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SOLAR RADIATION BASED PLUME TRANSMISSIVITY MEASUREMENTS[1]

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INTRODUCTION

The emission of fine particulate matter (PM) is a primary environmental and health concern [1]. In Canada, emissions of both PM10 (particulate matter less than 10 μm in aerodynamic diameter) and PM2.5 (less than 2.5 μm) are classed as criteria air contaminants (CAC) and are tracked in the National Pollutant Release Inventory (NPRI). However, there are crucial gaps in our ability to accurately obtain these data. This problem is especially urgent in the upstream oil and gas sector where the distinct lack of practical, reliable, and accurate approaches for measuring or even estimating PM emissions from open industrial sources is a critical issue.

PM may be emitted from a variety of processes but specific difficulties arise in measuring PM from open sources such as gas flares. In Canada and around the world, significant volumes of gas are flared annually both on land and offshore. In Alberta alone an estimated 908 million m^3 of gas were flared in 2003 [2] despite significant reductions over the previous several years. By nature, flares are unconfined and emit directly into the open environment through a flame. Sampling of any type of flare emission is extremely challenging as plumes are typically not homogeneous [3], the position of the turbulent plume is varying, the amount of dilution in the plume at a given location is typically unknown and varying, the wind speed or flux of pollutants through a sample location may be unknown and varying, and the combustion process itself may be unsteady. Optical sampling methods show the greatest promise as an approach toward meeting these challenges but to date little progress has been made in accurately measuring particulate emissions from flares. This paper reports preliminary development of a potentially novel technique of quantifying soot (PM) emissions in flare plumes based on monochromatic plume transmissivity measurements using direct and/or sky scattered solar radiation as the light source.

EXPECTED SOOT MORPHOLOGY AND EMISSION RATES

Unfortunately there is no information in the literature on the morphology of flare generated soot. However, Faeth and coworkers have studied soot morphology and optical properties from soot generated from turbulent diffusion flames, which should have very similar characteristics to a flare [e.g. 4-7]. Faeth and coworkers found primary particle diameter ranges from 30-50 nm and the mean number of particles per aggregate ranges from 364-467, depending on the flame conditions. Such soot would have aerodynamic diameters almost exclusively less than 1 μm [8]. Research on the morphology of soot generated from various sources has found similarly small aerodynamic diameters [e.g. 9, 10]. By extension, one could confidently expect that all soot produced by flares would be classed as PM2.5.

The size and complex shape of soot aggregates have a significant impact on the interpretation of measurements made using optical diagnostics. Generally Mie scatter theory is inappropriate for interpretation of measurements of soot aggregates but good correlation of theory and experiments has been achieved using Rayleigh-Debye-Gans theory for the light interaction with polyfractal agglomerates (RDG-PFA) [6]. However, if the soot morphology is not known, it is very difficult to correctly interpret light scatter measurements. This negatively impacts or even prohibits the use of some diagnostics such as LIDAR, where Mie scatter theory is often used [11]. RDG-PFA theory shows that light absorption by soot aggregates scales linearly with soot volume and is insensitive to soot morphology for soot

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morphologies characteristic of flames [6]. Therefore soot light absorption measurements should be relatively easy to interpret quantitatively.

Although characteristics of individual flares may have extreme variations, detailed analysis of AEUB data on reported flaring volumes suggests a common flow rate of solution gas in a flare is 475 LPM [12]. Variability also exists in the chemical composition of the flare gas which is a critical parameter in soot formation but available data show that the principle component in flare gas is typically methane with decreasing amounts of higher hydrocarbons [12, 13]. For a methane fueled flare with a flow rate of 475 LPM, one can calculate an expected carbon mass flow rate of 4.24 g/s.

While no measurements of soot yield made over turbulent methane diffusion flames were found in the literature, measurements for turbulent propane flames suggest soot yields of 0.5 to 2.0% of the fuel mass [14, 5]. Propane has a slightly higher sooting propensity than methane, but solution gas will also contain smaller amounts of higher hydrocarbons that could be expected to increase the sooting propensity of the fuel. Based on these limited data, a conservative estimate for the emission rate of soot from a solution gas flare is 4.24 mg/s (assuming a 0.1% soot yield). Ideally we wish to design a measurement system capable of resolving this relatively low emission rate while recognizing that larger sized solution gas flares, as well as much larger well-test flares, plant flares, and off-shore production flares could be expected to produce soot at significantly higher rates and hence require less sensitivity.

LITERATURE ON PLUME PM EMISSION CHARACTERIZATION

A literature review has shown that scientifically rigorous and widely accepted approaches for estimating soot emissions from unconfined sources, such as industrial plumes and flares, do not exist. The primary standard for getting qualitative estimates of PM emissions within plumes is still EPA Test Method 9, a visual opacity standard set forth by the U.S. Environmental Protection Agency [15].

EPA Method 9 relies on a trained human observer's ability to visually estimate the opacity of a plume. This approach has several serious shortcomings including the subjective nature of the technique, varying accuracies under different background conditions, non-quantitative averaging over the area of the visible plume and potential interference due to reflection of direct solar radiation off the plume [16]. Most importantly, it is not possible to directly or accurately relate the visual opacity characterization to actual particulate mass emission rate. Various researchers have offered improvements to the human observation based visible opacity standard [16-18]; however, none have attempted to move beyond the opacity classification to a quantitative measure of soot emission rates.

A potential alternate approach is LIDAR (Light detection and ranging), which offers a means of measuring scatter coefficients for gases and particulate along a laser trajectory [11]. Unfortunately, interpretation of soot scatter coefficients with the end goal of measuring the soot volume or mass is not feasible without accurate a priori knowledge of the soot morphology [6]. Additionally, the spatial resolution of the diagnostic is inappropriate for flare plume characterization [11].

In summary, there are not current techniques or diagnostics that are appropriate for quantitative measurement of soot emissions from flares. This is the motivation for the development of the quantitative emission diagnostic proposed below.

QUANTITATIVE SOOT EMISSION MEASUREMENTS USING PLUME TRANSMISSIVITY

Theoretical Analysis for a Transmissivity Based PM Diagnostic

The amount of soot emitted from a flare is most usefully described as a mass production rate. By integrating across the plume cross-section, the mass flow rate of soot in the direction of the plume propagation can be calculated as:

$$\dot{m}_{\mathrm{soot}} = u\rho_{\mathrm{soot}} \int_{A_{\mathrm{cs}}} \mathrm{f_v}\,\mathrm{d}A, \qquad (1)$$

where u is the plume velocity, ρ_{soot} is the soot density, f_v is the soot volume fraction, and the integral is performed over an area normal to the plume flow direction (i.e. the plume cross-section A_{cs}). A soot density of 1.8-1.9 g/mL is generally accepted [e.g. 19] and the plume velocity can be related to the measured wind velocity.

The remaining unknown in Eq.(1) is the integral term. It is this term that will be measured using plume transmissivity measurements. The propensity of a soot aerosol to absorb light at a specific wavelength λ, referred to as its absorption coefficient C_a^{λ}, is related to soot volume fraction through the relationship:

$$\mathrm{f}_v = \frac{C_a^{\lambda}\lambda}{6\pi \mathrm{Em}_{\lambda}}, \tag{2}$$

where Em_{λ} is a refractive index function of soot [e.g. 7]. Transmissivity (τ_{λ}) is defined as the ratio of light intensity of a beam exiting an attenuating medium (I_{λ}) to the light intensity entering the medium ($I_{o,\lambda}$), along a chord through the medium. Unlike the opacity measurements discussed above which use white light, quantitatively useful transmissivity measurements must be made at a single wavelength (i.e. chromatic measurements) since the absorption coefficient and refractive index functions are wavelength dependent. From Beer-Lambert's law, τ_{λ} is related to the spatially dependent absorption coefficient of a non-homogeneous medium through:

$$\ln\left(\tau_{\lambda}\right) = \ln\left(\frac{I_{\lambda}}{I_{o,\lambda}}\right) = \frac{-1}{1+\rho_{sa,\lambda}}\int C_a^{\lambda}(s)\mathrm{d}s, \tag{3}$$

where $\rho_{sa,\lambda}$ is the wavelength dependent ratio of scatter cross section to absorption cross-section [20]. If we orient the x-axis such that it is coincident with the optical axis, we can insert Eqs.(1) and (2) into Eq.(3) after breaking the area integral into an x and y components:

$$\dot{m}_{\mathrm{soot}} = \frac{-u\rho_{\mathrm{soot}}\lambda}{6\pi \mathrm{Em}_{\lambda}\left(1+\rho_{sa,\lambda}\right)}\int \ln\left(\tau_{\lambda}(y)\right)\mathrm{d}y \tag{4}$$

Therefore, the mass emission rate of soot can be determined from numerical integration of a series of transmissivity measurements perpendicular to plume flow direction.

In order to evaluate Eq.(4) the ratio of light scatter by soot to light absorbed, $\rho_{sa,\lambda}$, must be known or determined. $\rho_{sa,\lambda}$ is dependent on the soot morphology, refractive properties and the wavelength of light used in the light attenuation measurements. If the above parameters are known, $\rho_{sa,\lambda}$ can be calculated using Rayleigh-Debye-Gans Polyfractal Agglomerate (RDG-PFA) theory [6]. As an example, Koylu and Faeth found $\rho_{sa,\lambda}$ values which scaled from 0.20 to 0.40 (for $\lambda = 514$ nm) for light scatter from soot emitted from turbulent diffusion flames for a range of fuels [4]. They observed that $\rho_{sa,\lambda}$ increases with increasing sooting propensity of the fuels, with the smallest value of 0.20 corresponding to propane. Therefore it is expected that ρ_{sa} for soot produced from a methane turbulent diffusion flame will be below 0.20.

A second assumption of the diagnostic is that the visible light used for the light attenuation measurements is principally absorbed by soot. This will be true if soot particulate and gases are the only constituents of the plume. If liquid droplets or other aerosol are present in the plume, they could interfere with the measurements, increasing the perceived soot emission rate of the flare. Since the variation of the transmissivity of a soot aerosol with wavelength can be predicted using Eq.(2), measurement of the plume transmissivity at various wavelengths could be used to determine if any non-soot constituents are contributing to the light attenuation measurements.

In the form proposed here, the plume transmissivity diagnostic does not distinguish between PM2.5 and PM10 in the soot mass emission rate calculation. However, measurements of soot morphology in turbulent diffusion flames [4] show that soot aggregates rarely grow to sizes greater 1 μm. Thus, it is reasonable to assume that all particulate emissions from flares are PM2.5.

Required detection sensitivity for plume transmissivity measurements

A number of variables influence the needed sensitivity of plume transmissivity measurements. In particular, plume velocity u, plume characteristic width w_{plume}, expected soot emission rate $\dot{m}_{\text{soot}}$, and wavelength λ are relevant. The measurement system sensitivity can be characterized by the highest transmissivity $\tau_{\max}$ that the system can accurately measure. In order to understand the interplay of the system sensitivity with these variables, Eq.(4) can be solved for different scenarios. For the purpose of this sensitivity analysis, the plume is approximated as having a rectangular cross-section with a characteristic width w_{plume}, and constant transmissivity. With this assumption, Eq.(5) can be simplified to:

$$\dot{m}_{\text{soot}} = \frac{-u\rho_{\text{soot}}\lambda}{6\pi \text{Em}_{\lambda}\left(1+\rho_{sa,\lambda}\right)} \ln\left(\tau_{\lambda}\right) w_{\text{plume}} \tag{4a}$$

Graphs of the solution of Eq.(4a) for sensitivities of $\tau_{\max} = 0.95$, 0.99, or 0.999 and $\lambda = 520$ nm are shown in Figure 1. It is evident that the detectable soot mass flow rate decreases with increasing plume diameter and speed. This follows from the fact that the soot dispersion increases with both parameters. Therefore it is desirable to make measurements as close to the flame as possible (smallest plume diameter) and on low wind days (low plume speed). The figures also show that increasing $\tau_{\max}$ increases the sensitivity of the system. For a plume diameter of 1 m and a plume velocity of 10 km/hr, the minimum detectable soot mass flow rate drops from 35 mg/s for $\tau_{\max} = 0.95$ to 0.7 mg/s for $\tau_{\max} = 0.999$.

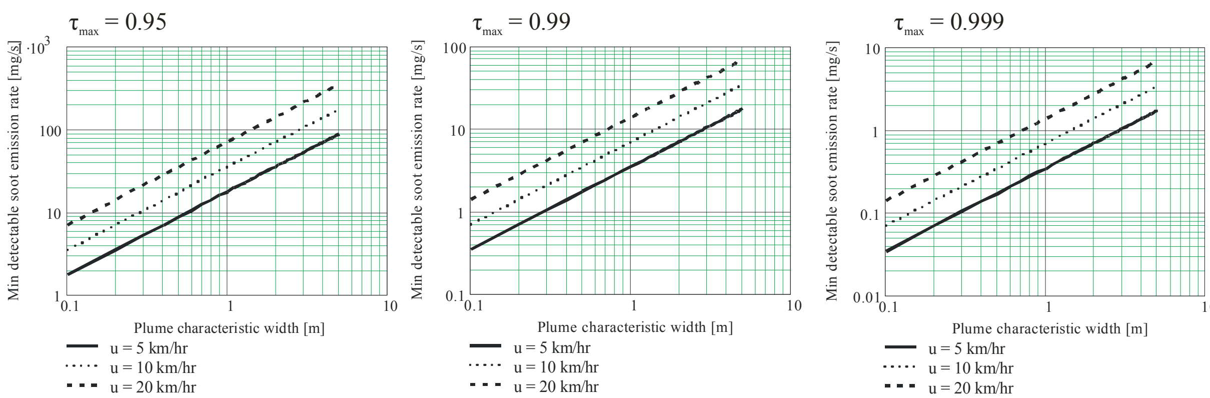

Figure 1 - Soot mass flow rate sensitivity

The target sensitivity of $\dot{m}_{\text{soot}} = 4.242$ mg/s soot mass flow rate could not be detected with a $\tau_{\max} = 0.95$ system. Under appropriate conditions (i.e. small plume and low plume velocity) a $\tau_{\max} = 0.99$ system could detect the particulate emission. A $\tau_{\max} = 0.999$ system would have sufficient sensitivity to measure soot mass flow rates expected from methane flares for a range of conditions.

Error Analysis for Plume Transmissivity Based Soot Emission Rate Measurements

The contribution of uncertainty from each variable in Eq.(4) to the total uncertainty in $\dot{m}_{\text{soot}}$ can be calculated by performing an error propagation analysis on Eq.(4). The analysis is valid if uncertainty sources are random in nature and normal in shape. From the analysis, we find that the relative uncertainty in the soot mass yield rate is:

$$\frac{\sigma_{\dot{m}}}{\dot{m}} = \sqrt{\left(\frac{\sigma_u}{u}\right)^2 + \left(\frac{\sigma_{\rho_{\text{soot}}}}{\rho_{\text{soot}}}\right)^2 + \left(\frac{\sigma_{Em_{\lambda}}}{Em_{\lambda}}\right)^2 + \left(\frac{\sigma_{\rho_{sa,\lambda}}}{1+\rho_{sa,\lambda}}\right)^2 + \frac{1}{N}\left(\frac{1}{\ln\tau_{\lambda}}\right)^2\left(\frac{\sigma_{\tau_{\lambda}}}{\tau_{\lambda}}\right)^2}, \tag{5}$$

where the σ terms represent the uncertainties in each of the variables and N is the number of transmission measurements used in the numerical integration of the integral in Eq.(4). The uncertainty in λ is assumed to be sufficiently small that it does not contribute to the uncertainty in $\dot{m}$. With the exception of τ_λ and $\rho_{sa,\lambda}$, the total relative error in $\dot{m}$ scales with the relative error of each variable in Eq.(7) summed in quadrature (i.e. as the sum of the squares). As an example, if the uncertainty in the plume velocity is 50% and this uncertainty dominates over the other error sources, the uncertainty in $\dot{m}$ will be 50%. Similarly, the total error scales with $\sigma_{\rho_{sa,\lambda}}$ as $\sigma_{\rho_{sa,\lambda}} \big/ \left(1+\rho_{sa,\lambda}\right)$. In contrast, the relative error in the transmissivity measurements is scaled by $1/\ln\tau_\lambda$. Therefore, as $\tau_\lambda \to 1$, $1/\ln\tau_\lambda \to \infty$ and the relative uncertainty in τ_λ is amplified. As an example, if $\tau_\lambda = 0.99$ and $\sigma_\tau = 0.001$, the relative uncertainty in $\dot{m}$ is 10%, if $\tau_\lambda = 0.999$ and $\sigma_\tau = 0.001$ (i.e. 0.1%), the relative uncertainty in $\dot{m}$ is 100%. The example is perhaps intuitive, however, it shows how the need for high measurement sensitivity (i.e. $\tau_{max} = 0.999$) puts high demands on the precision of the measurement system.

Although final uncertainty values can only be determined once a prototype instrument has been developed and tested, it is instructive to go through an example uncertainty estimation for reasonable conditions that might be experienced in the field. Table 1 summarizes the variables, associated uncertainties, and relative contribution to the total error for each of the variables of Eq.(4). The relative uncertainty contribution is the uncertainty in $\dot{m}_{\text{soot}}$ that would exist if the given variable was the only source of uncertainty in the measurement and is a good indicator of the anticipated importance of each variable in determining $\dot{m}_{\text{soot}}$. The individual uncertainties are combined according to Eq. (5) to give the overall uncertainty in $\dot{m}_{\text{soot}}$ shown in the last line of the table.

Table 1 - Example of uncertainty analysis†

Variable	Magnitude	Estimated Uncertainty	Relative Uncertainty Contribution to $\dot{m}_{\text{soot}}$
u	5 km/hr	1.2 km/hr	0.25
ρ_{soot}	1.8 g/ml	0.2 g/ml	0.10
Em_λ	0.285	0.056	0.20
$\rho_{\text{sa},\lambda}$	0.2	0.2	0.17
τ_λ	0.98	0.01	0.16††
$\dot{m}_{\text{soot}}$	**4.2 mg/s**	**1.7 mg/s**	**0.41**

† in this example λ = 520 nm and the characteristic width of the plume is 1 m
†† it is assumed that N = 10 for this calculation

In this example, the overall uncertainty in $\dot{m}_{\text{soot}}$ is estimated to be 1.7 mg/s, or 41%. The reader is cautioned that for the analysis, it is assumed that all uncertainty sources are random. This is significant for calculation of the relative uncertainty of τ_λ since this term scales as $1\big/\sqrt{N}$. If the uncertainty in τ_λ is systematic, it will not decrease with increasing N and the relative contribution to the soot emission rate uncertainty would be 0.49. A corollary to these observations is that the application of the diagnostic to a flare with a higher soot emission rate would significantly reduce the necessary sensitivity of the transmissivity measurement. If in the above error analysis, the soot loading was increased by a factor of 10, the permissible uncertainty in τ_λ could also be increased by an order of magnitude.

It is clear from the error analysis that most critical uncertainty in the diagnostic is in transmissivity measurements. In a laboratory environment, $\sigma_\tau = 0.001$ has been achieved [21]; however, in the field, with mobile detectors and uncertain sky conditions, σ_τ could be significantly larger. Further research, prototyping, and field tests would be needed to confirm if the uncertainties chosen in the example can be achieved in practice. Nevertheless, this example error analysis suggests that measurement of the soot emission rate from a flare could be possible with a reasonable uncertainty.

Light Source for Transmissivity Measurements

Existing plume opacity measurements use sky scattered solar radiation as a light source [e.g. 15]. On a clear day, the sky makes a good, relatively uniform light source which would suit it well for plume transmissivity measurements. However, direct solar radiation scatter by soot in the plume can bias the transmissivity measurements.

Conversely, the sun itself could be used as a light source for plume transmissivity measurements, thus avoiding the problem of direct solar radiation scatter. However, direct solar measurements are potentially more difficult to make due to the need to track the solar movement in the sky. No examples of direct solar transmissivity or opacity measurements of plumes were found in the literature; however, direct solar measurements are commonly used in atmospheric characterization of gases and aerosols [e.g. 22, 23].

It appears that direct and sky reflected solar radiation both have potential advantages and disadvantages for use as a light source for plume transmissivity measurements. It is therefore advised that both be considered further in the development of a field diagnostic.

CONCLUSIONS AND RECOMMENDATIONS

This project was initiated to complete a preliminary investigation of a potential novel technique for quantifying soot (PM) emissions in flare plumes. The proposed technique is centered on monochromatic plume transmissivity measurements using direct and/or sky scattered solar radiation as the light source for the measurements. The following is a list of our major conclusions:

- Existing soot measurement methods relevant to plumes of flares are based on EPA Test Method 9 and do not make quantitative measurements of soot emission rates;
- LIDAR cannot measure soot mass without accurate *a priori* knowledge of the soot morphology. Moreover, LIDAR spatial resolution is inappropriate for flare plume characterization;
- Analytical work suggests that soot emission rates could be quantified for plumes and flares using a novel solar based plume transmissivity technique;
- The potential technique would not discriminate particle size. However, this is not seen as a limitation for the target measurement of flares, since available data suggests all soot emissions should fall below the PM2.5 criterion;
- Initial sensitivity and uncertainty analyses support the potential for quantitative measurement of soot emissions from solution gas flares;
- As the soot emission rate increases, uncertainties diminish and the plume transmissivity technique becomes more viable such that emissions from larger plumes and flares would likely be easier to quantify;
- Both direct solar and sky reflected solar plume transmissivity measurements should be considered in greater detail

Although there are significant development challenges that may ultimately prevent any successful implementation of this approach, based on our research, we believe we have identified a potentially viable approach for quantifying PM emissions from unconfined sources such as plumes and industrial flares where no other comparable approaches currently exist.

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