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# Open-Path Mid-Infrared Remote Sensing of Atmospheric Gases Using a Broadband Optical Parametric Oscillator

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**Abstract:** Using active Fourier-transform spectroscopy with a mid-infrared ultrafast optical parametric oscillator we demonstrate quantitative, open-path, simultaneous concentration measurements of water, methane and ethane at over 30-m range with a simple target. © 2019 The Author(s)  
**OCIS codes:** 010.0280 Remote sensing and sensors; 300.6390 Spectroscopy, molecular.

## 1. Introduction

Fugitive hydrocarbon emissions cost the energy sector \$5B per year, account for 12% of greenhouse gas emissions and jeopardize safety and public health. Current gas monitoring methods lack the ability to take measurements in moderate winds, at varying heights and to provide simultaneous measurements of a diverse range of hydrocarbon species. The most advanced technique for remote emission measurements is differential absorption lidar (DIAL), in which intense IR pulses are fired into the atmosphere and returned to a ground-based detector by weak scattering from airborne particles. DIAL has many limitations due to the use of narrow-line ( $1\text{ cm}^{-1}$ ) dye-laser technology which restricts it to measuring only one chemical at a time, and scanning for multiple compounds that absorb at different wavelengths is time consuming. Furthermore, DIAL is a complex system and hard to maintain because it uses dye laser technology; it is also large and inefficient. By contrast, Fourier transform infrared (FTIR) spectroscopy, already a gold standard for laboratory chemical identification, is naturally broadband, offering far wider species coverage to DIAL, but the beam quality of the thermal sources in lab instruments is inadequate for free-space remote spectroscopy. Laser-based active FTIR spectroscopy therefore presents immediate opportunities for simultaneous and quantitative hydrocarbon emissions monitoring in and around petrochemical sites, at landfill sites and in agricultural contexts. Critically, the ability to measure methane and ethane simultaneously makes it possible to separate methane contributions from oil and gas sources—which are accompanied by a weaker correlated ethane emission—from biogenic sources (cattle, landfill, compost) which produce only methane.

Here we present an eye-safe active FTIR spectroscopy system based on a broadband ultrafast optical parametric oscillator (OPO) operating in the 3.1–3.5- $\mu\text{m}$  wavelength range (where common hydrocarbons display absorption peaks) and capable of acquiring gas absorption spectra from a simple target at ranges exceeding 30 metres. We demonstrate that simultaneous quantitative measurement of atmospheric background levels of water and methane is readily achieved by this system, even in the presence of strong absorption from a control cell of 1.5% ethane gas.

## 2. Experiment

The system employs an ultrafast OPO (*Spark-OPO*<sup>TM</sup>, Chromacity Ltd.) based on a fan-out MgO:PPLN (periodically poled lithium niobate) crystal, which provides 100-MHz pulses with instantaneous coverage from 3.1–3.5  $\mu\text{m}$  and an average power of >300 mW. The spectrometer is based on that described in [1], in which the OPO light is first coupled into a scanning Michelson interferometer before being launched into free-space and subsequently collected by a suitable telescope after scattering from a remote target. The scanning interferometer operates at around 1 Hz and achieves a resolution sufficient to resolve the narrow and complex absorption-line structure of light molecules like methane and ethane. Ethane and heavier hydrocarbon molecules also display a band continuum (associated with populated rotational levels) which suppresses the overall spectral intensity, so quantitative spectroscopy requires either a reliable reference spectrum or a method of inferring the original illumination spectrum. Since the effect of a remote target cannot be known, the only practical solution is to include the illumination spectrum as a free parameter when fitting the measured spectrum to library spectra of the molecules of interest, and this approach forms the basis of extracting concentrations from the measurements here.

We performed a measurement at a range of 30 meters in which the light launched from the OPO passed through a 20-cm gas cell containing a  $1.5\pm 0.15\%$  ethane:air mix. As the example data in Fig. 1 show, envelope co-fitting (Fig. 1(a), dashed line) is a successful strategy when confronted with a spectrum containing a mixture of sharp absorption lines and continuum features, making it possible to obtain environmental concentration values consistent with

independent humidity measurements (water), established ambient levels (methane) or known control concentrations (ethane).

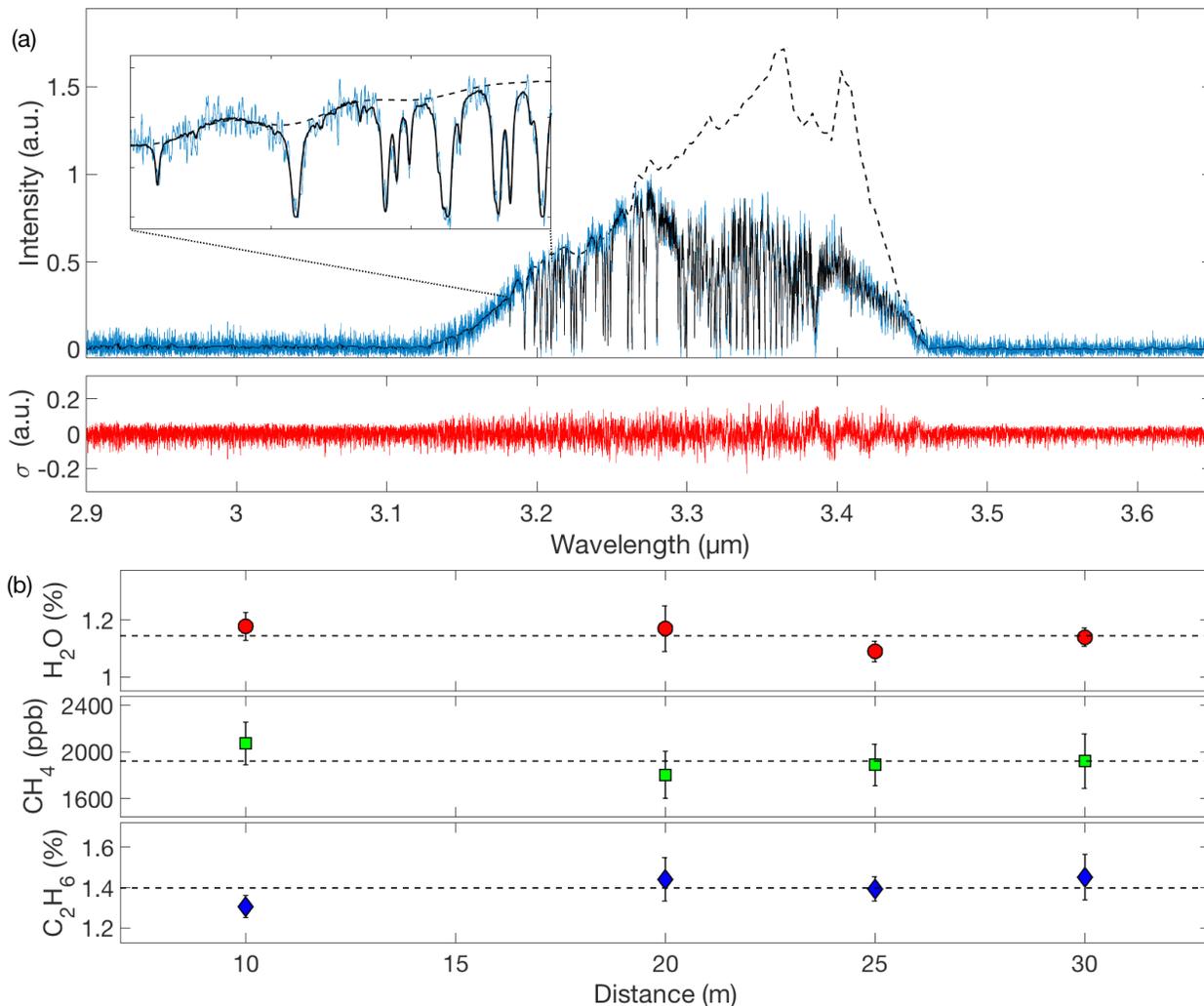


Fig. 1. (a) Normalized example spectrum from a set of 45 collected at a range of 30 meters from a reflector target. The upper plot shows the envelope of the illumination spectrum extracted from a fitting procedure which simultaneously minimized the rms error between the experimental spectrum (blue) and a synthetic spectrum (black) calculated from the envelope and a fitted mixture of PNNL absorbance data for water, methane and ethane [2]. In this example the best-fit concentrations determined were 1.15% (water), 1860 ppb (methane) and 1.37 % (ethane). The lower plot shows the rms fitting residual. (b) Measured concentrations at 10, 20, 25 and 30 meters, with data points showing the average values from approximately 45 spectra each, and the error bars showing the  $\pm 1$  standard deviation range. The methane background value is consistent with the current global ambient level of around 1900 ppb. Water and ethane values are consistent with the ambient relative humidity measured in the building and the filling concentration of the ethane cell.

### 3. Conclusions

We have demonstrated an active FTIR spectrometer capable of remote, quantitative, simultaneous multi-species hydrocarbon gas sensing, with potential applications in emissions monitoring in oil and gas, agriculture and waste management.

### 4. References

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