



HAL
open science

Turbulence and entrainment in an atmospheric pressure dielectric barrier plasma jet

Youssef Morabit, Richard D. Whalley, Eric Robert, Mohammad I. Hasan,
James L. Walsh

► **To cite this version:**

Youssef Morabit, Richard D. Whalley, Eric Robert, Mohammad I. Hasan, James L. Walsh. Turbulence and entrainment in an atmospheric pressure dielectric barrier plasma jet. *Plasma Processes and Polymers*, 2020, 17 (6), pp.1900217. 10.1002/ppap.201900217 . hal-02404643

HAL Id: hal-02404643

<https://hal.science/hal-02404643>

Submitted on 1 Dec 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Turbulence and entrainment in an atmospheric pressure dielectric barrier plasma jet

Journal:	<i>Plasma Processes and Polymers</i>
Manuscript ID	Draft
Wiley - Manuscript type:	Full Paper
Date Submitted by the Author:	n/a
Complete List of Authors:	<p>Morabit, Youssef; University of Liverpool, Department of Electrical Engineering & Electronics Whalley, Richard; Newcastle University, School of Mechanical and Systems Engineering robert, eric; CNRS/Univ Orleans, UMR 6606 GREMI Hasan, Mohammad; University of Liverpool, Department of Electrical Engineering & Electronics Walsh, James; University of Liverpool, Department of Electrical Engineering & Electronics</p>
Keywords:	

SCHOLARONE™
Manuscripts

Manuscript number DOI:10.1002/ppap.

Article type: Full Paper

Turbulence and entrainment in an atmospheric pressure dielectric barrier plasma jet.

Y. Morabit¹, R. D. Whalley², E. Robert³, M. I. Hasan¹ & J. L. Walsh^{1*}

¹ Centre for Plasma Microbiology, Department of Electrical Engineering & Electronics, University of Liverpool, L69 3GJ, UK.

² School of Mechanical and Systems Engineering, Newcastle University, NE1 7RU, UK.

³ GREMI UMR 7344 CNRS/Université d'Orléans, Orléans, 45067, France.

Abstract

Particle Imaging Velocimetry, Laser Induced Fluorescence and computational modeling are used to quantify the impact of plasma generation on air entrainment into a helium plasma jet. It is demonstrated that discharge generation yields a minor increase in the exit velocity of the gas. In contrast, the laminar to turbulent transition point is strongly affected, attributed to an increase in plasma-induced perturbations within the jet shear layer. The temporal decay of laser induced fluorescence from OH is used as an indicator for humid air within the plasma. The results show that plasma-induced perturbations increase the quenching rate of the OH fluorescent state; indicating shear layer instabilities play a major role in determining the physicochemical characteristics of the plasma.

1 **1 Introduction**

2 Atmospheric pressure plasma jets have been widely used in many healthcare and materials
3 processing applications, ranging from etching and deposition to microbial decontamination and
4 cancer therapy.^[1-5] Perhaps the most widely used plasma jet configuration is based on the dielectric
5 barrier discharge employing a noble gas such as helium or argon; typically, the gas is flushed
6 through a dielectric capillary and subjected to an applied voltage using one or more electrodes
7 placed inside and/or outside of the capillary. On application of a time-varying voltage of sufficient
8 magnitude to cause breakdown, a discharge forms within the capillary and propagates as a fast-
9 moving ionization wave along the noble gas channel, ultimately exiting the capillary and extending
10 into the surrounding quiescent air. From an application perspective, the interaction between the
11 propagating plasma plume and the surrounding air is of great importance as many atmospheric
12 pressure plasma-based applications rely on reactive oxygen and nitrogen species (RONS), many of
13 which are created when the noble gas plasma interacts with the surrounding air.

14
15 A large number of studies have considered the impact that plasma generation has on the
16 characteristics of the flowing noble gas. It has been widely reported that the generation of plasma in
17 a buoyant axisymmetric jet configuration leads to a rapid transition from laminar to turbulent
18 flow.^[6-9] It is commonly assumed that gas heating and Electrohydrodynamic (EHD) forces play a
19 role in creating turbulence within the flowing gas channel, with the latter being considered as the
20 dominant mechanism. Indeed, Park et al. used a pulsed plasma jet to demonstrate that EHD forces
21 are primarily exerted by space charge drifting in the applied electric field following streamer
22 propagation.^[10, 11] Whalley and Walsh demonstrated that the spatially developing velocity fields in
23 an inhomogeneous axisymmetric plasma jet flow are turbulent and self-similar, with characteristics
24 matching the turbulent velocity fields which develop naturally with increasing distance from the jet
25 exit. The study posited that the production of turbulence is related to the small-amplitude plasma-
26 induced body forces causing perturbations in the unstable shear layers at the jet exit, which grow as

27 they move downstream, creating turbulence.^[12, 13] Using an order-of-magnitude analysis, it was
28 predicted that the presence of a discharge should only increase the jet exit velocity by
29 approximately 10%.^[8]

30

31 To characterize the chemical species produced downstream of the jet orifice, multiple invasive and
32 non-invasive diagnostic techniques have been adopted, including tunable diode laser absorption
33 spectroscopy (TDLAS),^[14] mass spectroscopy (MS),^[15] Fourier transform infrared spectroscopy
34 (FTIR),^[16] Laser-Induced Fluorescence (LIF) and Two-photon Absorption Laser-Induced
35 Fluorescence (TALIF).^[17-19] Of all the techniques investigated, LIF based methods are particularly
36 convenient as they provide the high spatiotemporal resolution required to accurately investigate the
37 complex downstream chemistry in a plasma jet while facilitating the quantification of absolute
38 radical densities. Regardless of the diagnostic technique used, it is clear that a significant proportion
39 of the reactive chemical species produced within the downstream region of the jet are a result of
40 reactions between the noble gas plasma and molecular gas impurities entrained from the
41 surrounding environment, which is typically humid air. Given that the interplay between the
42 propagating plasma plume and ambient environment has a considerable impact on the
43 physicochemical properties of the plasma jet and, therefore, on the application efficacy,
44 understanding the underpinning mechanisms of how air becomes entrained within the flowing noble
45 gas channel is of vital importance.

46

47 This study investigates the entrainment of air into a helium plasma jet and explores the hypothesis
48 that small perturbations within the jet shear layer resulting from plasma generation not only result in
49 the early onset of turbulence but also act to increase entrainment of air into the laminar region of the
50 jet. Particle imaging velocimetry (PIV) was used to quantify the fluid dynamics of the helium jet
51 flow beyond the jet orifice and combined with LIF to provide a sensitive means of assessing the
52 entrainment of humid air into the discharge.

53

54 **2 Experimental and computation methods**

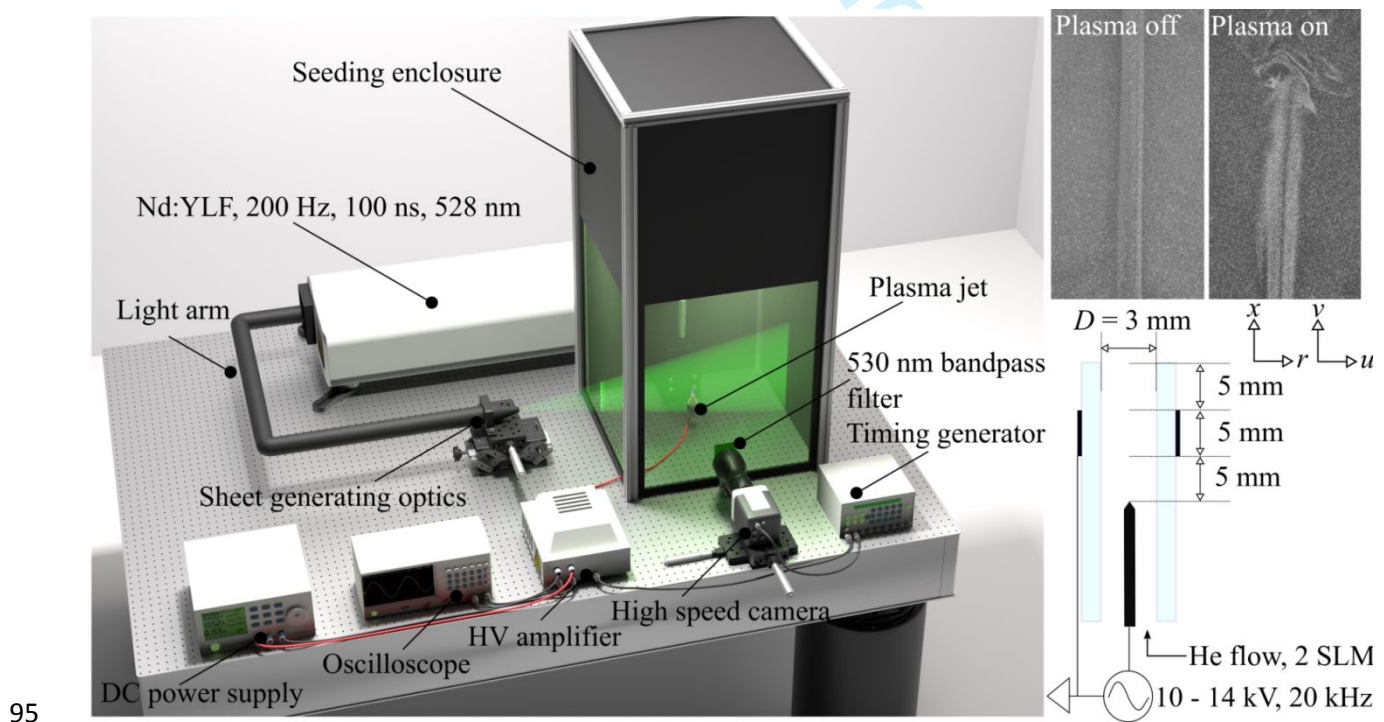
55 **2.1 Plasma Jet and PIV setup**

56 The plasma jet used in this study comprised of a quartz capillary with an inner diameter D , equal to
57 3 mm and outer diameter of 5 mm, a metallic strip was wrapped around the outer diameter of the
58 capillary to act as a ground electrode, as shown in **Figure 1**. A tungsten pin was positioned
59 coaxially within the quartz capillary and connected to a high-voltage sinusoidal power operating at
60 a frequency of 20 kHz. Two applied voltage cases were examined in this study, 14 and 10 kVpp,
61 with the latter being just above the point of gas breakdown. Helium gas with a purity of 99.999%
62 flowed through the capillary at 2 Standard Litres per Minute (SLM), giving a maximum exit
63 velocity, U_m , equal to 9.4 m s^{-1} .

64

65 To quantify the flow field created by the plasma jet, PIV measurements were undertaken using the
66 experimental setup shown in Figure 1. The plasma jet was inserted into a large sealed chamber
67 (volume $> 2 \text{ m}^3$), which was seeded using oil droplets with a nominal size of $1 \mu\text{m}$. The chamber
68 ensured that the plasma jet flow was not influenced by any external draughts. A similar
69 experimental arrangement was used in the previous works of the authors,^[8] where it was observed
70 that an insufficient number of seeding particles were entrained within the laminar region of the jet
71 to obtain reliable PIV measurements. To overcome this challenge, the helium flow into the jet
72 capillary was seeded using oil droplets, which also had a nominal size of $1 \mu\text{m}$. The addition of oil
73 droplets to the helium gas flow has an obvious potential to disrupt the discharge; while such
74 changes are difficult to assess, the breakdown voltage and length of plasma plume were found not
75 to change significantly. Furthermore, Rayleigh scattering was used to obtain an approximate
76 indication of the laminar region length in an un-seeded jet and a close agreement was observed
77 when compared to the PIV measurements on the seeded jet, suggesting the presence of the seeding
78 particles had a minimal impact on the fluid dynamic properties of the system. Given the nominal

79 size of the seeding particles, the Stokes number was found to be less than 0.1, thus ensuring that the
 80 particles followed the fluid flow closely with tracing errors being $< 1\%$.^[20]
 81
 82 Planar velocity measurements were conducted using a 2D particle image velocimetry system from
 83 TSI inc. The system consisted of a double pulsed Nd:YLF laser operating at 200 Hz with a pulse
 84 duration of 100 ns at a wavelength of 527 nm and was used to generate a 1 mm thick light sheet that
 85 was projected into the seeding chamber and across the plasma jet orifice. A high-speed Phantom
 86 Miro Lab 340 camera was positioned outside the seeding chamber normal to the laser sheet and
 87 synchronized with the laser such that each frame captured a single laser pulse. A spatial calibration
 88 was performed and the time delay between consecutive laser pulses (Δt) was set to 30 μs , a value
 89 chosen to capture the movement of oil droplets over a grid with spatial dimensions of 350 μm^2 ,
 90 enabling the velocity vectors to be computed using a recursive cross-correlation technique. For each
 91 measurement condition, the plasma jet was operated for several seconds before data capture to
 92 ensure steady-state conditions were achieved. Each dataset comprised of 800 frames that were used
 93 to make 400 individual velocity vector maps; in the case of time-averaged measurements, all 400
 94 vector maps were averaged and presented as a single figure.



96 **Figure 1.** Diagram showing the layout of the particle imaging velocimetry experiment and
97 configuration of the plasma jet device, with representative PIV images for the plasma off and
98 plasma on cases, vectors v and u represent the axial and radial velocity components, respectively.
99

100 2.2 LIF setup and OH density calibration

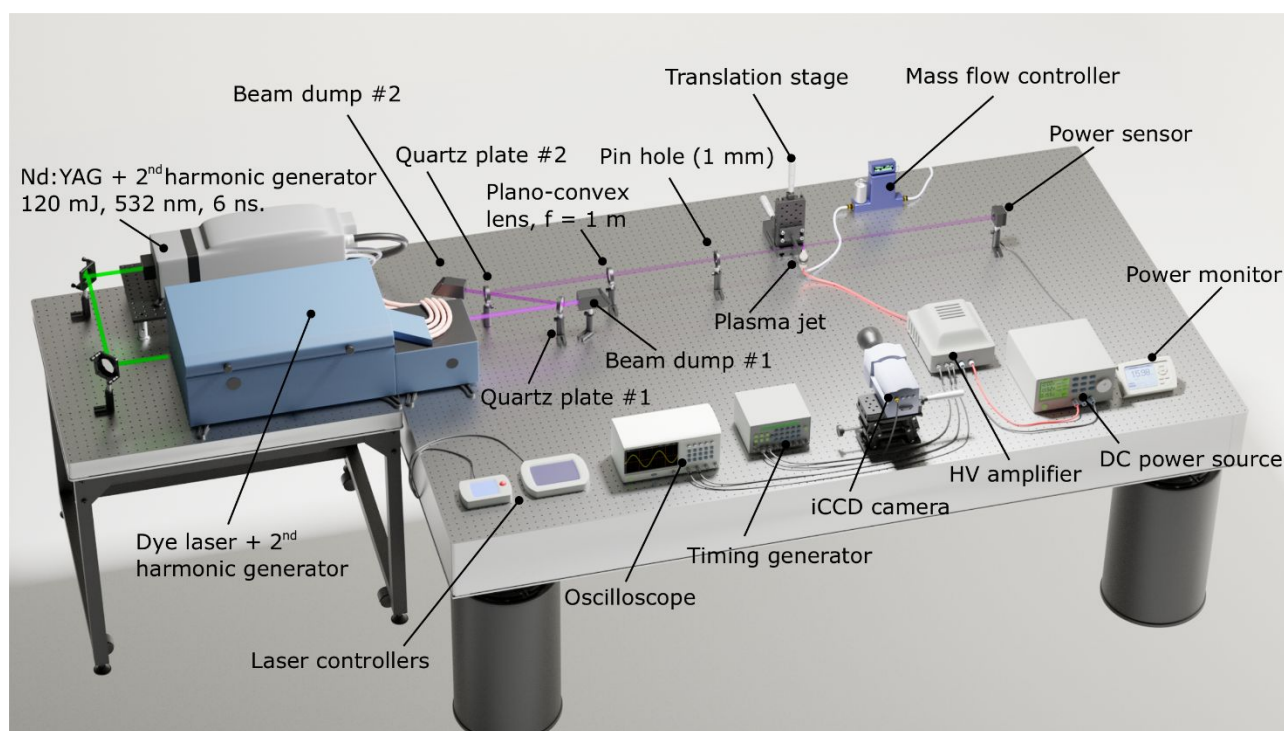
101 The LIF measurement system used in the investigation is shown in **Figure 2**, the system was used
102 to measure OH radical density and to obtain an indication of air entrainment within the plasma
103 plume through the measurement of the radiative decay time of the laser-excited OH(A) state. The
104 system comprised of a tunable dye laser (Sirah Cobra Stretch with second harmonic generation unit)
105 pumped by a 6 ns pulsed Nd:YAG laser with a wavelength of 532 nm, pulse energy of 120 mJ and a
106 repetition rate of 10 Hz. The dye laser was tuned to generate an output at a wavelength 282.58 nm
107 to excite the $P_1(2)$ transition from the $f_1(2)$ rotational level of OH X($v'' = 0$) to the $F_1(1)$ rotational
108 level of OH A($v' = 1$). Numerous other studies have used a similar excitation scheme due to the
109 $P_1(2)$ transition being spectrally separated from adjacent lines and the $f_1(2)$ level having the largest
110 relative population in the temperature range under investigation.^[21]

111
112 The 282.58 nm beam emitted from the dye laser was measured to have a pulse energy in excess of
113 17 mJ, a value several orders of magnitude above the range linear LIF measurements are typically
114 made (1 – 10 μ J).^[22] Operation beyond the linear region greatly complicates the interpretation of the
115 results, as the ground rotational level is significantly depleted by light absorption and partially
116 refilled by fast rotational redistribution, altering the LIF outcome dependent on the unknown gas
117 composition and temperature. To attenuate the laser energy to a suitable range an optical
118 arrangement similar to that employed by Ries et al. was adopted,^[17] two quartz plates were angled
119 to split the beam as shown in Figure 2, with a small fraction being reflected towards the plasma jet
120 and the majority of the beam passing through to beam dumps mounted behind each plate. Following
121 attenuation, the beam was directed through an uncoated quartz plano-convex lens with a focal

122 length of 1 m, and a pinhole of 1 mm was positioned to act as a spatial filter, further attenuating the
123 beam. Using this approach the maximum pulse energy was found to be approximately 15 μJ ; small
124 changes to the Q-switch delay of the pump laser were subsequently used to vary the pulse energy
125 between 1 and 15 μJ . Laser power was measured using a Thorlabs PM100D optical power and
126 energy meter equipped with a thermal volume absorber power sensor.

127

128 To capture the fluorescence of excited OH molecules, an Andor iStar740 iCCD camera was fitted
129 with a Jenoptik UV 105 mm f/4.5 imaging lens. The camera arrangement was positioned to face the
130 plasma jet perpendicular to the laser beam. For each image recorded, the iCCD camera was
131 configured to accumulatively capture 500 laser pulses, using an optical gate width equal to 8 ns for
132 each exposure. Following the approach of Verreycken et al.,^[21] no bandpass filter was used during
133 LIF measurements to avoid the need for additional corrections in the calibration procedure. A
134 consequence of this approach is the potential for interference of the fluorescent signal from other
135 emissions within the plasma and the Rayleigh scattering signal. A background subtraction of the
136 emission captured with the plasma energized and laser de-tuned was used to correct for light
137 emitted by the plasma and any scattered light. To ensure the measurements were conducted within
138 the linear LIF region, the measured LIF intensity was plotted against laser energy and a linear fit
139 applied. It was determined that the linear range was between 1 μJ to 10 μJ , a value in close
140 agreement with several past LIF investigations.^[17, 21]



141

142 **Figure 2.** Diagram showing the layout of the Laser-Induced Fluorescence experiment.

143

144 Absolute calibration of the LIF signal to determine the density of ground-state OH can be achieved
145 via several methods, including UV absorption, chemical modeling, and Rayleigh scattering.^[21, 23] In
146 this investigation, the Rayleigh scattering approach was adopted due to its high degree of accuracy,
147 and a similar methodology to that described by Verreycken et al. was adopted.^[21] Calibration by
148 Rayleigh scattering requires detailed knowledge of the rotational and vibrational energy transfer
149 rates, which vary significantly depending on the nature of the quenchers present; thus an accurate
150 appreciation of the gas composition is essential. Careful consideration must be applied in the case of
151 a plasma jet as the gas composition varies as a function of distance from the jet orifice; further
152 details on this are provided in section 2.3. In addition to the gas composition, the gas temperature
153 can also affect the interpretation of the LIF data. To investigate the influence of plasma generation
154 on gas temperature, an Omega FOB100 fiber-optic thermometer was used, the dielectric
155 temperature probe was positioned in the plasma at various points downstream of the jet orifice and
156 the temperature recorded. The temperature was found to vary little with spatial position, with a

157 maximum of 10 K above ambient located close to the capillary orifice, such observations are in-line
 158 with previous studies.^[8]

159 The remainder of the calibration process closely followed that reported previously by Verreycken et
 160 al. and will only be summarised in brief here.^[21] To obtain Rayleigh scattering data for calibration,
 161 the plasma jet was de-energized and supplied with Nitrogen gas at a flow rate of 2 SLM, while not
 162 strictly necessary, the nitrogen flow helped to ensure dust from the ambient environment did not
 163 enter the measurement region of interest. The laser power was varied from 2 to 15 μJ in 1 μJ
 164 increments, and the sum of the Rayleigh scattered signal intensity in a rectangular region of interest
 165 measuring 0.36 x 0.36 mm was calculated. The measured Rayleigh scattered signal, S_{Ray} (#counts),
 166 can be written as :

167

$$168 \quad S_{Ray} = \eta N_n \frac{\partial^{\beta=0} \sigma_0}{\partial \Omega} V_{Ray} I_L t_L \quad (1)$$

169

170 Where η is the calibration constant (#counts sr J^{-1}), N_n is the density of scattering particles (m^{-3}),
 171 $\partial^{\beta=0} \sigma_0 / \partial \Omega$ is the differential cross-section for Rayleigh scattering ($\text{m}^2 \text{sr}^{-1}$), V_{Ray} is the volume from
 172 which Rayleigh scattering is collected (m^3), I_L is the laser irradiance (W m^{-2}) and t_L is the temporal
 173 length of the laser pulse (s), which was measured by replacing the power meter shown in Figure 2
 174 with a fast photodetector. After taking into account the non-uniformity of the laser energy density,
 175 Equation (1) becomes:

176

$$177 \quad S_{Ray} = \eta N_n \frac{\partial^{\beta=0} \sigma_0}{\partial \Omega} E_L \Delta x \quad (2)$$

178

179 Where E_L is the laser energy (J), and Δx is the length of the detection volume (m). Following this,
 180 the calibration constant η was obtained from the slope α , of the measured Rayleigh intensity as a
 181 function of laser energy multiplied by pressure:

182

$$\eta = \alpha k_B T \frac{\partial^{\beta=0} \sigma_0}{\partial \Omega} \Delta x \quad (3)$$

183

184

185 Using the calibration factor, the intensity of the laser-induced fluorescence S_{LIF} , can be expressed

186 as:

187

$$S_{LIF} = \frac{1}{4\pi} \int \eta E n_{exc}(x, y, z, t) A \, dx dy dz dt \quad (4)$$

189

190 Where E is the energy gap according to the chosen transition, A is the Einstein emission coefficient
 191 (s^{-1}), and $n_{exc}(x, y, z, t)$ is the density of OH in the excited state. To determine the ground state OH
 192 density based on n_{exc} in Equation (4), the 4-level collisional radiative model reported Verreycken et
 193 al. was utilized.^[21] The model accounted for the variation in gas composition beyond the jet orifice,
 194 as described in the following section.

195

196 2.3 Computational model and statistical analysis methodology

197 To account for the varying composition of gas downstream of the jet orifice, a computational model
 198 was developed that solved for the velocity field of the gas mixture in addition to the mass fractions
 199 of its constituents, namely N_2 , O_2 , H_2O , and He. To obtain the velocity field, the model solved the
 200 mass continuity Equation (5) and the momentum conservation Equation (6). To compute the gas
 201 composition, the mass continuity equation, given by Equation (7), was solved for three species,
 202 while the mass fraction of the fourth was determined from the pressure constraint. All equations
 203 were solved in steady-state mode (*i.e.*, time-independent equations) :

204

$$\nabla \cdot (\rho \mathbf{u}) = 0 \quad (5)$$

205

$$\rho (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \nabla \cdot \left(\mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) - \frac{2}{3} \mu (\nabla \cdot \mathbf{u}) \mathbf{I} \right) - (\rho - \rho_0) \mathbf{g} \quad (6)$$

206

$$\rho (\mathbf{u} \cdot \nabla) \omega_i + \nabla \cdot (\Gamma_i) = 0 \quad (7)$$

207 Where ρ is the density of the gas mixture (kg m^{-3}), \mathbf{u} is the velocity field of the gas mixture (m s^{-1}),
 208 p is the gas mixture's pressure (Pa), μ is the gas mixture's viscosity (Pa s), I is the identity matrix,
 209 ρ_0 is the density of air (kg m^{-3}), and g is the gravitational constant (m s^{-2}), ω_i is the mass fraction of
 210 the i^{th} species, and Γ_i is the diffusive flux of the i^{th} species, which is calculated according to
 211 Maxwell-Stefan theory for diffusion as given by Equation (8-10).^[24, 25]

$$212 \quad \Gamma_i = \rho \omega_i \mathbf{V}_i \quad (8)$$

$$213 \quad \nabla x_i = \sum_{j=1}^4 \frac{x_i x_j}{D_{ij}} (\mathbf{V}_j - \mathbf{V}_i) + \frac{\nabla p}{p} (\omega_i - x_i) \quad (9)$$

$$214 \quad x_i = \frac{M_n}{M_i} \omega_i \quad (10)$$

215 Where \mathbf{V}_i is the diffusion velocity of the i^{th} species (m s^{-1}), x_i is the mole fraction of the i^{th} species
 216 (dimensionless), which is related to the mass fraction by Equation (8), and D_{ij} is the binary
 217 diffusion coefficients between the i^{th} and the j^{th} species ($\text{m}^2 \text{s}^{-1}$). A list of the binary diffusion
 218 coefficients used in the model is given in Table 1. It should be noted that Equation (9) is
 219 incorporated in the model as a constraint linking the flux term and the mass fraction term in
 220 Equation (7). In Equation (10), M_i and M_n are the molecular weight of the i^{th} species and the
 221 average molecular weight, respectively (kg mol^{-1}). Lastly, the computational domain and the
 222 boundary conditions used are described in the appendix.

223

224 *Table 1.* A list of binary diffusion coefficients used in the model.

Combination	Diffusion coefficient [$\text{m}^2 \text{s}^{-1}$]	Reference
$\text{N}_2 - \text{He}$	6.78×10^{-5}	[26]
$\text{N}_2 - \text{O}_2$	2.09×10^{-5}	[27]
$\text{N}_2 - \text{H}_2\text{O}$	2.54×10^{-5}	[28]
$\text{O}_2 - \text{He}$	7.36×10^{-5}	[26]
$\text{He} - \text{H}_2\text{O}$	8.36×10^{-5}	[28]
$\text{O}_2 - \text{H}_2\text{O}$	3.185×10^{-5}	[28]

225

226 As stated in the introduction section, it is hypothesized that plasma-induced turbulence affects the
 227 flow's velocity field and thus the gas composition. To account for such effects in the computational
 228 model, the Reynolds-Averaged Navier-Stokes (RANS) approach for modeling turbulence was
 229 followed, where a turbulent viscosity μ_T (also known as eddy viscosity) was added to the viscosity
 230 of the gas mixture. The eddy viscosity is a mathematical means to describe the loss of momentum
 231 of the flow as a result of turbulence as an "effective" viscosity that is added to the physical viscosity
 232 of the fluid. Similarly, a turbulent diffusivity D_T is added to the binary diffusion coefficients.^[29] The
 233 computation of the eddy viscosity is typically done using one of the conventional RANS turbulence
 234 models, such as the $k - \varepsilon$ model. Considering that such models were calibrated for flows without
 235 plasma, their use for plasma modified flows will yield results with unknown accuracy. To overcome
 236 this challenge, statistical analysis of the PIV data was conducted to obtain the necessary parameters
 237 to calculate the eddy viscosity resulting from the plasma generation. Following the $k - \varepsilon$ modeling
 238 approach, the turbulent kinetic energy k ($\text{m}^2 \text{s}^{-2}$) and the turbulent kinetic energy dissipation rate ε
 239 ($\text{m}^2 \text{s}^{-3}$) are defined by Equation (11) and (12).^[29]

$$240 \quad k = \frac{1}{2} \left(\overline{u'^2} + \overline{v'^2} \right) \quad (11)$$

$$241 \quad \varepsilon = 2\nu \overline{s'_{ij} \cdot s'_{ij}} \quad (12)$$

242 Where u' and v' are the time fluctuating velocity field components with respect to the average
 243 velocity field, which were calculated from PIV data by subtracting the time-averaged velocity field
 244 from each of the 400 instantaneous velocity maps captured during a measurement, then averaging
 245 the square of these fluctuations. In Equation (12), ν is the kinematic viscosity ($\text{m}^2 \text{s}^{-1}$), and s'_{ij} is the
 246 fluctuating deformation rate of the fluid (s^{-1}), which was calculated from the PIV data as outlined by
 247 Xu and colleagues.^[30] After calculating k and ε the eddy viscosity was calculated according to
 248 Equation (13).^[29]

$$249 \quad \mu_T = \rho C \frac{k^2}{\varepsilon} \quad (13)$$

250 Where C is a constant equal to 0.0016 and ρ is the self-consistent gas mixture density calculated by
251 the model. The turbulent diffusivity D_T is related to the eddy viscosity by Equation (14).^[25]

$$252 \quad D_T = \frac{\mu_T}{\rho S c_T} \quad (14)$$

253 Where $S c_T$ is the turbulent Schmidt number, obtaining an accurate value for this in a plasma-
254 modified flow is not possible; however, the turbulent Schmidt number is close to unity for a wide
255 variety of gas flows under very diverse conditions; hence it is assumed to be 1 in this
256 investigation.^[31] The model is subsequently solved with the experimental input of μ_T and D_T , which
257 were smoothed and mirrored, then added to their physical counterpart quantities. The model was
258 solved for both applied voltage cases investigated in this work, in addition to an unperturbed
259 laminar case, where the eddy viscosity and diffusivity were set to zero.

260
261 The radiative collisional model consisted of a system of Ordinary Differential Equations (ODE)
262 describing the LIF transitions. The system was solved at every point in the computational domain
263 close to the jet's orifice ($x < 20$ mm, $x/D < 7$), with the gas composition required for the ODEs being
264 taken from that calculated by the fluid flow model. To quantify the density of ground-state OH, the
265 4-level model reported by Verreycken et al. was adopted.^[21] The experimental parameters used in
266 this study are summarised in Table 2. To validate the developed computational model, the predicted
267 LIF signal decay times were compared to those measured experimentally at multiple points along
268 the jet axis; a close agreement was observed and is further discussed in the results section.

269

270

271

272

273

274

275

276 *Table 2.* Experimental parameters used in LIF measurements and absolute density calibration

Parameter	Description	Value
λ_L	Laser wavelength	282.58 [nm]
$\Delta\lambda_L$	Linewidth of the laser	0.95 [pm]
g_{int}	Overlap integral	0.017 [m]
A_L	Area of the laser beam	0.0746 [mm ²]
E_L	Laser energy per pulse	10 [μ J]
τ_L	Temporal FWHM of the laser pulse	6 [ns]
Δx	Length of the detection volume	0.359 [mm]
Δy	Width of the detection volume	0.359 [mm]
Δs	The spatial FWHM of the laser beam at the observation point	0.1795 [mm]

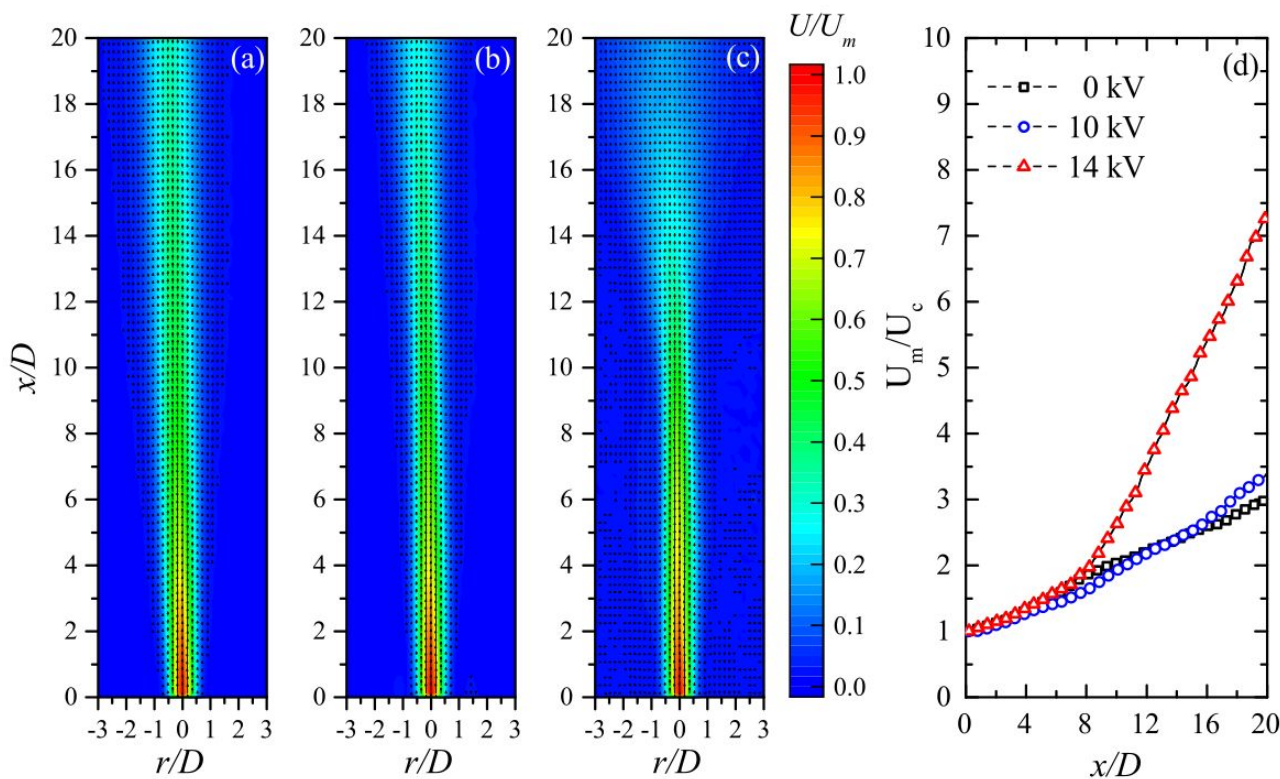
277

278

279 **3 Results and discussion**280 **3.1 Influence of plasma on jet velocity and turbulence generation**

281 PIV measurements were undertaken to quantify the velocity of the flowing helium gas and the
282 resulting perturbation of the surrounding quiescent air beyond the jet orifice. **Figure 3** (a-c) shows
283 the ensemble-averaged two-component velocity vector maps and velocity magnitude (U)
284 normalized to the measured exit velocity (U_m) for the 0 kV, 10 kV, and 14 kV cases, respectively.
285 Without a plasma discharge (0 kV case), the undisturbed helium flow appeared laminar over the
286 entire measurement region, confirmed by the ensemble-averaged centreline velocity (U_c) profile in
287 Figure 3(d). On the application of a 10 kV sinusoidal waveform, a weak discharge was observed to
288 form, indicating gas breakdown had been achieved; under such conditions, little obvious change to
289 the velocity profile was observed. An increase in applied voltage from 10 kV to 14 kV led to an
290 increase in the length of the visible plasma plume. Comparing Figure 3(b) and 3(c) highlights the
291 impact of the applied voltage on the flow structure downstream of the jet orifice, with the higher
292 applied voltage resulting in a significant reduction in the length of the laminar flow region. From
293 the ensemble-averaged centreline velocity, an abrupt change in gradient is observed at
294 approximately $8 x/D$, indicating a transition to turbulence. While such results are perhaps the first

295 quantitative measurements of velocity within a plasma jet, they remain highly consistent with
 296 previous observations made using qualitative methods such as Schlieren imaging.^[9, 32]
 297



298

299 **Figure 3.** Ensemble averaged particle imaging velocimetry measurements of the plasma jet

300 obtained at applied voltages of (a) 0 kV, (b) 10 kV, and (c) 14 kV, (d) shows the ensemble-averaged

301 centreline velocity (U_c) normalized to the measured exit velocity (U_m).

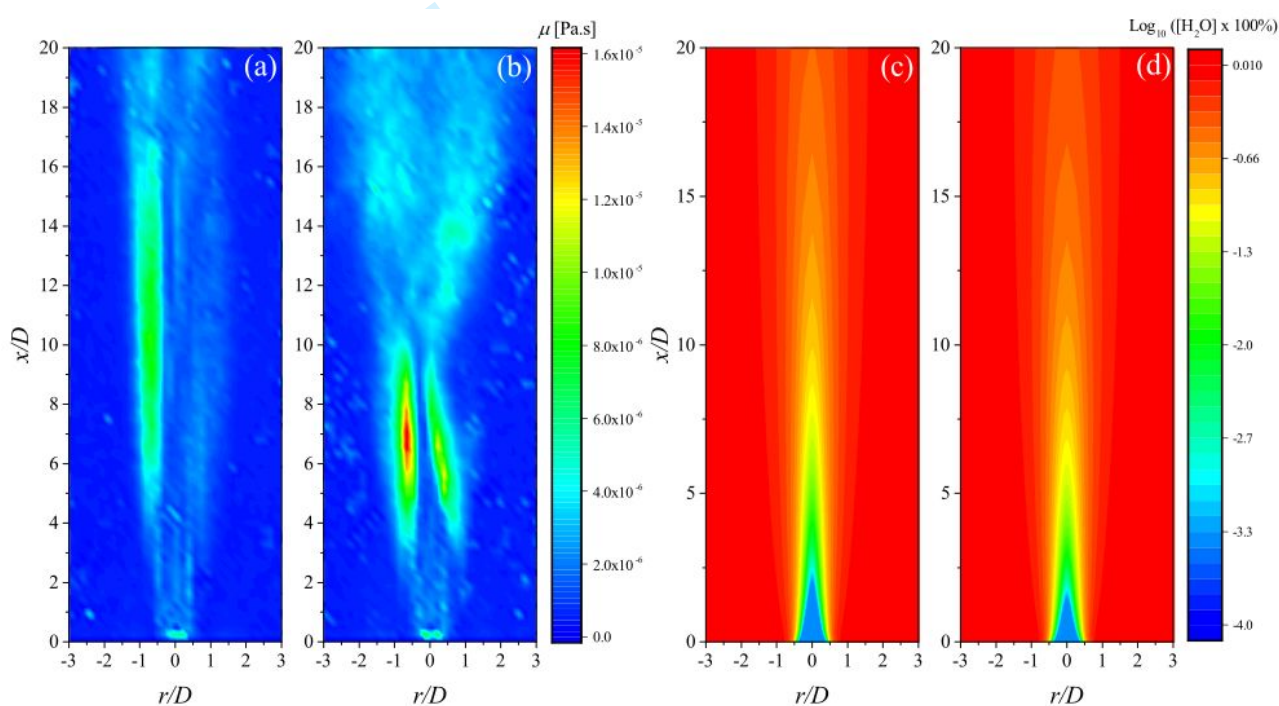
302

303 In absolute terms, the generation of plasma with an applied voltage of 14 kV was found to increase
 304 U_m by 0.94 m s^{-1} , representing a relatively modest increase of approximately 10% compared to the 0
 305 kV case. Such increases in velocity are in line with those predicted by others and are a consequence
 306 of gas heating and electrohydrodynamic forces induced by the plasma, with the latter mechanism
 307 being the most likely dominant factor.^[8, 10] Notably, the modest change in velocity associated with
 308 plasma generation cannot directly explain the rapid transition to turbulence observed when
 309 comparing Figure 3(a) and 3(c). Past studies on turbulence generation in axisymmetric round jets
 310 have revealed that turbulence initiates due to instabilities within the shear layers at the jet exit that

311 become amplified as they travel downstream.^[33-36] As the instabilities grow, they cause velocity
312 fluctuations, Reynolds shear stresses, and thus the production of turbulence.^[8] Many previous
313 studies have explored ‘excited’ jets that employ alternative means to perturb the jet flow in order to
314 investigate the mechanisms of turbulence generation.^[37-42] For example, the impact of sonic
315 excitation on the jet velocity profile shows a remarkable similarity to those observed in this
316 study^[38]; hence it is posited that plasma generation is an alternative means to excite an
317 axisymmetric round jet, resulting in the rapid onset of turbulence through increased shear layer
318 instability with little change to velocity.

319
320 While the growth of small-scale instabilities within the jet shear layer has a significant impact on
321 the laminar to turbulent transition, they also provide a mechanism to enhance entrainment of
322 quiescent air into the laminar region of the plasma jet.^[42] To investigate the influence of plasma
323 generation on instabilities in the jet shear layer, the eddy viscosity μ_T was calculated. The eddy
324 viscosity profiles for the two plasma cases investigated in this work are shown in **Figure 4** (a-b).
325 Three characteristic zones can be observed within the profiles, the first is close to the jet orifice,
326 where a region of low μ_T exists, which can be explained by the fact that this is the laminar region
327 where the amplitude of velocity fluctuations is small, leading to low turbulent kinetic energy k , and
328 consequently, a low μ_T as Equation (13) shows. The second zone (e.g., $4 - 10 x/D$ in Figure 4(b))
329 coincides with the transition region, as inferred from Figure 3(d), where the value of μ_T peaks. This
330 is attributed to the large scale fluctuations/eddies starting to appear in the transition region, leading
331 to high turbulent kinetic energy k , considering that such large fluctuations live long enough to be
332 transported downstream, the dissipation rate of the turbulent energy ε is relatively low in this
333 region, thus leading to a peak of μ_T as follows from Equation (13). The third zone (e.g. $> 10 x/D$ in
334 Figure 4(b)) coincides with the turbulent region, as inferred from Figure 3(d), which has a moderate
335 value of μ_T . As known from the energy cascade theory of turbulence,^[43] the large eddies generated
336 in the transition region break into smaller eddies in the fully turbulent region, the small eddies are

337 dissipated into heat due to the physical viscosity of the fluid.^[43] In this sense, the turbulent kinetic
 338 energy k is high, while the turbulent dissipation rate ε is also high, leading to a moderate value of μ_T .
 339 From Figure 4(a-b), it is clear that the eddy viscosity for the 14 kV case has a larger magnitude
 340 compared to the 10 kV case, which is consistent with the PIV results presented in Figure 3(b-c).
 341 When comparing the average value of μ_T in zone one ($x/D < 4$) for both cases it is found that μ_T for
 342 the 10 kV case is approximately 70%-80% of that for the 14 kV case, indicating that the plasma's
 343 perturbation to the flow in the laminar region is more significant for the 14 kV case in comparison
 344 to the 10 kV case.



345
 346 **Figure 4.** Eddy viscosity calculated from PIV measurements for the (a) 10 kV and (b) 14 kV
 347 excited plasma jet and the calculated entrainment of H_2O into the helium flow under (c) 0 kV and
 348 (d) 14 kV conditions.

349

350 As described in section 2.3, a higher value of the eddy viscosity of μ_T indicates a higher value of
 351 eddy diffusivity D_T , which adds to the physical diffusion coefficients, leading to increased
 352 entrainment of air into the helium jet for higher values of μ_T . To highlight the impact of the plasma-
 353 induced entrainment, Figure 4 (c) and (d) show the computed percentage of H_2O in the gas mixture

354 for the 0 kV and 14 kV cases, respectively. Notably, the 0 kV case represents an unperturbed
355 laminar flow where quiescent air is entrained due to the physical diffusion only. While in the case
356 of the perturbed flow shown in Figure 4(d), air entrainment occurs due to physical diffusion plus the
357 “effective” eddy diffusion due to turbulence. The centreline concentration of H₂O at the jet exit in
358 both cases was set to 0.00004%, a value obtained from the helium gas provider. Moving
359 downstream to 2 x/D, the level of H₂O in the unperturbed 0 kV case was found to increase to be
360 approximately 0.0005 %. In contrast, the H₂O concentration in the perturbed case was found to be
361 0.0034 %, representing a 7-fold increase. Further downstream at 6 x/D, the H₂O concentration in
362 the unperturbed case was found to be 0.046%, compared to 0.082% in the perturbed case,
363 representing a 1.8-fold increase. When moving downstream, the difference between the unperturbed
364 case and the 14 kV perturbed case diminishes as a result of the increasing density of the gas
365 mixture, which lowers the value of the eddy diffusivity as defined by Equation (4).

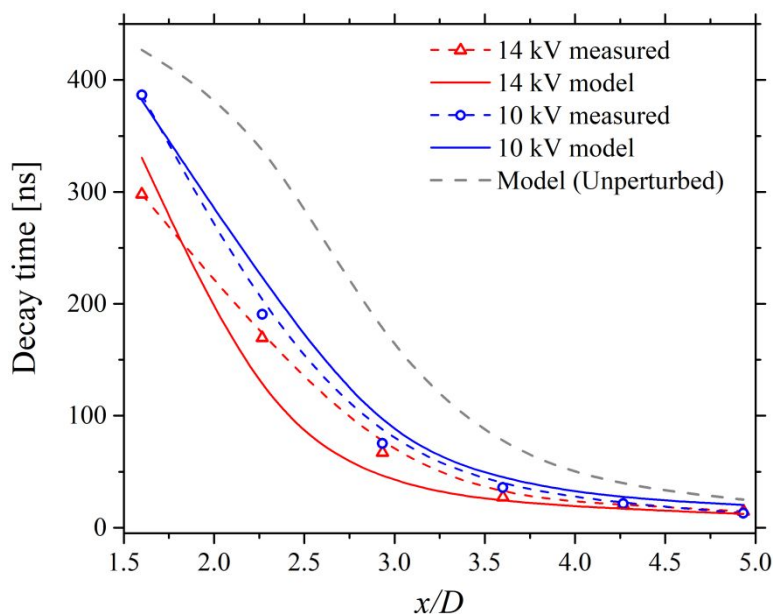
366

367 3.2 OH fluorescence decay rate and absolute density

368 As described by Yonemori et al. the decay rate of the LIF signal from OH provides a sensitive
369 indication of the helium-air mixing ratio as the quenching rate of the laser-excited state is strongly
370 influenced by the density of quenching species present within the flowing helium gas (e.g., N₂, O₂,
371 H₂O).^[44] In this study, a comparison between the measured LIF decay rate and computed decay rate
372 by the model was used to validate the adopted computational approach describing plasma-induced
373 entrainment, thus enabling the accurate quantification of absolute OH density downstream of the jet
374 exit. **Figure 5** shows the measured and computed LIF decay time as a function of downstream
375 spatial position for both the 10 kV and 14 kV cases; additionally, the predicted decay time for a
376 hypothetical unperturbed (laminar) helium flow is shown. The LIF decay time is shorter at all
377 spatial positions under both 10 and 14 kV excitation compared to what would be observed under
378 idealized laminar conditions. As highlighted in Figure 4 (c) and (d), plasma generation increases air
379 entrainment, which acts to increase the density of quenchers and thus increase the quenching rate of

380 the OH fluorescent state. This result provides experimental evidence supporting the hypothesis that
 381 plasma generation increases the entrainment of air within the helium jet at all spatial positions. The
 382 calculated decay times closely match the measured data points, with both cases showing very good
 383 agreement.

384



385

386 **Figure 5.** Comparison between measured and calculated decay time of the laser-induced fluorescent
 387 signal as a function of downstream distance from the jet orifice.

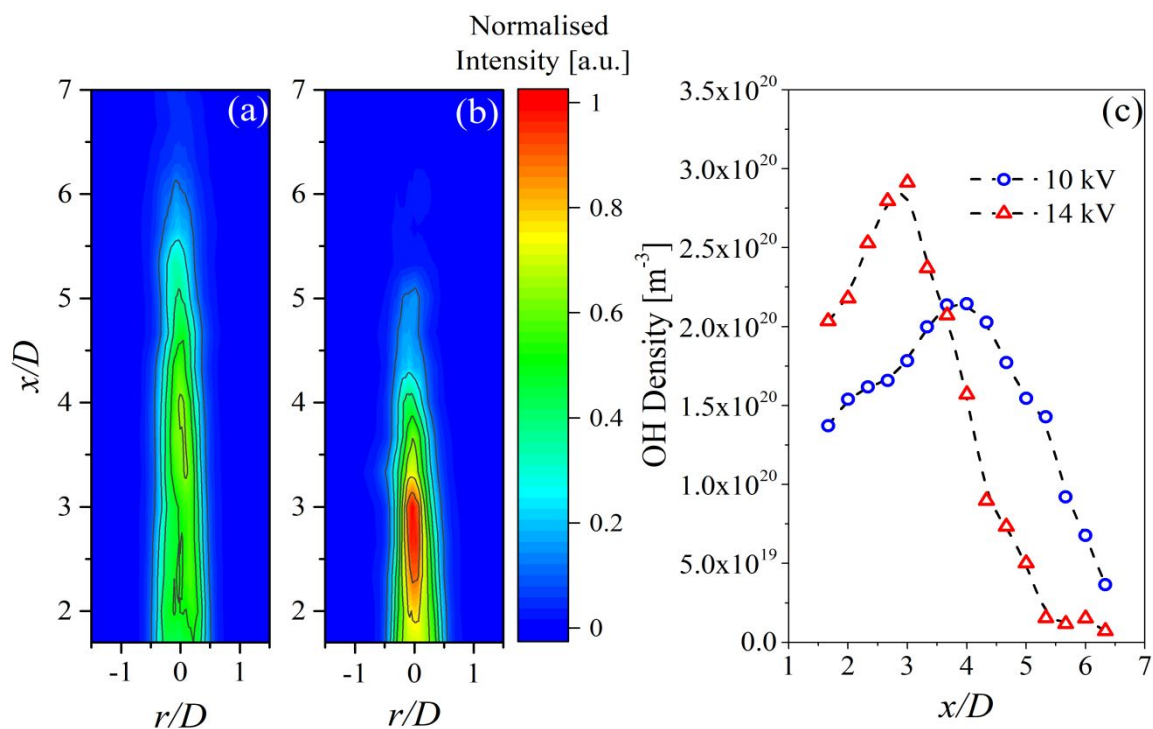
388

389 Using the experimental procedure outlined in section 2.2, the 2D LIF emission profile was compiled
 390 for both the 10 kV and 14 kV case and is shown in **Figure 6(a)** and **6(b)**, respectively. By
 391 comparing the centreline LIF intensity obtained from the 2D profile and the computed LIF intensity
 392 from the collisional radiative model, the absolute ground state OH density was calculated as a
 393 function of downstream position from the jet exit, shown in **Figure 6(c)**. Consistent with the
 394 observations made in many previous studies, an increase in applied voltage was observed to
 395 increase the peak OH density within the plasma plume.^[23] Given that hydrogen-based species
 396 densities increase when the air/water vapor fraction increases,^[45] it becomes likely that an increase
 397 in the applied voltage results in an increase in OH density as a result of not only more intense
 398 discharge conditions but also increased entrainment of air and H₂O. Critically, the position of peak

399 OH density was found to vary depending on the applied voltage. This phenomenon is attributed to
400 the interplay between the physical properties of the plasma (i.e., n_e and T_e) and the fluid dynamics
401 of the helium jet. With an applied voltage of 14 kV, perturbations within the jet shear layer caused
402 by plasma generation induce an early transition to a turbulent flow regime, as confirmed in Figure 3
403 and 4. Under such conditions, the generation of OH close to the jet exit is locally enhanced due to
404 enhanced H₂O entrainment combined with the relatively intense discharge conditions. Beyond the
405 local maxima, the elevated air mole fraction as a result of the induced turbulence acts to quench the
406 discharge and thus impedes OH generation. Conversely, at the lower applied voltage of 10 kV the
407 discharge is comparatively less intense, meaning less air entrainment and a weaker plasma is
408 formed, both factors contributing to limit OH production. However, less shear layer perturbation
409 causes less air entrainment into the helium flow; thus, the discharge can propagate further from the
410 orifice. Consequently, the concentration of ground-state OH is significantly higher far downstream
411 from the jet exit in the 10 kV case compared to the 14 kV case.

412
413 The presented findings have considerable implications from a practical perspective as it has been
414 demonstrated that the density of OH at a given downstream position is strongly influenced by both
415 the discharge characteristics and its interaction with the quiescent background gas. While it is
416 generally assumed that a higher plasma generation voltage results in enhanced production of OH,
417 Figure 6 clearly shows that this only holds true close to the jet orifice. With increasing voltage
418 comes increasing entrainment, which ultimately begins to quench the discharge and negatively
419 affects downstream OH production. Counterintuitively, Figure 6(c) indicates that at a position of 5.3
420 x/D (i.e., 16 mm from the jet exit), there is an order of magnitude more OH from a plasma
421 generated using 10 kV compared to one generated using 14 kV excitation. These findings
422 demonstrate that the complex interplay between the physicochemical properties of the plasma and
423 the fluid dynamic properties of the flowing noble gas must be carefully considered when designing
424 plasma jet sources for use in applications such as biomedicine and materials processing.

425



426

427 **Figure 6.** Composite 2D normalised LIF intensity for (a) 10 kV, and (b) 14 kV case. Absolute
 428 centreline OH density for the 10 kV and 14 kV case as a function of distance from the jet orifice.

429

430 4 Conclusion

431 This contribution has employed Particle Imaging Velocimetry, Laser-Induced Fluorescence, and
 432 Computational Fluid Dynamics to explore the interplay between the propagating plasma plume and
 433 the quiescent background air in an axisymmetric dielectric barrier discharge jet. Despite the rapid
 434 onset of turbulence observed following plasma ignition, it was demonstrated that the presence of the
 435 plasma had little impact on the jet exit velocity of the helium flow. Through statistical analysis of
 436 the measured velocity field from the plasma jet under varying excitation conditions, it was
 437 determined that plasma generation resulted in shear layer perturbations that grow downstream to
 438 initiate the early onset of turbulence.

439

440 Using the experimentally derived eddy viscosity, a computational model was developed to calculate
 441 the density of humid air entrained within the helium jet flow. To validate the model, the calculated

442 decay time of the laser-induced fluorescent state of OH was compared against those measured
443 experimentally and found to be in good agreement. Finally, the computational model was used to
444 convert the measured LIF intensity into an absolute OH density from which it was concluded that
445 OH production is strongly influenced by the interplay between the propagating plasma and the
446 background air. At high applied voltages, it was found that OH density increases close to the exit
447 but is rapidly reduced downstream as a result of the elevated air content quenching the discharge.

448

449 In summary, this study demonstrates an intricate link between the physicochemical properties of the
450 plasma and its interaction with the quiescent air. As many applications rely on the presence of
451 reactive oxygen and nitrogen species, which are predominantly formed when the plasma interacts
452 with the background environment, the results of this study provide valuable insight into the
453 underpinning mechanisms governing these interactions.

454

455 **Acknowledgments:** JLW & MIH would like to acknowledge the support of the Engineering and
456 Physical Sciences Research Council (Projects EP/S025790/1, EP/S017623/1, EP/R041849/1, and
457 EP/N021347/1). RDW would like to acknowledge the support of the NATO AVT-254 plasma flow
458 control group.

459

460 **Keywords:** LIF; PIV; plasma; turbulence.

461

462

463

464

465

466

- 467 [1] A. J. Knoll, P. Luan, A. Pranda, R. L. Bruce, G. S. Oehrlein, *Plasma Process Polym.* **2018**, *15*,
468 1700217. doi.org/10.1002/ppap.201700217
- 469 [2] X. Deng, A. Yu Nikiforov, T. Coenve, P. Cools, G. Aziz, R. Morent, N. De Geyter, C. Leys,
470 *Sci. Rep.*, **2015**, *5*, 10138. doi.org/10.1038/srep10138
- 471 [3] P. B. Flynn, S. Higginbotham, N. H. Alshraiedeh, S. P. Gorman, W. G. Graham, B. F. Gilmore,
472 *Int. J. Antimicrob. Agents*, **2015**, *46*, 1. doi.org/10.1016/j.ijantimicag.2015.02.026
- 473 [4] H. Tanaka, M. Mizuno, K. Ishikawa, S. Toyokuni, H. Kajiyama, F. Kikkawa, M. Hori, *Plasma*,
474 **2018**, *1*, 1. doi.org/10.3390/plasma1010014
- 475 [5] S. Bekeschus, P. Favia, E. Robert, T. Von Woedtke, *Plasma Process Polym.*, **2019**, *16*,
476 1800033. doi.org/10.1002/ppap.201800033
- 477 [6] S. Iseni, A. Schmidt-Bleker, J. Winter, K-D. Weltmann, S. Reuter, *J. Phys. D. Appl. Phys.*,
478 **2014**, *47*, 152001. doi.org/10.1088/0022-3727/47/15/152001
- 479 [7] T. Darny, J. M. Pouvesle, J. Fontane, L. Joly, S. Dozias, E. Robert, *Plasma Sources Sci.*
480 *Technol.*, **2017**, *26*, 105001. doi.org/10.1088/1361-6595/aa8877
- 481 [8] R. D. Whalley, J. L. Walsh, *Sci. Rep.*, **2016**, *6*, 31756. DOI:10.1038/srep31756
- 482 [9] R. Xiong, Q. Xiong, A. Yu Nikiforov, P. Vanraes, C. Leys, *J. Appl. Phys.*, **2012**, *112*, 033305.
483 DOI:10.1063/1.4746700
- 484 [10] S. Park, U. Cvelbar, W. Choe, S. Y. Moon, *Nat. Commun.*, **2018**, *9*, 371. DOI:10.1038/s41467-
485 017-02766-9
- 486 [11] M. I. Hasan, J. W. Bradley, *J. Phys. D: Appl. Phys.*, **2016**, *49*, 055203. DOI:10.1088/0022-
487 3727/49/5/055203
- 488 [12] M. Boselli, V. Colombo, E. Ghedini, M. Gherardi, R. Laurita, *Plasma Chem. Plasma Process.*,
489 **2014**, *34*, 853. DOI:10.1007/s11090-014-9537-1
- 490 [13] A. Michalke, *Journal of Fluid Mechanics*, **1965**, *23*, 3. DOI:10.1017/S0022112065001520
- 491 [14] A. V. Pipa, T. Bindemann, R. Foest, E. Kindel, J. Röpcke, K-D. Weltmann, *J. Phys. D. Appl.*
492 *Phys.*, **2008**, *41*, 194011. DOI:10.1088/0022-3727/41/19/194011

- 493 [15] K. McKay, J. S. Oh, J. L. Walsh, J. W. Bradley, *J. Phys. D. Appl. Phys.*, **2013**, *46*, 464018.
494 DOI:10.1088/0022-3727/46/46/464018
- 495 [16] A. Schmidt-Bleker, J. Winter, S. Iseni, M. Dünnbier, K-D. Weltmann, S. Reuter, *J. Phys. D.*
496 *Appl. Phys.*, **2014**, *47*, 145201. DOI:10.1088/0022-3727/47/14/145201
- 497 [17] D. Riès, G. Dilecce, E. Robert, P. F. Ambrico, S. Dozias, J. M. Pouvesle, *J. Phys. D. Appl.*
498 *Phys.*, **2014**, *47*, 275401. DOI:10.1088/0022-3727/47/27/275401
- 499 [18] S. Reuter, J. Winter, A. Schmidt-Bleker, D. Schroeder, H. Lange, N. Knake, V. Schulz-Von
500 Der Gathen, K. D. Weltmann, *Plasma Sources Sci. Technol.*, **2012**, *21*, 024005. DOI:10.1088/0963-
501 0252/21/2/024005
- 502 [19] A. Nikiforov, L. Li, N. Britun, R. Snyders, P. Vanraes, C. Leys, *Plasma Sources Sci. Technol.*,
503 **2014**, *23*, 015015. DOI:10.1088/0963-0252/23/1/015015
- 504 [20] C. Tropea, A. L. Yarin, J. F. Foss, *Springer Handbook of experimental fluid mechanics*,
505 Springer, NY, USA **2007**.
- 506 [21] T. Verreycken, R. M. Van Der Horst, N. Sadeghi, P. J. Bruggeman, *J. Phys. D. Appl. Phys.*,
507 **2013**, *46*, 464004. DOI:10.1088/0022-3727/46/46/464004
- 508 [22] G. Dilecce, L. M. Martini, P. Tosi, M. Scotoni, S. De Benedictis, *Plasma Sources Sci.*
509 *Technol.*, **2015**, *24*, 034007. DOI:10.1088/0963-0252/24/3/034007
- 510 [23] YF. Yue, XK. Pei, XP. Lu, *IEEE Trans. Radiat. Plasma Med. Sci.*, **2017**, *1*, 6. DOI:
511 10.1109/TRPMS.2017.2757037
- 512 [24] R. J. Kee, M. E. Coltrin, P. Glarborg, *Chemically reacting flow: theory and practice*, Wiley-
513 Interscience, Hoboken, NJ, USA **2003**. DOI:10.1002/0471461296
- 514 [25] R. B. Bird, W. E. Stewart, E. N. Lightfoot, *Transport Phenomena*, 2nd edition, John Wiley &
515 Sons, NY, USA **2002**.
- 516 [26] S. P. Wasik, K. E. McCulloh, *J. Res. Natl. Bur. Stand.*, **1968**, *73A*, 2.
- 517 [27] J. Rohling, J. Shen, C. Wang, J. Zhou, C. E. Gu, *Appl. Phys. B*, **2007**, *87*, 355.
518 DOI:10.1007/s00340-007-2595-9

- 519 [28] C. V. Paganelli, F. K. Kurata, *Respiration Physiology*, **1977**, *30*, 15. DOI:10.1016/0034-
520 5687(77)90018-4
- 521 [29] T. J. Chung, *Computational fluid dynamics*, Cambridge University Press, Cambridge, UK
522 **2002**.
- 523 [30] D. Xu, J. Chen, *Exp. Therm. Fluid Sci.*, **2013**, *44* 662.
524 DOI:10.1016/j.expthermflusci.2012.09.006
- 525 [31] C. Gualtieri, A. Angeloudis, F. Bombardelli, S. Jha, T. Stoesser, *Fluids*, **2017**, *2*, 17.
526 DOI:10.3390/fluids2020017
- 527 [32] M. Boselli, V. Colombo, M. Gherardi, R. Laurita, A. Liguori, P. Sanibondi, E. Simoncelli, A.
528 Stancampiano, *IEEE Trans. Plasma Sci.*, **2015**, *43*, 3. DOI: 10.1109/TPS.2014.2381854
- 529 [33] M. Lessen, *Nat. Adv. Comm. Aero., Wash.*, **1950**, 979.
- 530 [34] A. Michalke, *J. Fluid Mech.*, **1965**, *23*, 3. DOI:10.1017/S0022112065001520
- 531 [35] A. Michalke, R. Wille, *Proc. 11th Intern. Congr. Appl. Mech.*, **1965**.
- 532 [36] A. Michalke, *Prog. Aerosp. Sci.*, **1972**, *12*, 213. DOI:10.1016/0376-0421(72)90005-X
- 533 [37] H. Sato, *J. Phys. Soc. Jpn.*, **1956**, *11*, 702. DOI:10.1143/JPSJ.11.702
- 534 [38] H. Sato, *J. Phys. Soc. Jpn.*, **1959**, *14*, 1797. DOI:10.1143/JPSJ.14.1797
- 535 [39] E. Gutmark, C-M. Ho, *Phys. Fluids*, **1983**, *26*, 2932. DOI:10.1063/1.864058
- 536 [40] J. Cohen, I. Wygnanski, *J. Fluid Mech.*, **1987**, *176*, 191. DOI:10.1017/S0022112087000624
- 537 [41] S. W. Rienstra, *J. Sound Vib.*, **1983**, *86*, 4. DOI:10.1016/0022-460X(83)91019-2
- 538 [42] J. Westerweel, C. Fukushima, J.M. Pederson, J. C. R. Hunt, *Phys. Rev. Lett.*, **2005**, *95*,
539 174501. DOI:10.1103/PhysRevLett.95.174501
- 540 [43] B. Lautrup, *Physics of continuous matter: exotic and everyday phenomena in the macroscopic*
541 *world*, Institute of Physics, Bristol, UK **2005**.
- 542 [44] S. Yonemori, Y. Nakagawa, R. Ono, T. Oda, *J. Phys. D. Appl. Phys.*, **2012**, *45*, 225202.
543 DOI:10.1088/0022-3727/45/22/225202

- 544 [45]T. Murakami, K. Niemi, T. Gans, D. O'Connell, W. G. Graham, *Plasma Sources Sci. Technol.*,
- 545 **2013**, 22, 015003. DOI:10.1088/0963-0252/22/1/015003

For Peer Review