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► **To cite this version:**

Paule Lapeyre, Rodrigo Brenner, Michael Christopher Nagorski, Jean-Philippe Gagnon, Martin Chamberland, et al.. Quantifying Flare Combustion Efficiency using an Imaging Fourier Transform Spectrometer. 2024. hal-04418519

HAL Id: hal-04418519

<https://hal.science/hal-04418519>

Preprint submitted on 26 Jan 2024

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Quantifying Flare Combustion Efficiency using an Imaging Fourier Transform Spectrometer

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Keywords: Hyperspectral Imaging, Flare emissions quantification, Fenceline measurement, Imaging Fourier transform spectrometer, Remote Sensing

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1 **Abstract**

2 Mid-wavelength infrared (MWIR) imaging Fourier transform spectrometers (IFTSs) are a
3 promising technology for measuring flare combustion efficiency (CE), defined as the fraction of
4 carbon supplied to the flare as fuel that is converted into carbon dioxide. These devices generate
5 spectrally-resolved intensity images of the flare plume, which may then be used to infer column
6 densities of relevant species along each pixel line-of-sight. In parallel, a 2D projected velocity field
7 may be inferred from the apparent motion of flow features between successive images. Finally,
8 the column densities and velocity field are combined to estimate the mass flow rates for the species
9 needed to calculate the CE. Since the MWIR IFTS can measure all carbon-containing species in
10 the flare plume, it is possible to measure CE without knowing the fuel flow rate, which is important
11 for fenceline measurements.

12 This work demonstrates this approach on a laboratory heated vent, and then deploys the
13 technique on two working flares: a combustor burning natural gas at a known rate, and a steam-
14 assisted flare at a petrochemical refinery. Analysis of the IFTS data highlights the potential of this
15 approach, but also areas for future development to transform this approach into a reliable technique
16 for quantifying flare CE.

17 **1. Introduction**

18 The petrochemical industry uses flaring to transform high global warming potential (GWP)
19 hydrocarbons (HCs) into CO₂, principally to mitigate the impact of these emissions on climate
20 change. In the case of upstream oil and gas extraction, for example, simple flares are used to
21 dispose of natural gas as a by-product of oil extraction (so-called “associated gas”), while
22 downstream flaring often involves a wider range of hydrocarbon species and more costly and
23 sophisticated flare designs. Globally, 144 billion cubic meters of gas was flared in 2021 [1].
24 Recovery and reuse processes exist (gas to power, gas to hydrogen, gas reinjection [2]), but their
25 feasibility is subject to favorable economic and technical conditions. Consequently, vast amounts
26 of HCs will continue to be flared for the foreseeable future.

27 The performance of a flaring process may be quantified by its combustion efficiency (CE),
28 defined as the ratio of the mass flow rate of carbon affixed to carbon dioxide leaving the flare
29 plume to the mass flow rate of carbon affixed to fuel entering the flare stack. While CE is usually
30 assumed to be close to 100% [3], external factors may reduce this number significantly.
31 Crosswinds, for example, can induce an aerodynamic fuel steeping mechanism [4] that removes
32 the fuel from the combustion zone; while steam-assisted and air-assisted flows, which are intended
33 to suppress soot formation, can also cause excess HC emissions [5, 6]. Given the large volumes of
34 HCs flared annually, the impact of flaring on climate change is very sensitive to seemingly small
35 variations on CE. For example, Plant et al. [7] recently determined an average CE for lit flares
36 from upstream processing in Bakken, Eagle Ford, and Permian basins of 95% based on airborne
37 measurements, as opposed to the nominal value of 98% [3]. When unlit flares are considered, the
38 average CE drops to 91%; this 7% difference corresponds to the annual CO₂ emissions of between

39 2.9-8 million cars [7], with the higher number corresponding to methane having a CO₂(e) of 86
40 over a 20-year span.

41 This result highlights the importance of quantifying flare combustion efficiency. *In situ*
42 approaches like extractive gas sampling are viable in laboratory measurements [6, 8] but mostly
43 infeasible on operating flares. A further challenge is that, particularly in the case of fence-line
44 measurements, the fuel flow rate entering the flare is often unknown. Several studies have relied
45 on aerial mass balance measurements by flying an aircraft equipped with gas-analyzers to measure
46 CH₄ and CO₂ concentrations along a helical trajectory surrounding the plume [7, 9, 10]. Species
47 mass fluxes may be obtained by combining concentration measurements with wind speeds, and
48 the fuel supply rate is inferred through a carbon mass balance. While this approach is appropriate
49 for developing an overall sense of flare emissions, e.g., to develop an emissions inventory over a
50 large area, it is not suitable for assessing the performance of individual flares under specific
51 operating conditions.

52 Accordingly, flare operators, regulators, and equipment manufacturers urgently need
53 diagnostics that can quantify flare CE rapidly through optical standoff measurements. Normal
54 quantitative optical gas imaging (QOGI) cameras are not up to this task, since they cannot
55 distinguish multiple carbon-containing species in the plume. One of the earliest standoff
56 approaches is passive Fourier transform infrared spectroscopy (PFTIR) [11], in which an FTIR is
57 used to measure the spectral intensity emitted by the hot combustion products along a single line-
58 of-sight (LOS) through the plume. A spectroscopic model is then inverted to infer the species
59 concentrations, which are then related to a “local” combustion efficiency averaged along the LOS.
60 A major drawback of this approach is that it assumes that the plume is homogeneous in composition,

61 which is often not the case, and the CE estimate may vary considerably based on the LOS.
62 Consequently, an optical technique that provides both spectral and spatial resolution is desirable.

63 Zeng et al. [12], adapted a multispectral (MS) camera to visualize the radiant intensity emitted
64 by CO₂, CO, and unburned HCs in the flare plume; the spectral intensity measured by the camera
65 is then related to the relative concentrations of these species through a rudimentary spectroscopic
66 model, providing a local CE for each pixel. Finally, an overall CE for the flare is obtained by
67 averaging the CE for a subset of pixels outside of the combustion zone. While most MS cameras
68 generate images using filter wheels mounted external to the cold shield, these are generally
69 unsuitable for this application since the blackbody emission from the filters dominates the signal
70 component arising from thermal emission by the flare products. Instead, the specialized camera
71 used by Zeng et al. [12] features a microlens array that images the scene onto a number of
72 subdivided regions of the focal plane array (FPA), each of which is equipped with a distinct
73 broadband filter that is cooled along with the FPA. This amounts to a trade-off between spatial
74 and spectral resolution, and typically the number of spectral channels is 10 or smaller.
75 Unfortunately, since the detected spectral intensity is influenced both by species concentration and
76 temperature, it is necessary to measure both these quantities simultaneously, and the number of
77 channels in the MS camera may be inadequate to obtain robust estimates of all the parameters
78 needed to define the gas state within the flare plume.

79 Mid-wavelength infrared hyperspectral (MWIR-HS) imaging using imaging Fourier transform
80 spectrometers (IFTSs) combine the spectral resolution of PFTIR with the spatial resolution of the
81 MS camera to provide a detailed map of both temperature and species concentration within the
82 flare plume [13]. IFTSs use interferometry to generate a data cube of thousands of near-
83 simultaneous images, leading to a spectral resolution that far exceeds what is possible using a MS

84 camera without sacrificing spatial resolution, although at the expense of temporal resolution. The
85 spectroscopic data may be inverted to solve for the temperature and species concentration fields
86 simultaneously; these concentrations are then combined with a 2D map of intensity-weighted
87 velocities inferred from a sequence of raw broadband images in the interferogram to produce mass
88 flow rates.

89 This instrument has been used to measure gaseous emissions from a variety of industrial
90 combustion processes, including smoke stacks [14] and engine exhausts [15]. In the context of
91 emissions monitoring, key advantages of this approach include: (1) the spectral and spatial
92 resolution is generally sufficient to obtain the concentration and temperature gradients, which are
93 important in heterogenous plumes; (2) the spectral resolution may be further used to fingerprint
94 species when the plume has an uncertain chemical composition; and, in the case of emission
95 spectroscopy, (3) measurements may be carried out in day or night, and are not as sensitive to
96 nonuniform background illumination as are techniques based on absorption spectroscopy, e.g.,
97 using the sun or a cloud layer.

98 Savary et al. [16] were the first to use IFTSs to infer flare CE. In their work, the plume
99 temperature and species concentrations are assumed to be homogeneous along each LOS in order
100 to simplify the spectroscopic model. However, Grauer et al. [17] and Miguel et al. [18] used CFD
101 simulations to demonstrate that Gaussian distributions are more appropriate in the case of flare
102 plumes. Miguel et al. [18] also conducted several proof-of-concept measurements on lab scale
103 heated vents and a lab-scale steam and air-assist flare.

104 This work applies the technique developed by Grauer et al. [17] and Miguel et al. [18] to
105 conduct benchmarking experiments on a heated vent under conditions representative of a flare
106 plume, and then measure the combustion efficiency of a portable combustor (enclosed flare)

107 operating at as known firing rate and a steam-assisted flare at an oil refinery burning an unspecified
 108 fuel at an unknown rate. In the case of the combustor, the carbon mass flow rate in the plume is
 109 consistent with the estimated fuel supply rate, and the overall combustion efficiency is nearly
 110 100%. The refinery flare, in contrast, is over-assisted and a significant quantity of unburned
 111 hydrocarbons are visible in the plume. The paper concludes with an assessment of the strengths
 112 and weaknesses of this approach, and areas for improvement. In particular, while the IFTS
 113 provides rich spectral data, a major challenge concerns the inference procedure and the model
 114 errors that are made to reduce the ill-posed problem. Overall, however, these results highlight the
 115 promise of this technique, and its potential to reduce greenhouse gas emissions from flaring.

116

117 **2. Flare combustion efficiency**

118 Flare combustion efficiency is defined as the mass flow rate of the carbon affixed to CO₂ leaving
 119 the flare plume divided by the mass flow rate of carbon entering the flare stack. Since the latter
 120 quantity may not be readily available, particularly for fence-line monitoring, it may instead be
 121 inferred from a mass balance of all carbon-containing species in the flare plume

$$122 \quad \text{CE} = \frac{\dot{m}_{\text{C,CO}_2}}{\dot{m}_{\text{C,fuel}}} \approx \frac{\dot{m}_{\text{C,CO}_2}}{\sum \dot{m}_{\text{C,plume}}} \quad (1)$$

123 As shown in Figure 1, the mass flow rate of the k th species in the plume is found by integrating
 124 the product of the corresponding species column density (kg/m²) with the normal component of
 125 the 2D projected velocity along a control line that transects the plume in the image plane

$$126 \quad \dot{m}_{\text{C},k} = \frac{M_{\text{C}}}{M_k} \dot{m}_k = \frac{M_{\text{C}}}{M_k} \int_{\Theta} \rho_k(\theta) \mathbf{u}(\theta) \cdot \hat{\mathbf{n}} d\theta \quad (2)$$

127 where the mass flow rate is adjusted by the ratio of the carbon molar mass, $M_C = 12$ atomic mass
 128 units (amu), and the k th species molecular molar mass, M_k , to obtain the mass flow rates of carbon.
 129 The pixel column density of the k th species for the LOS at a location θ along the control line is

$$130 \quad \rho_k(\theta) = \frac{M_k p}{k_B N_A} \int_{\text{LOS}(\theta)} \frac{\chi_k(s, \theta)}{T(s, \theta)} ds \quad (3)$$

131 where p is the atmospheric pressure, k_B is Boltzmann's constant, N_A is Avogadro's number, and
 132 $T(s, \theta)$ and $\chi_k(s, \theta)$ are the temperature and mole fraction of the k th species evaluated at a location
 133 s along an LOS for a pixel located at θ on the control line. The procedures for calculating the
 134 species concentration, the plume temperature and the projected velocity field are described in the
 135 Sec. 4. In principle the denominator of Eq. (1) could include soot, although the contribution of soot
 136 to the overall carbon balance is usually negligible [19].

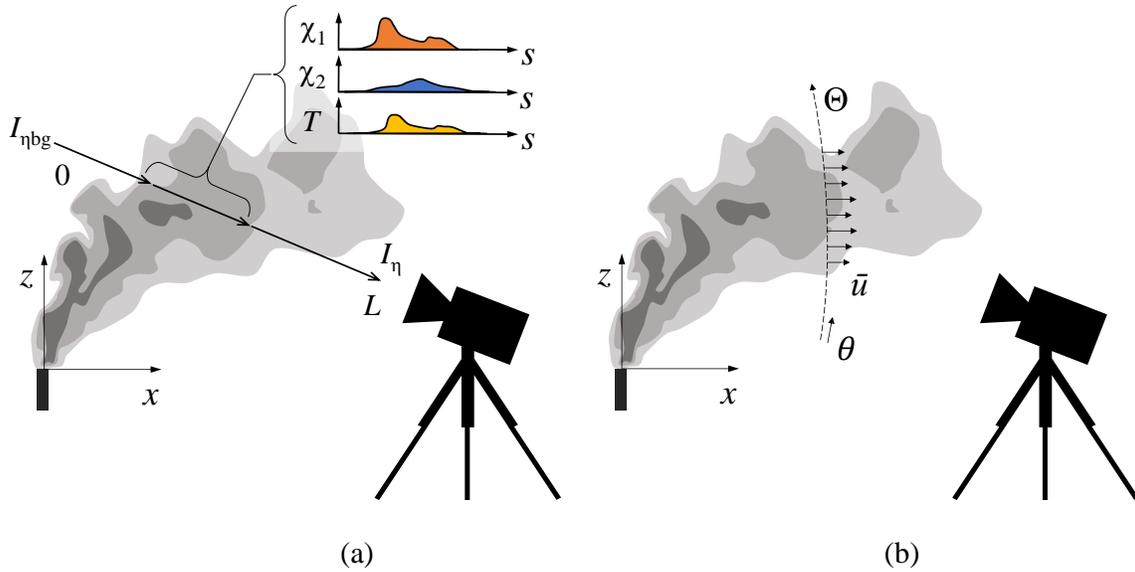


Figure 1: Schematic of a flare combustion efficiency measurement using an IFTS: (a) temperature and species concentration distributions are inferred from a spectroscopic model, Eq. (4); (b) these distributions are then used to derive column densities along a control line through Eq. (3), which is combined with a 2D intensity-weighted velocity map to obtain mass flow rates via Eq. (2).

137 **3. Measurement model**

138 The measurement model connects the quantities-of-interest, in this case the temperature and
 139 concentration distribution along each line of sight within the plume, to the spectral intensity
 140 measured by the IFTS. It is composed of a spectroscopic model, describing the intensity emitted
 141 by the scene that is incoming to the camera aperture, and a camera response model, describing the
 142 effect of the instrument on the incident scene intensity.

143

144 *3.1. Spectroscopic model*

145 The spectral intensity at wavenumber η entering the camera aperture, $I_{\eta,L}$, is defined by the
 146 radiative transfer equation (RTE) along each pixel line-of-sight

147
$$I_{\eta,L} = I_{\eta,bg} \exp\left(-\int_0^L \kappa_{\eta}(s) ds\right) + \int_0^L \kappa_{\eta}(s) I_{\eta,b} [T(s)] \exp\left(-\int_s^L \kappa_{\eta}(s') ds'\right) ds \quad (4)$$

148 where L is the length of line-of-sight from behind the plume to the camera aperture, $I_{\eta,bg}$ is the
 149 background intensity, $I_{\eta,b}$ is the blackbody intensity, and κ_{η} is the spectral absorption coefficient.

150 The background intensity may be determined from a pixel outside of the flare plume, although
 151 its contribution to $I_{\eta,L}$ is usually small compared to the emission term over the vibrational-
 152 rotational (V-R) bands of the species in the plume. For N_s participating species (e.g., CO₂, CO,
 153 H₂O, and various HCs) the spectral absorption coefficient is given by

154
$$\kappa_{\eta}(s) = \sum_{k=1}^{N_s} \kappa_{\eta,k} [\chi_k(s), T(s)] \quad (5)$$

155 The spectral absorption cross-sections of individual species may be found using a spectral line
 156 database (e.g. HITRAN [20]) or from empirical spectra. In the case of HITRAN-derived spectra,

157 the absorption coefficients of common species are precomputed for given concentrations and
 158 temperatures, assuming air broadening:

$$159 \quad \kappa_{\eta,k} [\chi_k(s), T(s)] = \sum_{ij} S_{k,ij} [T(s)] f [\eta, \eta_{ij}, T(s), p] \frac{\chi_k(s) p}{k_B T(s)} \quad (6)$$

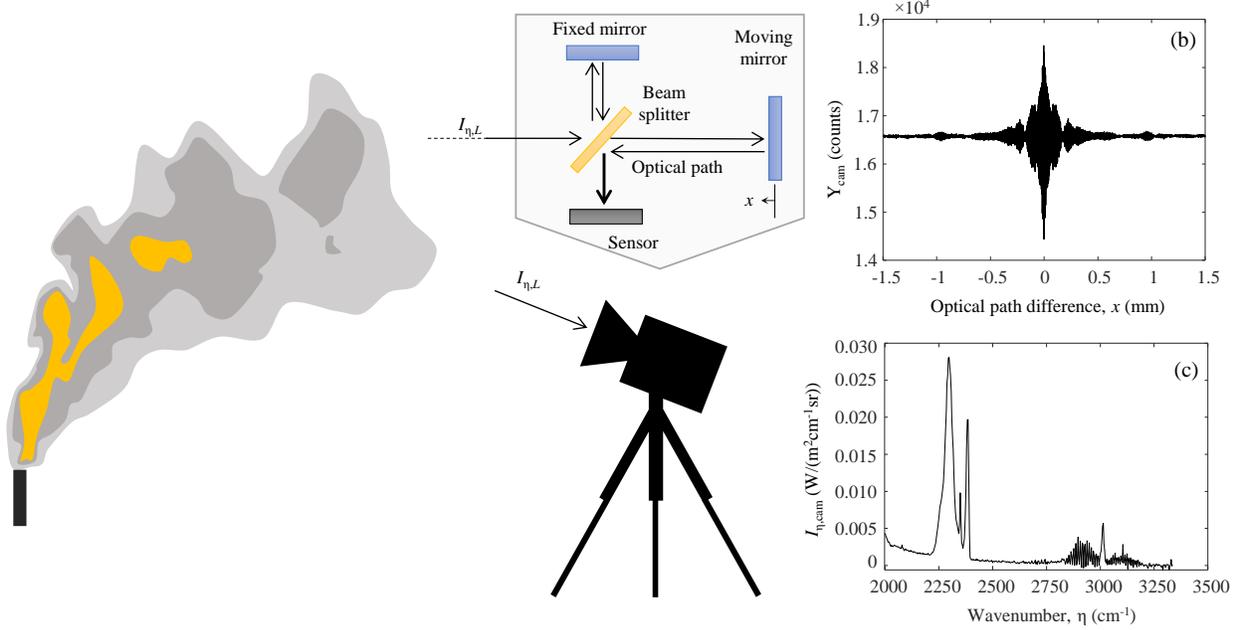
160 where $S_{k,ij}$ is the species spectral line intensity for the transition between the i th and j th energy
 161 states, and f is the Lorentz line shape function. (Doppler broadening is negligible at the
 162 temperatures and pressures relevant to flare plumes.)

163

164 *3.2. Camera response model*

165 Hyperspectral imaging is done using a Telops MW HyperCam IFTS with a detection spectrum of
 166 1900-3400 cm^{-1} , which includes V-R bands of CO_2 , H_2O , CO , and most HCs, and a maximum
 167 spectral resolution of 0.25 cm^{-1} . The focal plane array (FPA) is composed of 320×256 pixels with
 168 an instantaneous field-of-view (IFOV) of 0.35 mrad per pixel. The FPA is made of InSb sensors,
 169 which records the interferogram image generated by a Michelson interferometer as shown in Figure
 170 2 (a). As the moving mirror move across its range, an interferogram Y_{cam} is formed, as shown in
 171 Figure 2 (b). The mirror position defines the optical path difference (OPD), which characterizes the
 172 difference in pathlengths travelled by the two light beams generated by the beam-splitter. The beams
 173 are then recombined, generating interferences that are measured by the FPA to form an interferogram.
 174 A Fourier transform is performed on the interferogram in counts to obtain an intensity spectrum with
 175 arbitrary units. Finally, an absolute intensity spectrum is obtained by calibrating using a two-point
 176 calibration method [21] using two built-in blackbodies maintained at different temperatures.

177 The IFTS camera response model, referred to as the instrument line shape (ILS) function,
 178 relates the scene intensity entering the camera aperture $I_{\eta,L}$ to the measured intensity, and describes



(a)

Figure 2: IFTS measurement process. (a) The scene radiation $I_{\eta,L}$ enters the camera aperture and traverses the Michelson interferometer, generating interferences measured by the FPA InSb sensors for successive mirror position. (b) The measured raw signal is the interferogram Y_{cam} . Finally, (c) the interferogram is Fourier transformed and calibrated by a two-point calibration method to produce the absolute intensity spectra $I_{\eta, cam}$.

179 how the IFTS mirror position limited range affects the measurement. In theory, the interferogram
 180 $Y_m(x)$ is defined for $x \in \mathbb{R}$. However, the range of the moving mirror in the camera is limited to a
 181 maximum OPD, x_{MOPD} , which is related to the spectral resolution by $\delta\eta_{res}=1.20671/2x_{MOPD}$. The
 182 measurement model accounts for the finite range of the mirror and relates the intensity $I_{\eta,L}$ to the
 183 modelled measurement, $I_{\eta,m}$, by the convolution product:

$$184 \quad I_{\eta,m} = \mathcal{F}[Y_m(x) B(x, x_{MOPD})] = \mathcal{F}[Y_m(x)] * \mathcal{F}[B(x, x_{MOPD})] = I_{\eta,L} * ILS(\eta, x_{MOPD}) \quad (7)$$

185 where B is the boxcar function that characterized the finite OPD and becomes the ILS when the
 186 Fourier transformation is applied. Modelling the measured intensity then comes down to a
 187 convolution product between the intensity spectra, given by Eq. (4), and the ILS, which is
 188 $ILS(\eta, x_{MOPD}) = 2x_{max} \text{sinc}(2\pi x_{MOPD} \cdot \eta)$.

189 One of the challenges inherent to hyperspectral measurements of turbulent flows are rapid
 190 variations in intensity caused by fluctuations in the species concentration and temperature, which
 191 occur over a much shorter time scale than the time needed for the mirror to traverse its entire OPD
 192 range. These intensity fluctuations manifest as “scene change artifacts” (SCAs) in the transformed
 193 intensity spectrum [15]. Indeed, the interferogram model assumes that the intensity field remains
 194 constant as the interferogram is assembled:

$$Y_{\text{cam}}(x) = \int_{-\infty}^{+\infty} \frac{1}{2} S_{\eta} [1 + \cos(2\pi\eta x)] d\eta \quad (8)$$

196 where S_{η} is the intensity entering the interferometer, representing the scene intensity $I_{\eta,L}$ and the
 197 camera self emission. The turbulent fluctuations of the flare plumes are expressed in Eq. (8) through
 198 the time dependence of the intensity field $S_{\eta} \equiv S_{\eta}(t)$. Following the hypothesis that the true intensity
 199 field is stationary but fluctuating, SCAs may be suppressed by using a high band pass filter to
 200 isolate the low frequency intensity variations, Fig. 3 (a), and then removing them from the

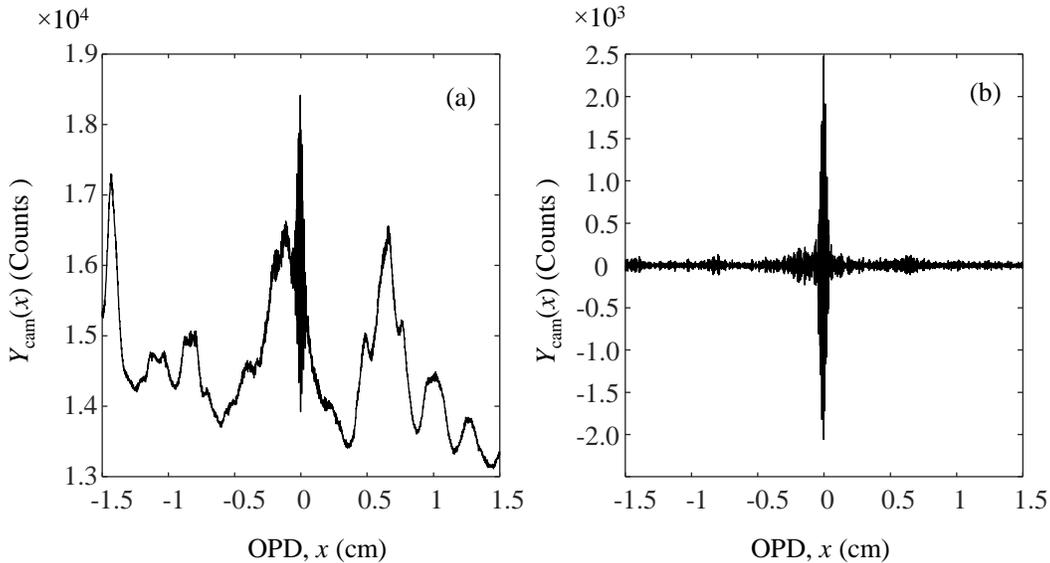


Figure 3: The raw interferograms $Y_{\text{cam}}(x)$ (a) are contaminated by scene change artifacts caused by turbulent fluctuations in the flow field as the mirror traverses through its OPD. These can be removed through a filtering procedure (b).

201 measured interferogram to obtain a new filtered interferogram, Fig. 3 (b). The filtered raw
202 spectrum Y , cleared from the intensity fluctuations, is then transformed to obtain a raw intensity
203 spectrum, S_η . An alternative approach, used in this work, consists of estimating a median
204 interferogram from a large set of raw datacubes.

205

206 **4. Inferring the mass flow rates**

207 Calculating the species mass flow rates from Eq. (2) relies on solving the column densities and the
208 projected velocity profile along the control line. The column densities are obtained from the
209 species mole fractions and plume temperature distributions along the LOS of each pixel on the
210 control line; these quantities, in turn, are found by inverting the spectroscopic measurement model.

211 The image projected velocity field is found from the apparent motion of pixels between sequential
212 broadband images.

213

214 *4.1. Inferring the mole fractions and plume temperature*

215 Inferring the species mole fractions and the temperature profiles along a pixel LOS amounts to
216 minimizing the residual between the measured and modeled spectral intensities for each LOS. This
217 inversion problem is mathematically ill-posed since an infinite combination of $\chi_k(s)$ and $T(s)$ could
218 be substituted into Eq. (4) to generate a given spectral intensity. Accordingly, it is necessary to
219 parameterize the distributions along the pixel LOS to reduce the ill-posedness of the problem.
220 Following the work of Miguel et al. [18], and Grauer et al. [17], we assume Gaussian profiles inside
221 the plume along the LOS, which are appropriate for free turbulent plumes, and constant values for
222 the atmospheric layer between the plume and the camera aperture:

223

$$\phi(s) = \phi_{\text{amb}} + (\phi_{\text{max}} - \phi_{\text{amb}}) \exp\left[-\frac{(s - s_{\text{pc}})^2}{2\sigma_p^2}\right] \quad (9)$$

224 where $\phi = \chi_k$ or T , σ_p is the characteristic plume thickness, and s_{pc} is the location of the plume
 225 center. The ambient concentrations and temperature may be derived from meteorological data, or
 226 determined by examining the spectra from a pixel outside of the flare plume, e.g., the flare stack.
 227 The plume thickness is assumed to be the same for all parameters and is estimated from the plume
 228 image, assuming that the plume is axisymmetric. The plume center is determined by the measured
 229 distance between the camera and the flare stack. Therefore, the number of unknown parameters to
 230 be solved for is N_s+1 , corresponding of the peak concentration of each species and the peak
 231 temperature; these are stored in the vector \mathbf{x} . The maximum likelihood estimate is then found for
 232 each pixel/LOS by

233

$$\mathbf{x}_{\text{MLE}} = \arg \min_{\mathbf{x}} \left\{ [\mathbf{m}(\mathbf{x}) - \mathbf{b}]^T \mathbf{\Gamma}_b^{-1} [\mathbf{m}(\mathbf{x}) - \mathbf{b}] \right\} \quad (10)$$

234 where \mathbf{m} is the measurement model, defined in Sec. 2, \mathbf{b} contains the set of spectral intensities at each
 235 wavelength $\{I_{\eta, \text{cam}}\}$, and $\mathbf{\Gamma}_b$ is the measurement error covariance matrix, which is assumed to be
 236 diagonal and is computed from the measurement variance for each IFTS spectral bin. Minimization
 237 is carried out using the nonlinear least-squares solver in MATLAB[®] with a function tolerance of 10^{-9}
 238 and a step tolerance of 10^{-9} [22]. In some cases, the spectral range of the data is restricted to a subset
 239 of the detection spectrum that contains species-of-interest, in order to reduce computational time and
 240 exclude features that are not included in the model, such as background intensity from the atmosphere.

241

242

243 *4.2. Inferring the velocity field*

244 The time-resolved 2D velocity field, $\mathbf{u} = [u, v]^T$, is derived from a set of successive raw
245 interferometer images via either the Horn-Schunk [23] or Lucas-Kanade optical flow algorithms
246 [24]. The intensity of each image is first normalized and the mean pixel intensity field is subtracted
247 to remove the stationary features. Optical flow is based on the hypothesis that any change in pixel
248 brightness between successive images is due entirely to advection. Accordingly, pixel intensity
249 becomes a conserved quantity and the velocity field is obtained by solving

$$250 \quad \frac{DE}{DT} = \frac{\partial E}{\partial x} u + \frac{\partial E}{\partial y} v + \frac{\partial E}{\partial t} = 0 \quad (11)$$

251 where E is the modified broadband pixel intensity and the derivatives are found from finite
252 difference approximations. Solving for the 2D velocity field requires additional information to
253 close the problem. In the Horn-Schunk algorithm, this is done by assuming that the velocity
254 components vary smoothly in space, which correspond to Tikhonov first-order regulation:

$$255 \quad \begin{cases} \nabla^2 u = \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \\ \nabla^2 v = \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \end{cases} \quad (12)$$

256 The Lucas-Kanade algorithm adopts a weighted window scheme to filter the velocity field
257 calculation, thereby suppressing noise amplification by the numerical derivative. In that case, the
258 weighting term is the equivalent to first-order Tikhonov regularization. The regularization
259 parameters are usually chosen heuristically, as is done here.

260

261

262 **5. Experimental analysis**

263 The technique is demonstrated by conducting measurements on a laboratory-scale heated vent,
 264 combustor, and a steam-assisted petrochemical flare.

265 *5.1. Laboratory-scale heated vent*

266 The performance of this technique is first benchmarked using the heated vent apparatus shown in
 267 Figure 4 (a), consisting of a 1.9 cm diameter nozzle positioned in front of four aluminum plates
 268 (30 cm × 30 cm each), coated with matte black paint (Krylon k01602 Ultra-flat black) and
 269 maintained at a set-point temperature of 281 K by a water/ethylene glycol mixture. A specified
 270 47:53 vol. mixture of CH₄ (53%) and CO₂ (47%) is generated using mass flow controllers (Brooks
 271 SLA5850 for CO₂ and Brooks GF40 for CH₄) at a total flow rate of 25.5 standard litres per minute

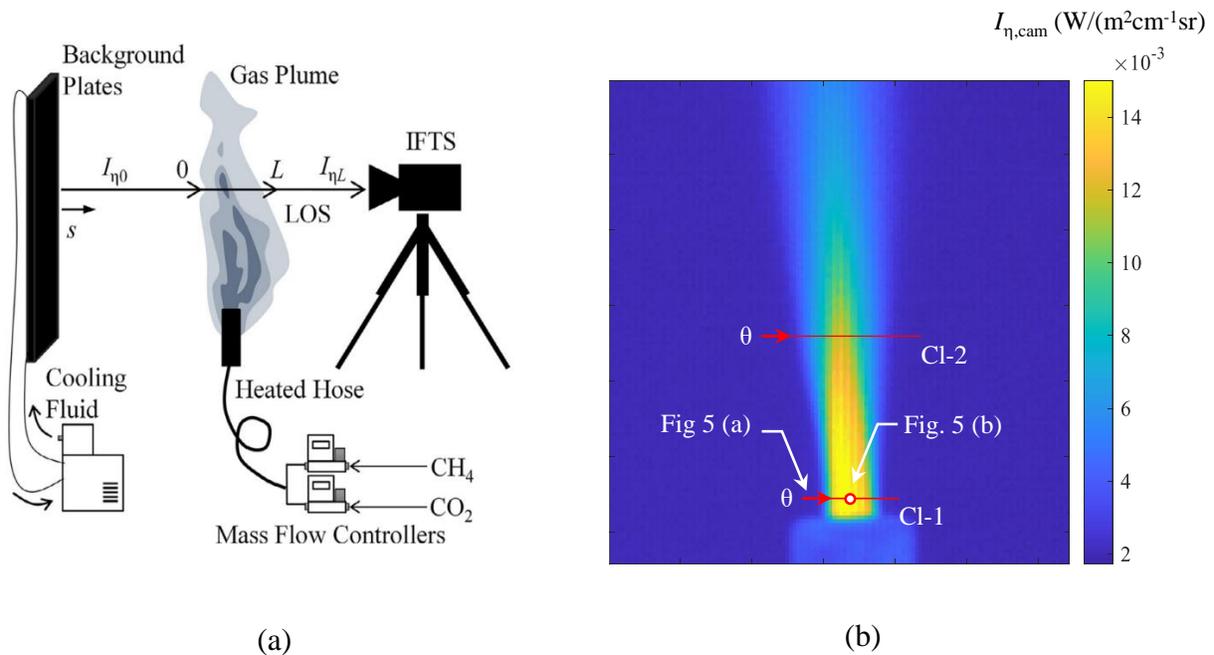


Figure 4: (a) laboratory scale apparatus, the hyperspectral camera is aiming at the gas plume. (b) The camera generates infrared images of the emitted radiation from the plume. The analysis is conducted over two control lines, CI-1 near the stack exit and CI-2 located 6.6 cm above the stack exit.

272 (SLPM). The gas mixture then flows through a heated hose before it is discharged from the nozzle.
273 The gas temperature is maintained at 350 K via a K-type thermocouple located at the nozzle exit.

274 Hyperspectral measurements were conducted with the camera aperture located 420 cm away
275 from the plume, using a 128 px \times 128 px subdomain of the FPA. The detection spectrum was set
276 between 1,900 cm^{-1} and 3,400 $^{-1}$ with a spectral resolution of 4 cm^{-1} and a fixed integration time of
277 100 μ s. However, during the inference, only a subset of the spectrum was used, containing the
278 species-of-interests absorption spectral ranges, from 2200 to 2470 cm^{-1} for CO_2 and from 2800 to
279 3300 cm^{-1} for CH_4 . The ambient CO_2 concentration was assumed to be 420 ppm and the
280 temperature of the atmospheric layer between the plume and the camera was measured at 295 K.
281 Inferring the ambient temperature from background pixels led to $T_{\text{amb}} = 297$ K, which is in good
282 agreement with the measurement.

283 The control line CL-1 was positioned near the stack exit, as shown in Figure 4 (b), so the gas
284 velocities could be estimated from the known mass flow rates and nozzle dimensions. Because the
285 control line is located above the stack exit, the velocity and concentration profiles are assumed to
286 be uniform along the control line region covering the stack nozzle. The absence of turbulent flow
287 features at the nozzle exit precludes calculating the gas velocity from optical flow, so the gas exit
288 velocity is estimated to be 1.5 m/s from the known mass flow rate and the nozzle diameter.
289 Therefore, calculating the mass flow rate at this position provides a means to isolate how
290 uncertainty in the spectroscopic model impacts the mass flow rate.

291 Figure 5 (a) shows the column density profiles for CO_2 and CH_4 , as well as the inferred plume
292 temperature for the control line “CL-1” in Fig. 4 (b). The inferred ambient temperature (297 K)
293 was used for this calculation. The plume temperature inferred from the IFTS data is around 400 K
294 instead of the measured 350 K at the stack exit. The column density profiles of CO_2 and CH_4 show

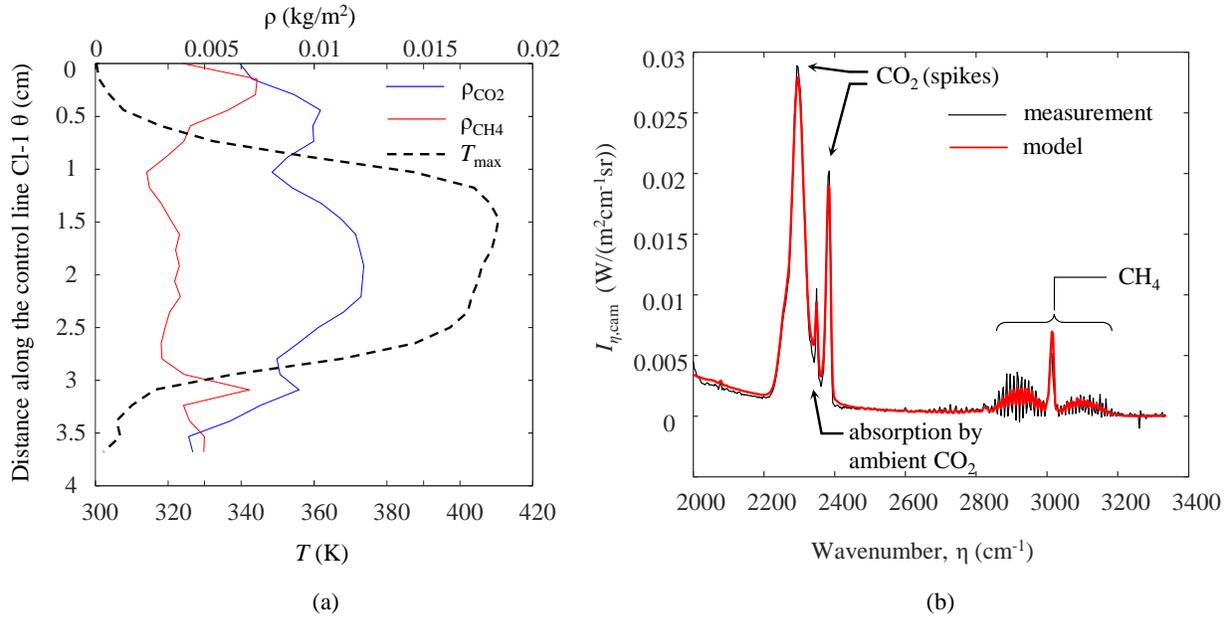


Figure 5: (a) Column densities and temperatures inferred from the IFTS camera data for the integration line Cl-1 shown in Fig. 4 (b). (b) Comparison of modeled and measured spectra at the pixel location shown in Fig. 4 (b).

295 spikes at the extremities of the control line, which is due to the fact that the contribution of thermal
 296 emission from the gases to the detected intensities is small at this location, and the inference
 297 becomes dominated by measurement noise and model error. Since the values do not cover the
 298 plume, they are excluded from the mass flow rate estimations, which is conducted from $\theta = 0.7$
 299 cm to $\theta = 2.9$ cm. Figure 5 (b) shows that the modeled spectrum at the sample point indicated in
 300 Fig. 4 (b) closely matches the measured intensity. Features for both CO_2 and CH_4 are visible at
 301 $2,200\text{-}2,400\text{ cm}^{-1}$ and $2,800\text{-}3,200\text{ cm}^{-1}$, respectively. The measured emission spectra from the
 302 plume reveals two CO_2 emission “spikes” at $2,250$ (“red spike”) and $2,400\text{ cm}^{-1}$ (“blue spike”).
 303 This feature arises from the fact that a portion of emission from 001 fundamental CO_2 band in the
 304 plume is absorbed by the ambient CO_2 between the plume and the camera, only leaving the hot
 305 lines at the wings of the band.

306 The control line Cl-2 was positioned 6.6 cm above the stack exit, as shown in Figure 4 (b), so
 307 the gas velocities are inferred from the Horn-Schunk optical flow algorithm. Gaussian profiles for
 308 the plume concentrations and temperature are assumed due to the turbulent nature of the plume at
 309 that location (see Eq. (9)). The inferred column density profile for CO₂ and CH₄ are shown in
 310 Figure 6 (a), as well as the maximum temperature distribution along Cl-2. Figure 6 (b) shows the
 311 corresponding velocity profile used to estimate CO₂ and CH₄ mass flow rates. The mass flow rates
 312 are computed from $\theta = 0.6$ cm to $\theta = 5$ cm to exclude the background pixels from the mass flow
 313 rate calculation.

314 Table 1 shows that the inferred mass flow rate for CO₂ and CH₄, along Cl-1 and Cl-2, are in
 315 good agreement with the mass flow controller settings.

316 Table 1: CO₂ and CH₄ carbon mass flow rates inferred from the heated vent.

	IFTS (Cl-1)	IFTS (Cl-2)	MFC	% diff. (Cl-1)	% diff. (Cl-2)
\dot{m}_{C,CO_2} ($\times 10^2$ g/s)	8.9	8.4	8.37	5.9	0.3
\dot{m}_{C,CH_4} ($\times 10^2$ g/s)	8.1	9.6	9.45	-13.9	1.6

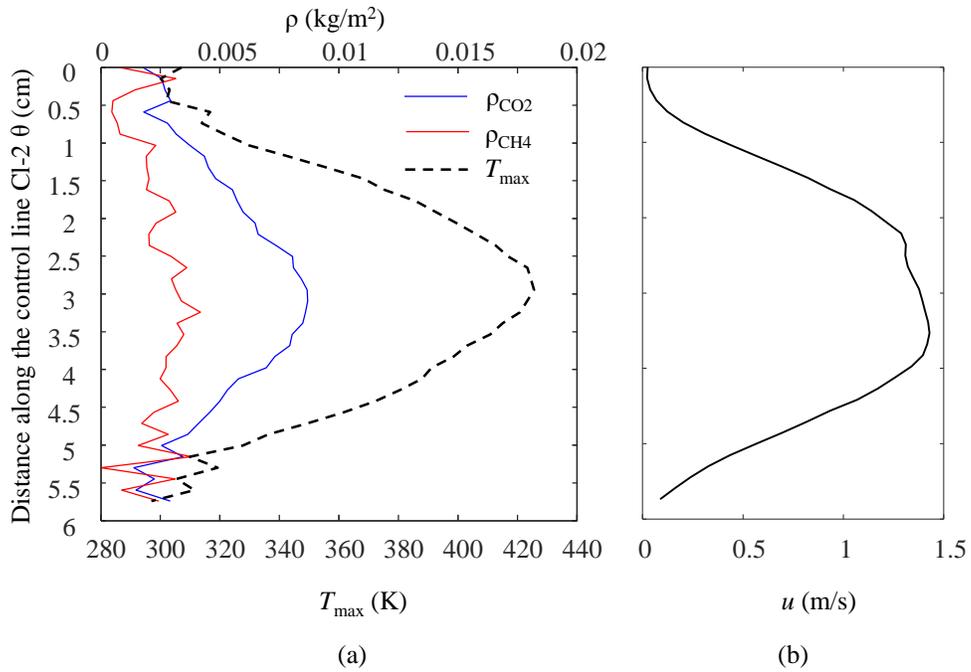


Figure 6: (a) Column densities and temperatures inferred from the IFTS camera data for the integration line Cl-2 shown in Fig. 4 (b). (b) Velocity profile inferred from the Horn-Schunk optical flow algorithm.

317

318

319 5.2. *IFTS measurement of a combustor*

320 Figure 7 (a) shows a Questor Q100 combustor that was operated at Carbon Management Canada's
321 Newall County Research Station. The combustor was supplied with natural gas (~94.2% CH₄,
322 3.4% C₂H₆, 1.1% C₃H₈, and 1.3% minor components by volume) at approximately 42.5 standard
323 cubic meters per hour. Local meteorological conditions (windspeed, wind direction, and
324 temperature) were measured by a Davis onsite weather station, while ambient CO₂, CH₄, and H₂O
325 concentrations were measured by a Picarro cavity ringdown spectrometer.

326 Figure 7 (b) shows the spectral intensity map at $\eta = 2283 \text{ cm}^{-1}$, corresponding to the CO₂ "red
327 spike." Measurements were conducted at a distance of 150 m from the combustor stack exit, using
328 a 128 px×128 px subdomain of the FPA, corresponding to a (6.6×6.6) m² area of the scene. The
329 detection spectrum was set between 1900 cm⁻¹ and 3400 cm⁻¹ with a spectral resolution of 4 cm⁻¹ and
330 a fixed integration time of 50 μs. Figure 8 shows the intensity spectra for pixels "A" above, "B"
331 within, and "C" beneath the plume, along with the absorption coefficients of species that may be
332 in the plume (H₂O, CO, CO₂, CH₄). The spectra for pixels "A" and "B" show blackbody-like
333 behavior at low wavenumbers corresponding to an average temperature of approximately 255 K
334 under clear conditions. As one would expect, the spectral intensity for the plume pixel "B" is much
335 larger than those of "A" and "C" due to the thermal emission of the hot species. The intensity
336 spectra indicate the presence of CO₂, H₂O, CO, but shows no detectable CH₄, suggesting that the
337 combustor is operating with a high CE. Note also that the red and blue CO₂ spikes are more
338 pronounced compared to Fig. 5, due to the greater column density of ambient CO₂ between the
339 camera and the plume.

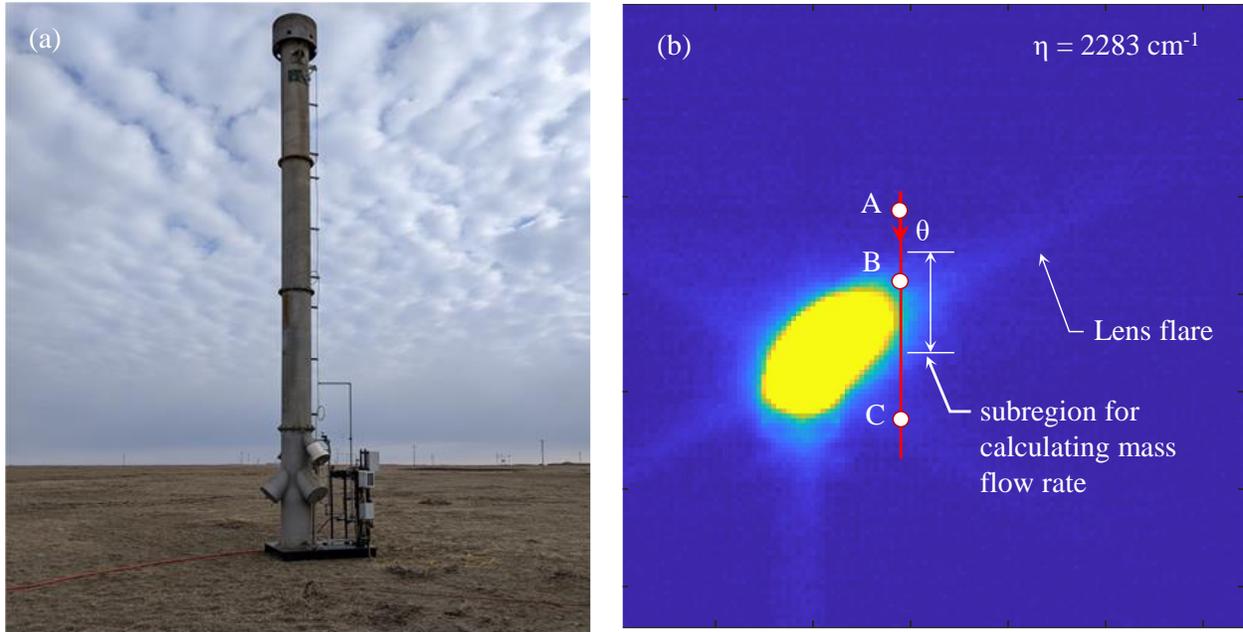


Figure 7: (a) Questor Q100 combustor; and (b) the intensity map at $\eta = 2283 \text{ cm}^{-1}$, corresponding to the CO₂ red spike. Spectra for locations A, B, and C along the integration line are shown in Fig. 8. Lens flare artifacts extend diagonally from the region of peak intensity.

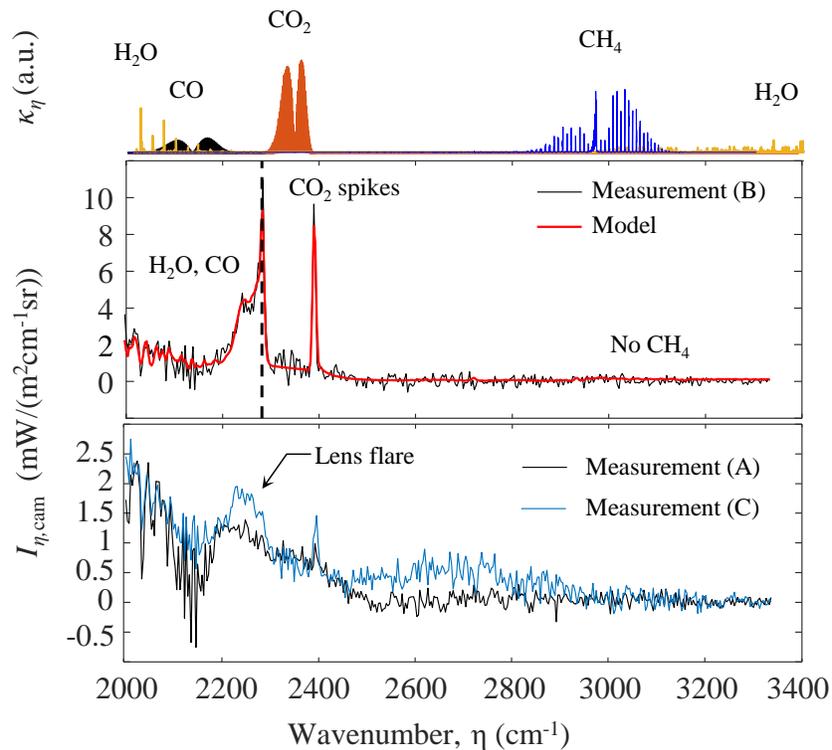


Figure 8: Spectra for pixels “A”, “B”, and “C” shown in Fig. 7. Note the difference in scale between the top and the bottom plots.

341 The infrared intensity map in Fig. 7 (b) also reveals lens flare artifacts, which extend diagonally
 342 from the brightest spot on the image. These artifacts affect all of the broadband images used to
 343 construct the interferogram, and are therefore difficult to remove through image post-processing.
 344 The impact of lens flare on the recovered spectrum is highlighted in the bottom panel of Figure 8,
 345 where the spectra from pixels “A” and “C” are superposed. The intensity spectra from pixel “C”
 346 shows two small CO₂ spikes, as we would expect inside the plume but not outside the plume. It
 347 also shows an increase of intensity at 2,150 cm⁻¹ corresponding to CO emission lines. From the
 348 spectra at pixel “C”, it is difficult to identify that those spectral signatures come from an optical
 349 artifact and not from species emissions. This artifact will cause the CO and CO₂ column densities
 350 of these pixels to be over-estimated.

351 The “hole” between the red and blue CO₂ spikes for highlights the importance of accounting
 352 for the atmospheric layer between the flare plume and the camera in the spectroscopic model.
 353 Ambient conditions can often be inferred from the hyperspectral data by selecting pixels from the
 354 flare stack region. In this particular measurement, however, the stack pixels are strongly affected
 355 by the lens flare, so onsite concentrations of CO₂ and H₂O and temperature are used instead (Table
 356 2). Given the long pathlength between the flare plume and the camera, it is crucial to include the
 357 ¹³C¹⁶O₂ isotopologue, which has a natural abundance of approximately 1% and a red-shifted
 358 absorption spectrum compared to that of the main ¹²C¹⁶O₂ isotopologue.

359 Table 2: Ambient conditions set for the combustor hyperspectral measurement

Parameter	Onsite measured values
CO ₂	426 ppm ¹
H ₂ O	2914 ppm (RH = 44%) ¹
<i>T</i> _{amb} (K)	289 K ²

¹Picarro station

²Davis weather station

360 Figure 9 shows the column density profiles for of CO₂ and CO, inferred using Eq. (3), the
 361 plume temperature, as well as the velocity profile found from the Lucas-Kanade algorithm. Since
 362 there were no CH₄ features in the intensity spectra, a subregion of the detection spectrum, between
 363 2000 and 2470 cm⁻¹, was used for the inference, and a blackbody spectrum corresponding to 255
 364 K is assumed for the background.

365 During the field trial wind speed was approximately 2.5 m/s during the measurement period,
 366 which is in good agreement with the peak velocity inferred from the optical flow algorithm. The
 367 velocities and column densities along the control line are used to infer the species mass flow rate,
 368 Eq. (2), and then multiplied by their respective carbon mass ratios (CO₂: 12/44, CO: 12/28) to
 369 obtain the carbon mass flow rate and estimate the combustion efficiency via Eq. (1). Figure 9
 370 shows that the column densities increase drastically outside the plume compared to their densities

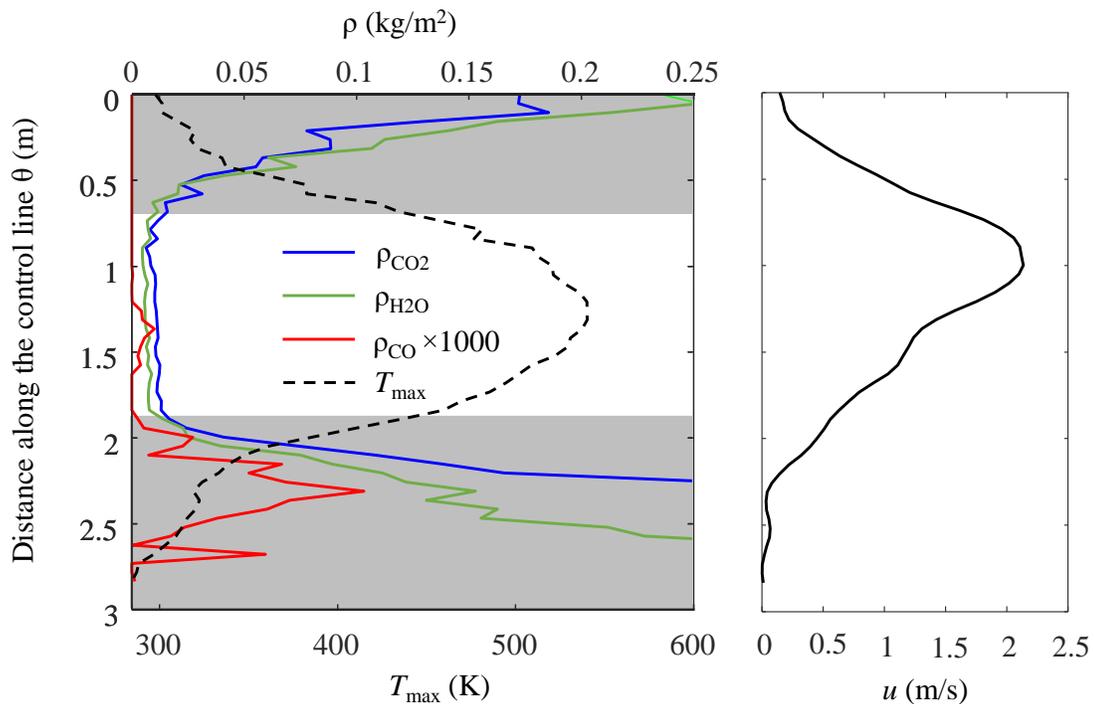


Figure 9: Column densities and peak temperature inferred from the spectroscopic model, and velocity profile inferred from the optical flow algorithm, for the combustor shown in Fig. 6. The shaded regions are excluded from the mass flow rate calculation.

371 inside the plume ($0.7 \text{ m} < \theta < 1.9 \text{ m}$). This behaviour is caused by the lens flare effects depicted in
 372 Fig. 7 (b), and of the mischaracterization of the background intensity when modelled by Eq. (4).
 373 Quantification of the species mass flow rate and of combustion efficiency is therefore conducted
 374 only for pixels covering the plume. Results are summarized in Table 3. When only considering the
 375 part of the control line that crosses the plume ($0.7 \text{ m} < \theta < 1.9 \text{ m}$), as labeled in Fig. 9, the
 376 combustor carbon mass flow rate is estimated at $\dot{m}_C = 5.8 \text{ g/s}$, which is in good agreement with the
 377 known natural gas input carbon mass flow rate of 6.1 g/s (42.5 SCM of natural gas per hour). This
 378 corresponds to a CE greater than 99%.

379

380 Table 3: CO₂ and CO mass flow rates of the combustor, inferred using the IFTS.

Species, k	$\dot{m}_{C,k}$ (g/s)
CO ₂	5.8
CO	~ 0

381

382 *5.3. IFTS measurement of a steam-assisted flare at a refinery*

383 A key application for this technique lies in conducting fenceline measurements on flares that burn
 384 unknown fuel compositions and at unknown rates, as is the case of a steam-assist flare at a
 385 petrochemical refinery in Southern Ontario, shown in Fig. 10.

386 A $128 \text{ px} \times 128 \text{ px}$ subdomain of the FPA was used to image the flare, which corresponds to a
 387 $(22.4 \times 22.4) \text{ m}^2$ area for a measurement distance of 580 m. The spectral resolution was set to 4
 388 cm^{-1} . With these parameters and an integration time of 100 μs per image, the imaging framerate
 389 was 747 Hz, which required 3.17 s to form a complete interferogram. Figure 11 (a) shows the
 390 spectral intensity map for 2960 cm^{-1} , a part of the spectra that contains features corresponding to
 391 the principle C-H stretching mode of many hydrocarbons, while Figure 11 (b) shows the spectral

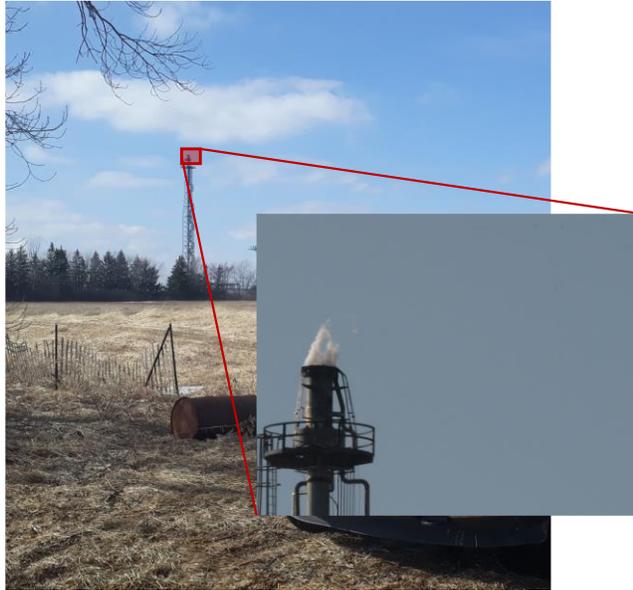


Figure 10: Steam assisted flare at a refinery. Measurements were carried out at a standoff distance of approximately 580 m.

392 intensity map for another wavenumber, $\eta = 2224 \text{ cm}^{-1}$, corresponding to the combustion products
393 CO and CO₂. For that wavenumber, the map illustrates the lens flare artefacts and the flare halo
394 that affect the infrared image. As the lens flare transects the control line pixels, this artifact will
395 interfere with the inversion procedure and the column densities estimates.

396 Figure 12 (b) shows the intensity spectra for pixels above and within the plume (“A”, “B”) and
397 for a pixel at the plume extremity (“C”), along with absorption coefficients for H₂O, CO₂, and CO,
398 all of which are expected to be in the flare plume. The intensity spectra indicate the presence of
399 CO₂, H₂O, CO, while the feature between 2800-3000 cm⁻¹ visible in the spectra for pixels “B” and
400 “C” indicates the presence of an unknown hydrocarbon. In the case of flares used to dispose of
401 process gas for upstream oil and gas production this signal would likely be CH₄, but the absence
402 of the R-branch lines at higher wavenumbers indicates the presence of a heavier hydrocarbon. The

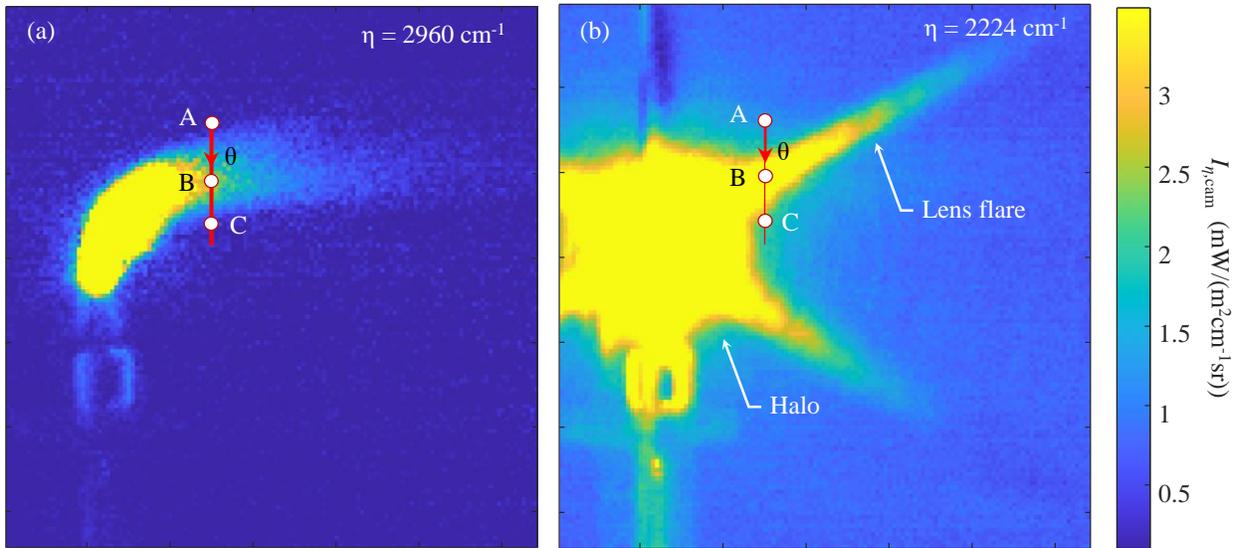


Figure 11: (a) Intensity map at 2224 cm^{-1} of the flare shown in Fig.10; this wavenumber corresponds to CO and CO₂ emission lines. For that wavenumber, lens flare and glowing of the flare is observed on the image. (b) Intensity map at 2960 cm^{-1} of the flare shown in Fig. 10; this wavenumber corresponds to the principle C-H stretching mode of hydrocarbons. The National Pollution Release Inventory suggests that the flare is likely burning butane, which is consistent with the detected spectra (Fig. 12 (b)).

403 National Pollutant Release Inventory for this facility [25] suggests that the HC is butane, C₄H₁₀.
 404 Like many heavier hydrocarbons, line parameters for butane are not available in the HITRAN
 405 database. Instead, a comparison of the experimentally-derived molecular absorption cross-section
 406 [20, 26] and the IFTS emissions spectrum confirms that this emission feature likely corresponds
 407 to butane. This empirically-derived absorption spectrum is incorporated into the spectroscopic
 408 model. This result highlights the importance of spectral and spatial resolution when calculating
 409 flare CE, particularly for downstream oil and gas operations since the absorption cross-sections of
 410 hydrocarbons generally do not scale with the number of carbons per molecule.

411 As mentioned for the combustor measurements, the spectra in Fig. 12 (b) corresponding to
 412 pixels “B” and “C” feature two CO₂ emission “spikes” caused by emission the hot CO₂ in the flare
 413 plume and its absorption by the ambient CO₂. For each pixel “A”, “B” and “C”, the spectra show

414 an increase in intensity around wavenumber 2224 cm^{-1} that correspond to the halo and lens flare
 415 artefacts. For pixels inside the plume like “B” and “C”, the model attempt to recover that fictitious
 416 signal by an increase in the CO_2 concentration. In Figure 12 (a), are represented the column
 417 densities and plume maximum temperature along the control line. The CO_2 column density profile
 418 is increasing for pixels located at the plume extremities (e.g. pixel “C”), illustrating the effect of
 419 lens flare and the overcompensation of the model (spectra “C” in Fig. 12 (b)). The CO_2 and C_4H_{10}
 420 spikes at the control line top and bottom pixels can be explained by those artefacts but also by the
 421 mischaracterization of the background, as shown by the intensity spectrum “A” in Fig. 12 (b).

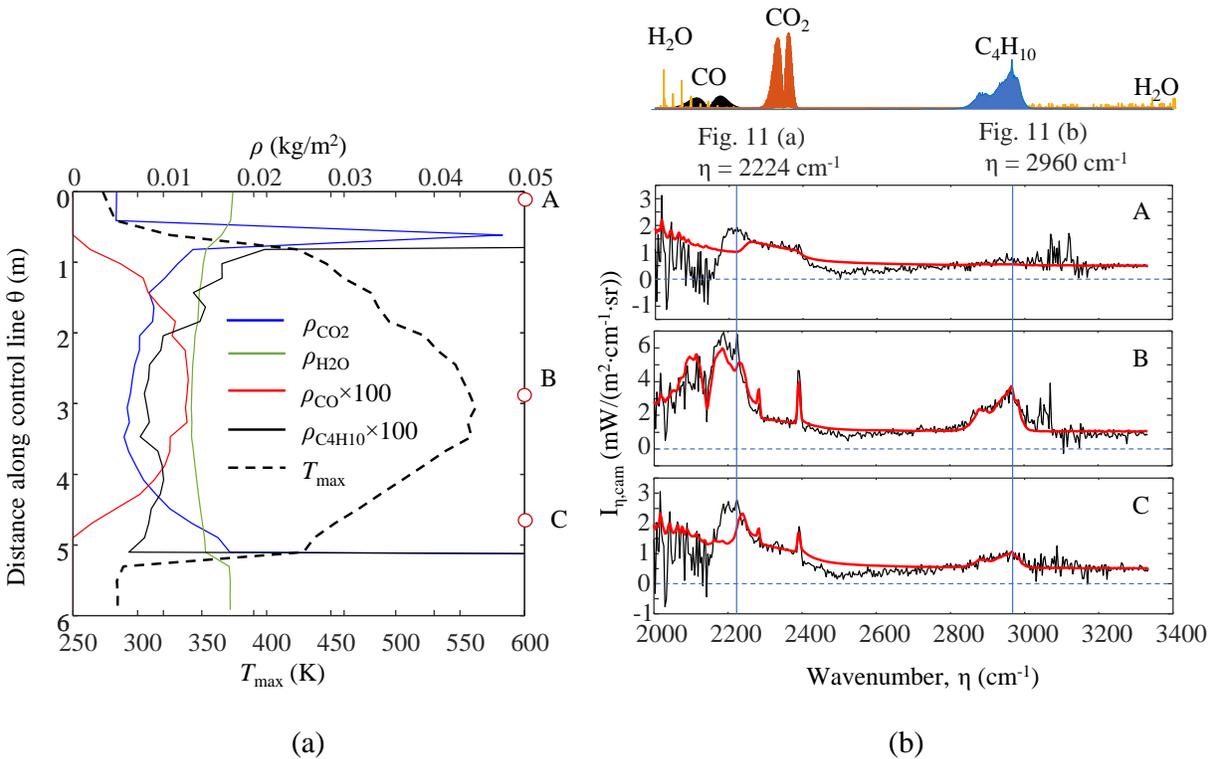


Figure 12: (a) Column densities and peak temperature inferred along the control line shown in Fig. 11. The spikes in column density at the extreme ends of the integration line are likely caused by lens flare artifacts and mischaracterization of the background. The positions “A”, “B” and “C” labeled along the control line correspond to the ones in Fig. 11; (b) Intensity spectra for pixels at positions “A”, “B” and “C”, along with the absorption coefficient of candidate species. Blue lines for wavenumbers corresponding to Fig. 11 images, show the emission from butane ($\eta = 2960\text{ cm}^{-1}$) as well as the lens flare and glowing effect on the spectra ($\eta = 2224\text{ cm}^{-1}$).

422 The ambient CO₂ and H₂O concentration and temperature needed to interpret the spectra are
423 shown in Table 4. Apart from the background and lens artefact, the modeled spectra are in
424 excellent agreement with the measured spectra.

425

426 Table 4: Ambient conditions during the hyperspectral measurement of the petrochemical flare.

Parameter	Value
CO ₂	413 ppm ¹
H ₂ O	3666 ppm (RH = 60% ²)
T_{amb} (K)	280 K ³ (280.3 K ²)

¹Based on average atmospheric abundance

²Environment Canada

³Onboard thermocouple

427

428 The velocities are found from the Lucas-Kanade algorithm, with a maximum normal velocity
429 of 4.5 m/s. Column densities and velocities are substituted into Eq. (2) to obtain the mass flow
430 rates shown in Table 5. Finally, these mass flow rates are multiplied by their respective carbon
431 mass ratios (CO₂: 12/44, CO: 12/28, C₄H₁₀: 48/58) and then substituted into Eq. (1) to obtain an
432 overall flare combustion efficiency of 97%.

433 Table 5: CO₂, CO, and C₄H₁₀ carbon mass flow rates for the petrochemical flare.

Species, k	$\dot{m}_{\text{C},k}$ (g/s)
CO ₂	27.2
CO	0.5
C ₄ H ₁₀	0.3

434

435 6. Conclusions and future work

436 This study presents a technique for estimating flare combustion efficiency using a MWIR HS
437 camera. The mass flow rates needed for this calculation are found by combining species column
438 densities inferred using a spectroscopic model with a 2D velocity field obtained using an optical

439 flow algorithm. Crucially, this technique provides the capacity to infer flow rates and flare
440 efficiency through fenceline measurements, without requiring any knowledge about the fuel flow
441 rate supplied to the flare. This tool may be used by operators, flare equipment manufacturers, and
442 regulators to assess the combustion efficiency of flares *in situ* through standoff measurements.
443 This information is crucial to understand and minimize the impact of flaring on climate change.
444 While the approach focused on calculating CE, it may also be used to quantify emissions of other
445 types of flare emissions, including black carbon [27] and sulphur dioxide [28]. The combined
446 spectral and spatial resolution afforded by the IFTS is particularly well-suited in cases where the
447 species may be distributed heterogeneously in the flare plume and in cases where the species
448 present in the plume may be unknown or uncertain.

449 Despite the strong potential of this technique, there are a number of challenges that must be
450 overcome in order to improve the reliability of this approach. First and foremost, it is necessary to
451 quantify the uncertainties attached to the column densities and mass flow rates used to estimate
452 the CE, and to carry out benchmark validation measurements using an independent measurement
453 technique. The inversion of the spectroscopic model is deeply ill-posed, since an infinite set of
454 concentration and temperature distributions exist that, when substituted into Eq. (4), could explain
455 the spectroscopic data. The imposition of Gaussian profiles may be reasonable in a time-averaged
456 sense, but the instantaneous intensity field may be influenced strongly by turbulent fluctuations
457 between the concentration and temperature field (so-called “turbulence-radiation interactions”
458 [29]). A second source of model error comes from the use of intensity-weighted projected
459 velocities obtained from the optical flow algorithm. When the intensity variations are caused by
460 the absorption of background radiation it can be shown that the projected velocity field is a “mass-
461 weighted velocity”, which is appropriate for calculating a mass-flow rate [30, 27]. In this case,

462 however, the intensity variations are due to coupled fluctuations in local temperature and
463 concentration, and the resulting velocity field may not be mass-weighted. In both regards, working
464 with Bayesian inversion methods could help with both of those objectives, as it formalizes the
465 inclusion of prior information and account for quantification of the measurement and model
466 uncertainty propagation to the estimates.

467 There also remain several issues related to hardware implementation. Hyperspectral images of
468 the combustor and petrochemical flares were contaminated with lens flare features, likely caused
469 by coherent scattering of the most intense source of incident intensity (e.g., hot CO₂) by defects in
470 the optics. These artifacts are difficult to remove since they contaminate the broadband images
471 that are Fourier-transformed into the recovered spectra. Turbulence-induced SCAs and lens flare
472 both impact the intensity spectrum, and therefore the inferred temperature and species column
473 densities. While the impact of SCAs may be mitigated through filtering, flare artifacts are
474 considerably more challenging to remove. Future work will focus on developing image processing
475 techniques to remove lens flare, and experimental procedures for avoiding these features in the
476 first place.

477 **Data availability statement**

478 The data that support the findings of this study are openly available in “Replication Data for:
479 Quantifying Flare Combustion Efficiency using an Imaging Fourier Transform Spectrometer” at
480 <https://doi.org/10.5683/SP3/JLMYWE>.

481 **Acknowledgments**

482 This research was sponsored by NSERC's FlareNet (NETGP 479641-15). The authors are grateful
483 to Defence Research and Development Canada for providing their Imaging Fourier Transform
484 Spectrometer.

485

486 **References**

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