations shown in Fig. 8 are also for nitrogen-fuel mixtures, but are presented  
354 as equivalence ratio also using equation (5).

355 Fig. 9 shows very local equivalence ratios that exceed a value of 8 even after the spray becomes dilute after the end  
356 of injection. According to the thermodynamic analysis given in Fig. 8, these mixtures must have liquid even at  
357 equilibrium. Therefore, it is expected that liquid persists. However, the simulation may belie this conclusion if the  
358 degree of stratification is unnatural and sensitive to modeled spray parameters. To gain a better understanding of the  
359 mean mixture prediction, a quantitative comparison of the liquid volume fraction and equivalence ratio is presented  
360 in Fig. 10, where the average of these data cutting through the center of all 8 plumes is plotted versus radial position.  
361 The measured liquid volume fraction after tomographic reconstruction (and also the average of 8 plumes) is presented  
362 as a solid line. The averaged liquid volume fraction curve derived from simulation was a bit noisy because of uneven  
363 parcel distribution in the spray so binned-pixel average (10 by 10) value was used for presentation. Fig. 10(a) shows  
364 that the liquid volume fraction from the simulation is higher and has a wider distribution compared to the experimental  
365 data. While the assumed droplet size creates some uncertainties in the experimental LVF data, the overall trend is that  
366 the simulation predicts less evaporation compared to the experiment. It is also obvious that the spray is rich enough  
367 to have liquid and vapor mixture during injection.

368 Simulation results at 1.2ms aSOI (Fig. 10(b)) show a mismatch in the plume liquid center and width compared to the  
369 experiment, with the experiment showing more of a plume collapse towards the injector axis compared to the  
370 simulation. This plume movement throughout injection will be discussed in more detail later. The mean (and spatial  
371 average of all plumes) equivalence ratio is predicted to be less than two. If the system were truly mixed and at  
372 equilibrium, there should be no liquid. Since liquid is measured and predicted in the simulation, there is high  
373 confidence that mixture stratifications exist and that vaporization is not dictated by mixing in the mean sense.

### 374 **3.2. Effects of ambient condition on plume direction**

375 The availability of three-dimensional LVF data, created by tomographic reconstruction of line-of-sight PLV data,  
376 enables detailed analysis of the radial and axial movement of individual plumes. Figure 10 showed a snapshot of the  
377 LVF data for the G2 condition at only two timings, but the overall motion of the plumes for other axial positions and  
378 timings is available for analysis. Outlines of the liquid spray with a LVF threshold of  $0.5 \cdot 10^{-3}$  are presented along the

379 Y-Z cut plane in Fig. 11 for iso-octane at G2, G3, and G3HT conditions. It is emphasized that the threshold of  $0.5 \cdot 10^{-3}$   
380 corresponds to LVF and should not be confused with the threshold of  $0.2 \cdot 10^{-3} \text{ mm}^3(\text{liquid})/\text{mm}^2$  set for PLV in  
381 Fig. 7. Error analysis showed that more than 100 injections were sufficient to acquire converged data (no variation in  
382 plume direction angle and LVF according to number of injection) with low noise. The contours are given at several  
383 timings aSOI, but full movies of the same data are available to view at [41]. The nozzle drill angle is indicated as a  
384 grey dotted line that forms a  $37^\circ$  from the injector axis (refer Fig. 1 for injector Z-axis). The macroscopic spray  
385 characteristics discussed using PLV results in the last section are consistent with the planar data cutting directly  
386 through plume centers (Y-Z plane cuts through centers of Plumes 3 & 7). For example, the faster evaporation for  
387 G3HT and longer axial liquid penetration for G2 conditions is easily shown along the plane. What is more clearly  
388 manifested in cut plane data, however, is the G2 condition plumes deflect more to axis centerline. Plumes at all  
389 conditions contract towards the centerline with increasing time aSOI, departing from the hole drill angle, but the  
390 deflection is higher for G2 conditions. One effect of the plume direction contracting in time, even prior to the end of  
391 injection, is that plumes show a curved or bent shape, retaining a ‘memory’ of the earlier plume direction but also  
392 contracting towards the injector axis.

393 A quantitative measurement of the plume direction angle is provided by defining the plume center as that with near-  
394 maximum LVF. The maximum is a justified choice because it removes artifacts created by plumes that are not  
395 orthogonal to an axial plane, as is the case for Spray G [36]. For example, rotated ideal-distribution (Gaussian) plumes  
396 defined with center at full-width at half maximum have a bias towards a larger plume direction, and one that does not  
397 correspond to the actual center. We therefore define plume center as the mean of 99% peak LVF positions to eliminate  
398 this bias, and compute a plume direction back to the hole origin from this center. Results are shown in Fig. 12 for data  
399 analyzed at  $Z = 30\text{mm}$  from the nozzle tip. As the plume arrives at 30 mm, the plume direction angle is already  
400 approximately  $3^\circ$  smaller as compared to the drill angle regardless of ambient conditions. The plume direction angle  
401 of G3 and G3HT are more or less the same, while it contracts by several degrees for the G2 condition during injection,  
402 and more so after the end of injection—results that are consistent with data rendered on a plane in Fig. 11. These  
403 results show a greater degree of plume movement towards the centerline, but not complete spray collapse at G2 (flash-  
404 boiling) conditions. The Mie-scattering images presented at the top of the figure show greater plume growth and more  
405 plume-to-plume interaction with smaller area between plumes at G2 conditions. The quantitative measurement of  
406 plume cone angle will be discussed in Section 4.

407 Adding understanding of the plume dynamics between conditions, we show LVF values on the  $z=30\text{mm}$  X-Y plane  
408 and the average through the center of all eight spray plumes as a function of radial position in Fig. 13. A more narrow  
409 spacing between plumes is shown in the X-Y plane at G2 conditions. G3 and G3HT conditions also show some LVF  
410 between plumes, but at lower intensity. The averaged LVF radial profiles given in Fig 13 (b) show that peak LVF  
411 during injection (solid lines) is only slightly less for G2 conditions compared to G3 conditions but there are greater  
412 differences after the end of injection (dashed lines), once more confirming a faster final vaporization for G2 conditions.  
413 The LVF radial shift towards the centerline for the G2 condition is pronounced during and after injection. As part of  
414 the general collapse of the spray, plumes clearly merge together more rapidly for the G2 condition with individual  
415 plume structure between plumes nearly absent by 1.07 ms aSOI. The LVF peak at G2 conditions also translates  
416 approximately 4mm from that at 0.54 ms aSOI, compared to only 1-2 mm shift for G3 or G3HT conditions. While  
417 lacking a complete collapse and mixing to the injector axis, all indications are that plume collapse is more severe for  
418 the G2 condition than other cases. Recall that G2 and G3HT condition did not show noticeable difference in spray  
419 liquid width in Fig. 7(d), movement in plume center under G2 condition is considered as substantial change. This  
420 result suggests that plume contraction had a dominant role for smaller spray liquid width for G2 condition, on the  
421 other hand, similar result was derived by faster evaporation under G3HT condition as it shows much lower peak of  
422 LVF than G2 condition.

### 423 3.3. Effect of fuel on macroscopic spray characteristics

424 Experiments were performed for the entire Table 4 test matrix, but for brevity, we show the effect of fuel type on  
425 macroscopic spray properties only at the G2 condition. At this experimental condition, the pressure ratios ( $P_a/P_v$ ) are  
426 0.17, 0.29, 0.67, and 0.71 for e30, olefinic, di-isobutylene, and iso-octane, respectively. Using the same definitions  
427 for liquid and vapor penetration used above (see Fig. 7), a comparison between fuels is provided in Fig. 14. Generally,  
428 the overall spray morphology of iso-octane and the di-isobutylene blend is similar to each other since they showed  
429 minor differences in fuel properties. The statistical data in Figs 14 (c) and (d) showed slightly longer liquid and vapor  
430 penetration lengths for di-isobutylene but the difference is still minor. However, distinct differences in fuel spray  
431 characteristics are found for olefinic and e30 blends. The e30 blend shows a shorter penetration length and a narrower  
432 liquid width at the beginning of injection, accompanied by wider radial dispersion at 15 to 25 mm axial positions. But  
433 e30 eventually overtakes the other fuels in axial liquid and vapor penetration after 1.5ms aSOI. The olefinic fuel is

434 somewhat intermediate to e30 and the lighter fuels in terms of maximum axial liquid penetration, and in the contour  
435 of PLV plot that shows a clustering of liquid towards the center. The olefinic and e30 blends have significant times  
436 for persistence of liquid, to 4.9ms and over 5ms, respectively, despite heated fuel (90°C) as an injection boundary  
437 condition. The slower evaporation of e30 is entirely expected because of the extensive vaporization cooling associated  
438 with its heat of vaporization compared to other fuels, as well as the presence of heavy species in the multi-component  
439 blend compared to single-component surrogates with lower distillation temperature. However, we will show below  
440 that e30 vaporization is also affected by impaired mixing because of plume collapse.

441 The vapor envelope measurements indicate the same general shape but the olefinic and e30 fuels tended to have a  
442 more narrow tail in vapor distribution after the end of injection. Using the three-dimensional LVF data, we will show  
443 below that these changes in vapor distribution are the result of significant spray collapse for olefinic and e30 fuels.

#### 444 **3.4. Effect of fuel on plume direction**

445 As shown above, the availability of three-dimensional LVF data provides a much better understanding of spray  
446 dynamics. A comparison of all fuels at the G2 condition is shown in the Y-Z plane Fig. 15 (a), using the same threshold  
447 used for iso-octane ( $0.5 \cdot 10^{-3}$ ) at all experimental conditions (in Fig. 11). While plumes move towards the centerline  
448 for iso-octane and di-isobutylene fuels, liquid moves all of the way to center for e30, and to a large degree for the  
449 olefinic fuel. For e30, individual plume identification is almost immediately lost after start of injection, while there is  
450 clearly evidence of plumes connected at the centerline at  $z = 20\text{mm}$  for the olefinic fuel by 0.5 ms aSOI as shown in  
451 Fig. 15 (b). The strong collapse to a single jet structure (for e30), produces a repeatable head vortex structure in the  
452 leading edge of the spray, such that it appears even in the ensemble-average. While some plume bend is observed for  
453 other fuels, including the olefinic fuel, the head-vortex structure is not as prominent.

454 The LVF boundary at 1.26 ms aSOI highlights the strong differences in liquid distribution for each fuel. While iso-  
455 octane and di-isobutylene have evaporated to lower LVF concentrations by the timing, the olefinic and e30 blends  
456 show significant liquid remaining near the centerline. One reason for the persistence of liquid is the high boiling point  
457 constituents, and high heat of vaporization (e30), but a second contributing factor is that fuel plumes are clustered  
458 together at the center and experiencing poor mixing. A complete merging of plumes (spray collapse for e30) creates  
459 a merged jet with cumulative momentum higher than that of individual plumes, and this can also explain the greater  
460 axial penetration for e30, along with greater potential for liquid impingement on engine wall surfaces. The significant

461 departure of liquid from the original drill angle projection (dashed line) is an indication of how different the fuel  
462 delivery becomes under spray collapsing scenarios. We emphasize that the 3D diagnostic for local LVF provides this  
463 level of detail and new understanding.

464 Using the 3D LVF data, a quantitative comparison of plume direction angle for all fuels is also provided in Fig. 16,  
465 using the same definitions as that used in Fig. 12 for the  $z=30$  mm position. The trend for a decrease in plume direction  
466 angle with time is exactly consistent with the pressure ratio ( $P_a/P_v$ ) for each fuel. While appearing nearly identical, the  
467 measured plume direction angle of di-isobutylene is approximately  $1^\circ$  smaller than that of iso-octane, a result that is  
468 within the resolution of the optical setup and statistically significant because of the well-converged average data. This  
469 result shows that the di-isobutylene had slightly more plume collapse than iso-octane, potentially facilitated by the  
470 fast evaporation of the light component (4% 1-hexene) in the blend, even though at low concentration. On the other  
471 hand, the olefinic and e30 blends have massive differences in plume direction, consistently smaller than the drill angle  
472 even from the time of first arrival at  $z=30$  mm. The smaller initial plume direction angle indicates the spray contraction  
473 by plume collapse is well underway even at early stages of injection, consistent with Fig. 15. The plume direction  
474 angle for these fuels abruptly decreases and is not measurable once plumes combine and collapse.

475 The fuel effect on plume collapse is illustrated with even more detail by considering results along the X-Y plane in  
476 Fig. 17, with analogy in presentation to Fig. 13. Fig. 17(a) shows significant plume-to-plume interaction for the  
477 olefinic and e30 blends during injection at 0.54 ms aSOI, illustrating a symptom of spray collapse. A close look also  
478 shows higher LVF between plumes for di-isobutylene compared to iso-octane, potentially indicating slightly more  
479 plume-to-plume interaction leading to the subtle decrease in plume direction for di-isobutylene compared to iso-  
480 octane. The LVF distributions across plumes presented in Fig. 17(b) shows wider plumes for the olefinic and e30  
481 blends compared to the lighter fuels at 0.54 ms, indicating distortion and stretch towards the center. Plume direction  
482 (at maximum LVF) is discernable at this time for the olefinic and e30 blends, but at 1.07 ms the peak LVF is at the  
483 centerline, indicative of full spray collapse and undefined plume direction for these fuels (see Fig. 16). Interestingly,  
484 the peak LVF is actually nearly the same or higher, despite the fact that 1.07 ms is after the end of injection and there  
485 is an expectation for further dilution with time. This rising LVF in time can only be accomplished if mixing with air  
486 is impaired, as is the case for central collapse for all plumes. By contrast, at 1.07 ms LVF decreases quickly and  
487 significantly for iso-octane and di-isobutylene, assisted by ambient gas surrounding each of the eight separated plumes.

488 A comparison of the overall spray processes between iso-octane and e30 blends is exhibited in Fig. 18. Mie-scattering  
489 images are shown at the top and the 3D LVF maps rendered in transparency are given at the bottom. The Mie-scattering  
490 images from the iso-octane indicate light plume-to-plume interaction, not severe enough to induce complete collapse.  
491 On the other hand, the e30 blend suffers from strong plume collapse from the beginning, so that each plume could not  
492 be identified. At 0.1ms aSOI, e30 shows specific patterns in Mie-scattering images, but these patterns are not created  
493 by individual plumes. Rather, the strongest signature is at the position bisecting the plume position, as if the injector  
494 were rotated. This observation is another indication of strong plume-to-plume interaction. The Mie-scatter images and  
495 3D LVF images show significant liquid at the jet centerline by 0.5ms aSOI, consistent with Fig. 15. The complete  
496 spray collapse to the center and rollup of the head vortex for e30 are illustrated dynamically in the 3D LVF images  
497 (and movie available at [41]).

#### 498 **4. Plume vapor cone angle**

499 A potential hypothesis to explain the fuel effects demonstrated in this paper is that fuel sprays with low ( $P_a/P_v$ ) have  
500 significant vapor formation from flash boiling that tends to enlarge individual plume cone angle. With larger plume  
501 cone angle, interaction between plumes is accelerated, air entrainment is shut off, and spray collapse occurs. To  
502 substantiate this hypothesis, it is necessary to have a direct assessment of plume cone angle. While radial LVF  
503 distributions may be used to understand liquid plume growth, the width of the liquid region may not be representative  
504 of plume growth because of confounding factors such as vaporization dependencies upon distillation properties as  
505 well as ambient conditions. These confounding factors are overcome if able to measure the plume vapor cone angle  
506 corresponding to the lowest fuel mixture fraction.

507 To accomplish this goal, we use the vapor boundary from line-of-sight schlieren imaging, in concert with the plume  
508 center measured from 3D tomographic reconstruction, to obtain the plume vapor cone angle. The process is illustrated  
509 schematically in Fig. 19. While schlieren imaging only measures the outer vapor envelope of two plumes (Plumes 3  
510 and 7 in the primary  $0^\circ$  rotation view), the average plume center and average vapor envelope can be combined to  
511 approximate vapor plume cone angle. The plume vapor cone angle was measured near the nozzle at  $z=8.5$  mm to  
512 avoid distortion from the head vortex zone shown in the figure. Fig. 19 shows a schematic diagram in which the vapor  
513 boundary with 0.5 intermittency is presented as red solid line. Averaged plume centers (from 3D LVF analysis) are  
514 also indicated as triangles on the line of plume direction (dotted blue lines). A line orthogonal to the plume direction

515 angle intersects the vapor boundary and is shown with a circle. At this moment, the angle formed by two lines, circle  
516 to orifice outlet and averaged plume center to orifice outlet, can be considered as half of the plume vapor cone angle.  
517 Combining the measurement of plume center (via 3D tomographic reconstruction) and vapor envelope offers a new  
518 technique to measure plume vapor cone angle in realistic multi-plume GDI sprays. A technique to measure plume  
519 cone angle, rather than total spray angle or width, is critical because interaction and merging between plumes also  
520 affects total spray angle or width.

521 The measured plume vapor cone angle presented in Fig. 20 (a) is an average value from top and bottom plumes during  
522 200 $\mu$ s to 680 $\mu$ s aSOI for all conditions and fuels. As can be seen, the pressure ratio has a significant effect on plume  
523 cone angle, which has been observed in past single-hole studies [42]. A strong inflection occurs near a pressure ratio  
524 of 0.5. Fuels with a high portion of light species have high  $P_v$  and lower pressure ratio ( $P_a/P_v$ ), and are more susceptible  
525 to higher plume growth. As shown in earlier sections, conditions with higher plume growth also show more tendency  
526 for spray collapse, as confirmed by 3D LVF measurements, generating the hypothesis that initial plume growth is a  
527 key factor for spray collapse. With measurement of both plume direction and plume cone angle, facilitated by the 3D  
528 measurements of this study, we can investigate this hypothesis through a correlation between these two quantities.  
529 Fig. 20 (b) shows plume direction angle measured at  $z=30$ mm and 0.37ms aSOI according to averaged plume vapor  
530 cone angle presented in Fig. 20 (a). This result clearly shows that smaller plume direction angle, as a sign of more  
531 plume-to-plume interaction, is induced by a larger plume vapor cone angle over a broad range of conditions and fuels.  
532 For example, under G3 conditions the e30 blend (purple triangle) has a slightly smaller plume vapor cone angle and  
533 a corresponding larger plume direction angle than the olefinic fuel under G2 conditions (orange circle). Subtle effects  
534 such as the greater plume interaction and smaller plume direction for di-isobutylene compared to iso-octane are also  
535 consistent with lower pressure ratio and greater plume cone angle for di-isobutylene. While the conclusion and relation  
536 of greater spray growth with lower pressure ratio is consistent with past literature observations for single-hole injectors  
537 [42], or in some cases multi-hole nozzles [43], we emphasize that this study is the first to provide measurement of  
538 individual plume growth, rather than total spray angle, for realistic multi-plume GDI sprays.

## 539 **5. Conclusion**

540 In this study, a series of high-speed Mie-scattering, extinction, and schlieren imaging was carried out in a constant-  
541 flow spray vessel under G2, G3 and G3HT conditions, consistent with realistic intake-injection conditions in an

542 engine. Four different fuels: iso-octane, di-isobutylene surrogate, olefinic, and a 30% ethanol blend were injected  
543 using an ECN Spray G 8-hole injector. CFD Lagrangian parcel simulations were also performed to understand mixing  
544 and liquid residual time. We applied computed tomographic reconstruction using extinction imaging to measure the  
545 3D liquid volume fraction of all plumes, and schlieren imaging to identify the vapor boundary for some of the plumes.  
546 Measurements of individual plume dynamics were resolved in time and space, and include statistics such as plume  
547 direction angle, plume cone angle, liquid penetration, vapor penetration, and so forth. The dataset offers unique  
548 information on plume growth and movement in the midst of a realistic multi-hole spray, and are available to  
549 view/download [41]. The major findings from this study as follows.

550 1) Liquid persists under a flash-boiling G2 condition for more than 3ms even with iso-octane fuel, showing a  
551 longer liquid penetration than the non-flashing G3 condition and enough time and penetration distance to  
552 cause liquid impingement in engine. Equilibrium mixture calculations can explain the long persistence at G2  
553 conditions as liquid is expected if mixtures have an equivalence ratio greater than a value of four. At 30 mm  
554 downstream of the injector, CFD simulations predict mixtures with  $\varphi > 4$  during injection, but  $\varphi < 4$  at  
555 0.4 ms after the end of injection, even though liquid is measured and predicted at this same time. These data  
556 support the notion that the system is not mixing-limited or at equilibrium. Fuels containing heavy species  
557 such as olefinic and e30 showed even longer liquid residual time, up to the end of the experimental acquisition  
558 at 5ms, which is likely to cause piston/cylinder wetting.

559 2) Using tomographically reconstructed liquid volume fraction data centered upon a plume, a noticeable plume  
560 movement towards the injector axis (decrease in plume direction angle) occurs under G2 conditions. The iso-  
561 octane spray under G2 conditions shows an approximately  $6^\circ$  smaller plume direction angle at the end of  
562 injection ( $z=30\text{mm}$ ) as compared to the drill angle ( $37^\circ$ ). For all conditions and fuels, an analysis of measured  
563 plume cone angle versus plume direction angle shows excellent correlation for a smaller plume direction  
564 angle when there is a larger plume cone angle. The logical conclusion is that a larger plume cone angle  
565 enhances aerodynamic interaction between plumes, pulling plumes closer to one another and reducing plume  
566 direction angle relative to the injector axis. For larger plume cone angles, the plumes may combine to undergo  
567 complete spray collapse.

- 568 3) Plume vapor cone angle measurements were made possible in a multi-plume spray by a combination of 3D  
569 liquid distribution measurements to find the plume center, combined with schlieren imaging to find the vapor  
570 boundary for two of the plumes. The plume cone angle with various fuels and conditions shows a strong  
571 inflection at a pressure ratio of 0.5 ( $P_a/P_v$ ), or where the ambient pressure is half of the vapor pressure. The  
572 plume vapor cone angle increases sharply at a pressure ratio less than 0.5.
- 573 4) Strong plume interaction and collapse may leave high liquid fuel concentrations in the spray core region. For  
574 a volatile 30% ethanol fuel (e30) at flash-boiling conditions, peak fuel concentration actually increases after  
575 the end of injection, as the result of plume combination and collapse during injection.

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## 677 **Appendix**

678 In this section, effects of droplet-size distribution on projected liquid volume, liquid volume fraction, and plume  
679 direction will be discussed. First, PLV was calculated according to different droplet diameters as shown in Fig. 21.  
680 An optical thickness of 0.081, which corresponds to PLV of  $0.2 \cdot 10^{-3} \text{ mm}^3(\text{liquid})/\text{mm}^2$  for  $7\mu\text{m}$  droplet diameter was  
681 held constant in the calculation. According to eq-2, and demonstrated in Fig. 21, the calculated PLV for the same  
682 optical thickness decreases nearly linearly with droplet diameter. Thus, the actual liquid envelope, defined with PLV  
683 of  $0.2 \cdot 10^{-3} \text{ mm}^3(\text{liquid})/\text{mm}^2$  would shrink if droplet diameter is smaller than  $7\mu\text{m}$ , or it would grow if droplet diameter  
684 is larger than  $7\mu\text{m}$ . We note that droplet sizing measurements for ECN Spray G show a rather flat distribution when  
685 spanning the radial position within the plume [36]. Droplet size decreases from  $10\mu\text{m}$  to about  $7\mu\text{m}$  at the edge of the  
686 plume, so our assumption of  $7\mu\text{m}$  droplet size had the best application for positions at the jet periphery. However, as  
687 droplet size is not measured for all conditions and fuels, we can consider the likely scenario of decreasing droplet size  
688 (from vaporization) at the spray periphery. For example, using the standard criteria of PLV of  $0.2 \cdot 10^{-3}$   
689  $\text{mm}^3(\text{liquid})/\text{mm}^2$ , liquid penetration and width decrease by about 5% if droplet diameter decreases from  $7\mu\text{m}$  to  $2\mu\text{m}$ ,  
690 at  $Z=15\text{mm}$  and  $0.78\text{ms}$  aSOI under G2 condition.

691 We also note the significant uncertainty associated with droplet size for prediction of LVF through tomographic  
692 reconstruction. Even though we used a value of  $7\mu\text{m}$  from direct measurements, there is some measured variation in  
693 droplet size towards larger droplet sizes (about  $10\mu\text{m}$ ) at the plume centerline. Should  $10\mu\text{m}$  be a more appropriate  
694 assumption, the LVF at the plume center would increase by 30%. However, experimental measurements to date are  
695 limited, leading to significant uncertainty were the droplet sizes to vary from our assumptions. Fig 22 indicates the  
696 effect of droplet diameter on LVF predictions with droplets as large as  $20\mu\text{m}$  or as small as  $1\mu\text{m}$  [36]. While the peak  
697 liquid volume fraction value changes depending upon the droplet size, the experimental measurements of droplet size  
698 to date do not show a significant change in droplet size with radial position that would cause an incorrect determination  
699 of the plume center, and plume direction. However, we recommend that droplet size distribution measurements be  
700 performed for as many locations as possible during the entire spray process, while working to overcome challenging  
701 limitations with respect to optically thick or optically thin (overly dilute) regions of the fuel spray in the complex  
702 vaporizing environment.