



Gaussian model for emission rate measurement of heated plumes using hyperspectral data



Samuel J. Grauer^{a,*}, Bradley M. Conrad^b, Rodrigo B. Miguel^a, Kyle J. Daun^a

^a Department of Mechanical and Mechatronics Engineering, University of Waterloo, 200 University Avenue West, Waterloo, ON N2L 3G1, Canada

^b Department of Mechanical and Aerospace Engineering, Carleton University, 1125 Colonel By Drive, Ottawa, ON K1S 5B6, Canada

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ABSTRACT

This paper presents a novel model for measuring the emission rate of a heated gas plume using hyperspectral data from an FTIR imaging spectrometer. The radiative transfer equation (RTE) is used to relate the spectral intensity of a pixel to presumed Gaussian distributions of volume fraction and temperature within the plume, along a line-of-sight that corresponds to the pixel, whereas previous techniques exclusively presume uniform distributions for these parameters. Estimates of volume fraction and temperature are converted to a column density by integrating the local molecular density along each path. Image correlation velocimetry is then employed on raw spectral intensity images to estimate the volume-weighted normal velocity at each pixel. Finally, integrating the product of velocity and column density along a control surface yields an estimate of the instantaneous emission rate. For validation, emission rate estimates were derived from synthetic hyperspectral images of a heated methane plume, generated using data from a large-eddy simulation. Calculating the RTE with Gaussian distributions of volume fraction and temperature, instead of uniform distributions, improved the accuracy of column density measurement by 14%. Moreover, the mean methane emission rate measured using our approach was within 4% of the ground truth. These results support the use of Gaussian distributions of thermodynamic properties in calculation of the RTE for optical gas diagnostics.

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1. Introduction

Gaseous emissions impact human health and safety, ecological stability, and long-term climate patterns. For instance, aerosolized SO₂ and NO₂ produced by fossil fuel power stations are converted to sulfuric and nitric acids, which further decompose into toxic compounds [1]. Sulfuric aerosols cause human respiratory morbidity; and subsequent acidic products damage nearby soil [2]. Power stations must therefore scrub and monitor effluent gas flows to minimize these effects [3]. Moreover, anthropogenic sources of CO₂, CH₄, and NO₂ contribute positive radiative forcing due to the greenhouse effect and feedback mechanisms [4]. CH₄ and NO₂ are of particular interest, as they have a mass-based greenhouse warming potential that is 86 and 268 times greater than that of CO₂ over a 20-year period, respectively [5]. As a result, the EPA has issued regulations, to decrease emissions of CH₄, NO₂, volatile organic compounds, and other toxic molecules [6]. Many nations have enacted similar rules to meet global climate commitments,

which frequently mandate that polluters monitor their emissions to ensure compliance. Increased effort is therefore devoted to the development of emission diagnostics: to quantify the release of hazardous gasses, inform our fundamental understanding of turbulent plume dispersion, and develop targeted mitigation strategies [7].

Several key factors complicate probe-based measurements of emissions from industrial chimneys, flare stacks, and landfills. Chimneys and flare stacks are typically elevated, and feature dangerous compounds and high-temperature gas flows, which pose safety risks and restrict physical access to emissions [8]. While landfill hydrocarbon emissions are more readily accessible, they are distributed across large areas and feature considerable variation over space and time [9]. Physical sampling through probes provides an important source of information for these scenarios, but the accuracy of emission fluxes inferred from point concentration measurements is limited by the inherent locality of these measurements and sensor uncertainty, among other issues [10].

Advances in spectroscopy and opto-electronic engineering have enabled cost-effective, stand-off measurement of gas plumes using optical instruments [11]. Optical diagnostics are categorized as active devices, which require illumination, and passive devices,

* Corresponding author.

E-mail address: sgrauer@uwaterloo.ca (S.J. Grauer).

which employ background illumination or thermal emission from a heated gas. Line-of-sight (LOS) techniques include differential absorption light detection and ranging [9,12], tunable diode laser absorption spectroscopy [13], and UV [14] and Fourier-transform IR (FTIR) spectroscopy [15] (see [16] for a comprehensive review of these techniques). Spectroscopy-based diagnostics exploit the unique spectral characteristics of target molecules to provide accurate path-integrated concentration data, and can realize accuracies below 1 ppm-m [17]. LOS data is used in conjunction with plume mapping to identify the source of emissions and estimate concentration distributions over a large area [18]. However, extrapolation of single-path measurements yields uncertainties similar to those of maps derived from probe data. Broadband [19,20] and multi-spectral [21–23] IR imaging capture one or more 2D images of spectrally-integrated intensity, which enable the instantaneous estimation of column density data throughout a field of view (FOV). This approach, called gas correlation imaging, can be combined in parallel with image correlation velocimetry to calculate the total emission rate [24]. The number of target species that can be resolved by gas correlation methods is governed by the number and spectral range of measurement bands; and estimate accuracy decreases in the presence of multiple absorbing species.

Hyperspectral gas detection is an emerging technique, based on 2D images with spectrally-resolved data for each pixel, called a *hypercube*. Hyperspectral diagnostics incorporate the advantages of both LOS spectroscopy and gas correlation imaging [25]. Cameras that generate hyperspectral images for remote sensing typically couple an imaging Fourier-transform spectrometer with an IR focal plane array. Fourier transformation of the interferogram produced by this combination yields a hypercube. Remote sensing of emissions by this method has been conducted for plumes containing CO_2 , CH_4 , H_2O , NO_2 , NO , SO_2 , HCl , C_2F_6 , CHF_3 , CO , and R-134a [26–32]. Savary et al. [33] proposed that hyperspectral imaging be applied to monitor flare combustion efficiency, improving on the multispectral approach of Zeng et al. [23]. And recent algorithmic development has focused on the robust detection of multiple species [34], including Bayesian methods [35]. Gålfalk et al. [36] summarize state-of-the-art remote sensing of gas plumes by hyperspectral imaging, and present a novel algorithm for multi-species emission rate estimation. Each of these studies assumes a uniform distribution of temperature and concentration throughout a column of gas. By contrast, Ren and Modest [37] proposed an algorithm to estimate the spatial distribution of temperature along a single LOS for a gas of known composition. The authors show that, given a single perspective, spectral data is a crucial input to inference of the gas state along the optical axis. However, their method requires full knowledge of the gas composition, and is not generally applicable in the remote sensing context.

This paper reports a novel model for quantification of heated, gas-phase emissions using hyperspectral images, such as those needed to carry out field-measurements of flare combustion efficiency. We assume Gaussian distributions of concentration and temperature along a LOS to model spectral intensity measurements, as opposed to the spatially-uniform gas column that has been universally employed to date [26–36]. First, we estimate a characteristic plume width along each LOS from broadband IR data. Next, using this estimate, in conjunction with hyperspectral data, we perform a nonlinear regression to identify peak values of volume fraction and temperature for each pixel. The resulting thermodynamic state distributions are used to calculate column density, and image correlation velocimetry (ICV) is used to obtain the volume fraction-weighted normal velocity along a control surface that transects the plume. Finally, integration of the product of column density and normal velocity along this control surface yields a total mass emission rate. The procedure is executed in a

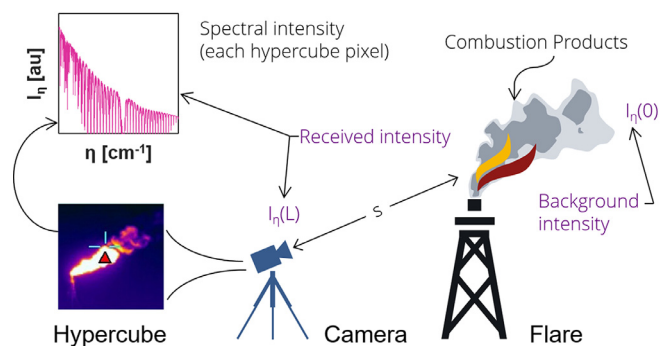


Fig. 1. Schematic for hyperspectral imaging of flare emissions. The camera generates a hyperspectral image of the flare, in which each pixel contains spectrally-resolved intensity data. This data is used to infer the molecular number density along each LOS.

time-resolved manner along multiple arcs, which enables robust estimation of the time-averaged emission rate.

We validate this approach through simulated detection of a heated, turbulent CH_4 plume with a commercial imaging device. Thermodynamic state data was obtained from a large-eddy simulation (LES) of the plume, and light incident on the camera's aperture was calculated by a ray tracing procedure via the pinhole camera analogy. Spectral absorption data along rays throughout the gas were obtained through line-by-line calculation using the HITRAN database [38], and the radiative transfer equation (RTE) was then calculated along the camera rays to determine the spectral intensity at each pixel. The incident intensity was downsampled with an instrument lineshape (ILS) function to account for the camera's Michelson interferometer [39]. The diffraction-limited nature of the camera's optics [40] was simulated with a Gaussian point spread function, and white Gaussian noise was added to simulate intensity fluctuations due to a photon shot process. The result was a series of synthetic, time-resolved hyperspectral images, akin to those from a Telops Hyper-Cam. Our proposed model was executed on 0.5 s of artificial hyperspectral data, sampled at 50 Hz, to estimate the mass emission rate of CH_4 through 20 selected control surfaces. Gaussian closure of the measurement model improved the accuracy of column density measurement by 14% compared to uniform-concentration calculations. In our simulated experiment, we measured a CH_4 emission rate of 5.2 g/s over a 0.5 s interval; this estimate lay within 4% of the 5.0 g/s ground truth. Our results support the use of Gaussian distributions of thermodynamic properties to calculate the RTE in hyperspectral gas diagnostics.

2. Measurement model

Fig. 1 illustrates a typical remote sensing scenario, in which an imaging Fourier transform spectrometer captures an image of a gaseous emission plume. The interferometer conducts a scan during image capture to construct an interferogram, which is Fourier-transformed to obtain spectrally-resolved intensity data for each pixel. The resulting set of monochromatic images is called a hypercube.

Pixels throughout the image relate to a LOS through the camera's FOV, and the intensity recorded for a pixel relates to the radiative transfer along the corresponding LOS. We infer the presence of a target molecule in the FOV using a model of the spectral intensity along each LOS.

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