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Broadband chemical species tomography: Measurement theory and a proof-of-concept emission detection experiment



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ABSTRACT

This work introduces broadband-absorption based chemical species tomography (CST) as a novel approach to reconstruct hydrocarbon concentrations from open-path attenuation measurements. In contrast to monochromatic CST, which usually involves solving a mathematically ill-posed linear problem, the measurement equations in broadband CST are nonlinear due to the integration of the radiative transfer equation over the detection spectrum. We present a transfer function that relates broadband transmittances to a path-integrated concentration, suitable for tomographic reconstruction, and use a Bayesian reconstruction technique that combines the measurement data with *a priori* assumptions about the spatial distribution of the target species. The technique is demonstrated by reconstructing a propane plume, and validating the results by point concentration measurements made with a flame ionization detector.

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1. Introduction

The release of fugitive hydrocarbons into the atmosphere poses health and safety hazards [1–3] and contributes to climate change [4]. Most often, emission fluxes from industrial sources are estimated using point concentration measurements (e.g., flame ionization detectors (FIDs) [1,5], catalytic pellistor sensors [6,7], semiconductor sensors [8], and closed-path absorption spectroscopy [9,10]) coupled with empirical emissions factors and mass balance calculations [11,12]. These practices are notoriously unreliable, however, and are limited by the inherent locality of point concentration measurements [13]. Moreover, point-measurement surveys are costly, time-consuming, and often require personnel to work under hazardous conditions.

Such shortcomings motivate the development of stand-off optical methods for quantifying fugitive emissions. Optical methods exploit the fact that hydrocarbons absorb, emit, and scatter radiation at select wavelengths that are transparent to other ambient species. One of the most accurate of these techniques is differential absorption light detection and ranging, DIAL, which infers the concentration of airborne hydrocarbons from the difference between backscattered laser light measured at two wavelengths [14]. DIAL provides highly accurate environmental measurements of volatile organic compounds (VOCs) in the C_2 to C_{22} range, including alkanes, alkenes, aromatics, benzene, and toluene, as well as simple

molecules like methane (CH_4) [12,15–20], but is too costly and complex to use on a daily basis.

Open-path (OP) absorption measurements infer the path-integrated concentration of the target species, measuring the intensity attenuated by a gas between a transmitter and receiver. The path-integrated gas molecular concentration is assumed to scale with the average absorption or extinction coefficient *via* the Beer-Lambert law. OP techniques based on tunable diode laser absorption spectroscopy (TDLAS), differential absorption spectroscopy (DOAS), and OP Fourier-transform infrared (OP-FTIR) spectroscopy have been extensively deployed for quantifying fugitive emissions [21,22]. TDLAS measurements are generated by scanning a monochromatic source over a short spectral range. The reference intensity is inferred from wavenumbers that do not correspond to an absorption line. Transmittance data is resolved for multiple lines, which may be used to infer the concentration of multiple species as well as temperature and pressure [23]. DOAS is similar to TDLAS, except it uses ambient solar radiation or a broadband thermal source, which is spectrally-resolved at the detection stage by a scanning monochromator, spectrograph, prism spectrophotometer, or interferometer (as in FTIR) [24,25]. OP-FTIR is essentially DOAS with a broadband thermal source and interferometer-equipped detector.

Individual OP measurements only provide path-integrated concentration data. Therefore, in order to accurately quantify an emission flux or image and localize a source, multiple measurements must be combined and augmented with assumptions about the target species, resulting in a 2D or 3D concentration map. For instance, in radial plume mapping (RMP) a measurement plane is defined downwind from an emission source, and three or more OP

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measurements are made across the plane. A presumed Gaussian or bi-Gaussian plume profile is then fit to the measurements to obtain a spatial concentration map, which is combined with anemometry measurements to obtain a flux estimate [26]. Numerous RPM experiments have found the distribution shape to strongly influence the net emission estimate [27–29]; moreover, a source can vary in shape and location over time. As such, it is desirable to relax shape constraints on the gas distribution.

Absorption chemical species tomography (CST) maps the spectral absorption coefficient, in a domain using OP measurements between a transmitter (*i.e.*, a laser, broadband thermal source, or LED) and receiver, without assuming a specific distribution shape [30]. McCann et al. [31] and Cai and Kaminski [32] recently reviewed the state-of-the-art in CST. While lab-scale experiments make up the bulk of CST studies, this technique has also been deployed to monitor environmental emissions. For instance, Piper et al. [33] used a 16-beam OP-FTIR array to map the concentration of sulfur hexafluoride over a 70×70 m area. Todd et al. [34] reconstructed the ammonia distribution over a 107×255 m lagoon with a 17-beam OP-FTIR array. Laepple et al. [35] constructed a similar 16-path system from two DOAS telescopes and retro-reflectors, which they used to image the distribution of nitrogen dioxide over a highway in Germany. More recently, UV-DOAS schemes were constructed to image the volcanic emission of sulfur dioxide (SO_2) [36,37]. Rivera et al. [38] and Zhang et al. [39] applied optical emissions tomography to industrial settings, reconstructing a SO_2 distribution with absorption spectrometers and a CH_4 distribution with a TDLAS system, respectively. The number of optical paths in each of these studies was small due to the high cost of tunable light sources and spectrally-resolved detectors. Increasing the number of paths can dramatically improve the accuracy of reconstructions, however, which motivates the search for an alternative, low-cost approach to spectroscopic tomography.

To the best of the authors' knowledge, CST on molecular gases has exclusively relied on spectrally-resolved absorption lines. (Polychromatic measurements are also spectrally-resolved, *i.e.*, using TDLAS, DOAS, and FTIR devices.) However, broadband transmittance measurements also indicate the path-integrated concentration of an emissions plume *via* low-cost, commercially-available OP detectors. Moreover, many of these devices are intrinsically safe, and can therefore be deployed at industrial facilities without modification. However, broadband transmittances are a nonlinear function of the gas burden. As such, in order to apply CST reconstruction techniques to broadband transmittances, a new procedure is required to process the measurement data.

This work reports the development of such a procedure and the results of the first broadband CST experiment. Broadband transmittances were measured with an incandescent source (a silicon nitride global) and a photovoltaic detector equipped with a bandpass filter. The filter range was aligned with the main ro-vibrational HC stretching band at $3.3 \mu\text{m}$. Bandpass filters omit most incoming radiation but nonetheless combine light over a spectrum that includes many spectral lines, giving rise to the nonlinear dependence of transmittance on path-integrated concentration. We therefore developed a transfer function to account for this spectral convolution, specific to a target species over a set spectral range. Transfer functions were constructed for CH_4 , ethylene (C_2H_4), and propane (C_3H_8), over a range corresponding to the bandpass filter window, using a spectral line database [40] and high-resolution spectral transmittance-reference data [41].

The technique is demonstrated by imaging a propane plume, perturbed by advection normal to the flow. Tomographic measurements were made with a 35-path array and linearized with the C_3H_8 transfer function. A Bayesian framework was employed to reconstruct the plume, using a Tikhonov prior to promote spatially-smooth estimates. The reconstructed plume structure agreed with

point concentrations made using a flame ionization detector (FID). Our results support the development of broadband CST for the quantification of anisotropic, time-varying hydrocarbon emissions.

2. Broadband tomography

2.1. Monochromatic absorption tomography

Fig. 1 illustrates a monochromatic OP measurement at wavenumber η through an absorbing gas. The measurement model assumes that the field is discretized into pixels that contain a uniform concentration of gas.

The CST measurement model derives from the radiative transfer equation (RTE), which is simplified by neglecting thermal emission and scattering. This results in the Beer–Lambert law

$$I_{L\eta} = I_{0\eta} \exp \left\{ - \int_0^L \kappa_\eta[\mathbf{r}(u)] du \right\}, \quad (1)$$

where $\mathbf{r}(u)$ is a vector function that points at a position along the path corresponding to a parametric distance u between the transmitter ($u=0$) and the receiver ($u=L$); $I_{0\eta}$ is the spectral intensity of the source; $I_{L\eta}$ is the spectral intensity at the detector; and κ_η is the spectral absorption coefficient, which is proportional to the local target species concentration (discussed further in Section 2.2). Eq. (1) is rearranged into a Fredholm integral equation of the first kind (IFK)

$$b_i \equiv \ln \left[\frac{I_{0\eta}}{I_{L\eta}} \right] = \int_0^L \kappa_\eta[\mathbf{r}(u)] du, \quad (2)$$

where b_i is the path-integrated absorption coefficient along the i th optical path. A total of m paths transect the domain, and the corresponding measurements b_i are arranged to form the vector $\mathbf{b} \in \mathbb{R}^m$.

Distributions of estimated quantities, *e.g.* species concentration, must be represented in some parametric form. A common choice is to represent the unknown gas concentration using n square pixels [24], as in Fig. 1, each of which contains a uniform concentration of the target species. Estimates of κ_η are therefore represented as a vector of coefficients $\mathbf{x} \in \mathbb{R}^n$ and Eq. (2) is approximated by a “ray-sum”

$$b_i \approx \sum_{j=1}^n a_{ij} x_j, \quad (3)$$

where a_{ij} is the chord length of the i th path in the j th pixel, shown in Fig. 1. Collectively, the ray-sums for each path yield the ray-sum matrix $\mathbf{A} \in \mathbb{R}^{m \times n}$, resulting in the matrix equation $\mathbf{A}\mathbf{x} = \mathbf{b}$. In most CST experiments the number of paths m is limited due to equipment costs and restrictions on optical access. If the uniform pixel concentration assumption is to be reasonable, pixels must be small so n , in turn, will be large. Under those circumstances, $m < n$ and the system is rank-deficient, according to the rank-nullity theorem. In other words, since there are more variables than equations, an infinite set of \mathbf{x} vectors will satisfy any measurement vector \mathbf{b} . As such, *a priori* information must be added to the measurement data to estimate \mathbf{x} .

Bayesian tomography is a framework through which measurement information is synthesized with *a priori* information, producing an estimate of a species distribution. In Bayesian CST, the measurement data and inferred parameters are conceptualized as random variables, characterized by a probability density function (pdf). Measurements are then used to update the distribution parameters and estimate \mathbf{x} . Bayesian CST begins with Bayes' relation

$$\pi(\mathbf{x}|\mathbf{b}) = \frac{\pi(\mathbf{b}|\mathbf{x})\pi_{pr}(\mathbf{x})}{\pi(\mathbf{b})} \propto \pi(\mathbf{b}|\mathbf{x})\pi_{pr}(\mathbf{x}), \quad (4)$$

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