Letter

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Development of an open-path gas analyser for plume detection in security applications

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Abstract: We present here an open-path analyser, initially intended for security applications, specifically for the detection of gas plumes from illicit improvised explosive device (IED) manufacturing. Subsequently, the analysers were adapted for methane measurement and used to investigate its applicability for leak detection in different scenarios (e.g. unconventional gas extraction sites). Preliminary results showed consistent measurements of gas plumes in the open path.

Keywords: gas analysis; improvised explosive device; open-path analyser; quantum cascade lasers; trace vapour detection.

1 Introduction

This paper documents the development of an open-path analyser, based on a commercially available gas detection system (Cascade Technologies CT3000), designed to measure gas concentrations over a beam path potentially up to hundreds of meters. The aim was to demonstrate the potential for these instruments to detect terrorist threats at the stage of preparing improvised explosive devices (IEDs) even before the IED is completed ('left of the bomb').

The localisation of a gas source could ultimately be developed using multiple analysers in a similar fashion [1].

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2 Background

The principle of absorption spectroscopy using Beer-Lambert's law [2, 3] has long been known and understood. Cascade Technologies has developed a range of gas analysers that exploit this principle using highly efficient quantum cascade lasers (QCL) [4].

To make measurements, a high bandwidth photodetector is used to obtain a signal from a pulsed QCL. As the laser scans in wavelength, the pulse can then be analysed to show an absorption feature where specific wavelengths of light are absorbed by the gas of interest [5].

Conventional semiconductor lasers used for mid-infrared (IR) spectroscopy (commonly lead salt lasers) rely on electron-hole recombination across the doped semiconductor band-gap to emit single photons at best.

The QCL, which is based on a different principle, is a more efficient light source. They are made of very thin layers of semiconductor material, which create a set of square quantum-well potentials in the conduction band. These are offset following the application of an electric field.

Electrons cascade down this series of between 20 and 100 quantum wells, potentially producing a photon at each step. This electronic waterfall provides a step change in lasing efficiency, enabling a QCL to emit several watts of peak power in pulsed operation.

The thickness of the semiconductor layers determines the lasing wavelength. Although conventional semiconductor laser technology is often limited by material barriers, QCLs are available emitting from the near-IR region through to the tetrahertz domain [6].

QCLs began to be used in spectroscopy in the late 1990s as a spectroscopic source covering the mid-IR region of electromagnetic spectrum (3–25 μm). Multiple groups have performed gas measurements with a wide variety of techniques and wavelengths [7].

Direct absorption spectroscopy can be performed using either the interpluse or intrapulse approach.

Interpulse spectroscopy [8] uses the QCL in pulsed mode to facilitate its use at room temperature. Ultrashort current pulses to the laser are superimposed on a

slowly varying current or temperature ramp to tune the laser wavelength through the spectroscopic transition of interest. Early research found that pulsing the laser in this way caused a frequency chirp and consequent broadening of the laser linewidth and reduction in resolution. To minimise this effect, it was necessary to limit the pulse width to less than a few tens of nanoseconds while keeping the pulse amplitude near threshold. The typical tuning range for this technique is of the order of 1 to 2 cm^{-1}, with repetition rates ranging from tens of hertz through to kilohertz.

The interpulse method has been employed with considerable success in spectroscopy. However, threshold current limitation, noise caused by shot-to-shot variation, and attainable duty cycle have prevented this approach from achieving the very highest level of sensitivity currently available to other spectroscopic techniques.

Intrapulse spectroscopy [9] also uses the laser in pulsed mode to allow room temperature operation. The key difference from the interpulse technique is that the frequency chirp is maximised and harnessed to provide a fast scan through a spectral region of interest, with no need for a subthreshold ramp. Pulse widths up to several microseconds are employed with pulse amplitudes well above lasing threshold to produce a top hat current pulse. This causes localised heating of the laser and consequent downchirp, which is typically between 4 and 10 cm^{-1} wide (see Figure 1). The spectral resolution in this case is defined by the instantaneous linewidth of the laser as it sweeps in wavelength, given by

$$\Delta\nu = \sqrt{C\frac{d\nu}{dt}} \tag{1}$$

where $\frac{d\nu}{dt}$ is the laser chirp rate and C is a form factor defined by the pulse shape [10]. Typical QCL frequency downchirps will have better than 0.01 cm^{-1} spectral resolution, which is at par with many Fourier transform spectrometers. Repetition rates of up to 1 MHz can be used giving high-duty cycles and the resulting spectra are averaged to provide excellent signal-to-noise ratio levels at a high measurement rate.

Operating the laser in this quasi-continuous-wave intrapulse regime provides another less obvious but significant spectroscopic advantage. The fast chirp rate can be used in conjunction with a careful optical design to ensure incoherent optical feedback. This is used to prevent laser feedback noise and optical fringing, which tend to be the common noise floors for most practical implementations of the optical spectrometer design.

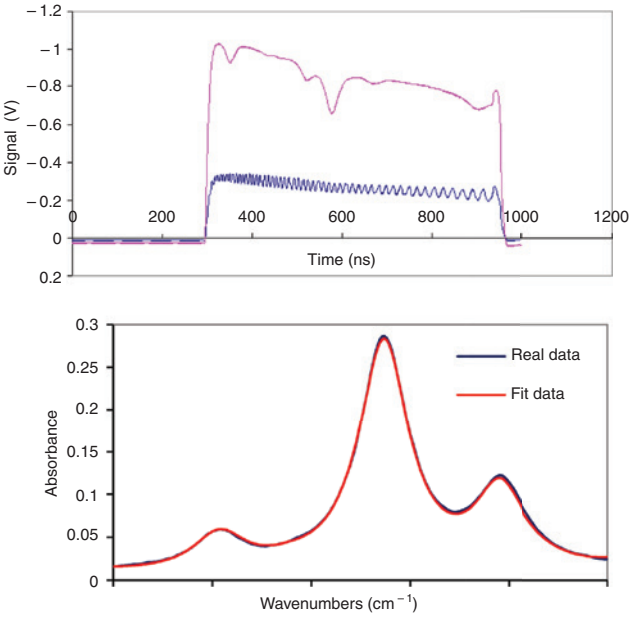

Figure 1: Raw pulse (top, pink) recorded using a 4.5 μm QCL measuring a sample of CO_2.
A 0.048 cm^{-1} Ge etalon signal (top, blue) confirms that the tuning is just over 2 cm^{-1}. The resulting calibrated absorbance spectrum (bottom, blue) can then be fitted (bottom, red) to calculate the concentration of CO_2 present.

The removal of this noise floor, without the need of complex fringe removal techniques such as Brewster plate spoilers or expensive optical isolators, enables the laboratory performance of this technology to be easily transferred to real-world applications. Cascade Technologies has patented the use of the intrapulse technique and the key technological advantages it provides.

Although absorption spectroscopy is less sensitive than photoacoustic absorption spectroscopy (PAS) [11], the fact that PAS needs an enclosed cell means that it cannot readily be applied to open-path measurements.

3 Laser absorption spectroscopy for security applications

In security applications, the ability of our analysers to detect either vapour from volatile explosives or decomposition products of more stable explosives allows a wide variety of potential threats to be measured with a single analyser [12]. Cascade Technologies' current designs of commercial system can operate up to six lasers simultaneously, where each laser can be used to detect one to three different gases.

With an increasing number of IED-based terrorist attacks in recent years, the detection of IEDs during the manufacturing stage is a more desirable approach than the detection of a completed IED minutes before it is detonated. The open-path analyser can be covertly deployed on rooftops, recording measurements of compounds likely to be given off during IED manufacture. With such a network of sensors covering a city, it would be possible to concentrate counterterrorism measures in the area where these gases are detected.

4 Instrument design

Cascade Technologies' analysers operate using multiple lasers at wavelengths specifically selected for the required gas, measurement range, and application. The nominal wavelengths of the lasers used for this work are detailed in Table 1. All lasers and the Vigo PCI-2TE-8 photodetector are temperature controlled by Peltier coolers to maintain the required wavelength, such that the entire system can be operated at room temperature without any cryogenic cooling.

Although most Cascade Technologies analyser designs obtain high sensitivity at low gas concentrations by directing the laser beam through a multipass optical cell to increase the pathlength, a variation has been developed, which instead directs the laser beam over an open path to a retroreflector that can be located more than 100 m away.

The open-path analyser was initially developed for security applications, where it may not be possible to place an analyser close to the gas source or even to identify where the gas source is likely to be. The sensor uses a laser/detector unit on a tripod combined with a reflector unit on a similar tripod positioned at a known distance from the sensor. The reflector is typically a corner-cube retroreflector, although one of the systems used a plane mirror during these tests instead. The system has been field tested with separations up to 150 m between the sections. The operating layout of the analyser for these tests is shown in Figure 2.

Table 1: Nominal laser wavelengths used for these measurements.

Gas measured	Nominal laser wavelength (μm)
Nitromethane	10.9
H_2O_2	7.8
Methane	7.4

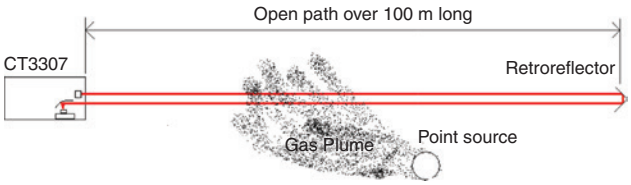

Figure 2: Operational layout of Cascade Technologies' open-path analyser.

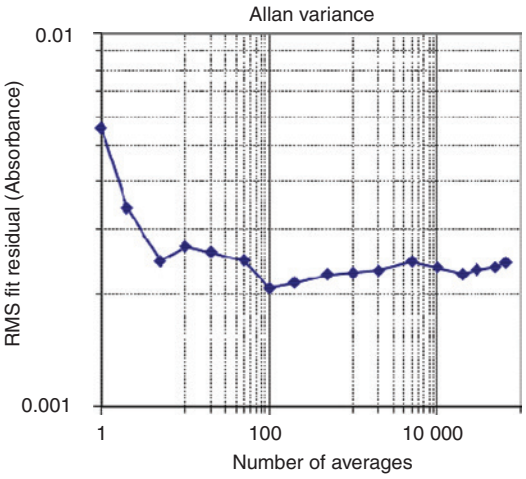

Figure 3: Allan variance plot for the open-path analyser with increasing numbers of pulses averaged together before analysis.

Although a single sensor/reflector can be used to monitor a specific section of a given scene, three of such systems used simultaneously and positioned strategically give the ability to remotely monitor a much wider area. Each system has an open-path capability of up to 150 m, allowing them to be positioned covertly on rooftops of buildings for the strategic monitoring of explosives or precursors to explosives in the vapour phase during illicit manufacturing. These sensors are capable of fingerprinting (detecting) and monitoring the concentration down to parts per billion (ppb) levels in real time (2 Hz detection rate) of several compounds of interest simultaneously. To date, work relating to the IR air monitoring system has resulted in the characterisation, fingerprinting, and test of four target compounds linked to the illicit manufacturing of IEDs.

The sensitivity of the analysers depends on the path length and, in the case of the open-path analyser, on the proportion of the path length occupied by the gas of interest. The typical detection limit of a Cascade Technologies analyser would be a spectral feature with an absorbance on the order of 2×10^{-3} as demonstrated in the Allan variance plot in Figure 3.

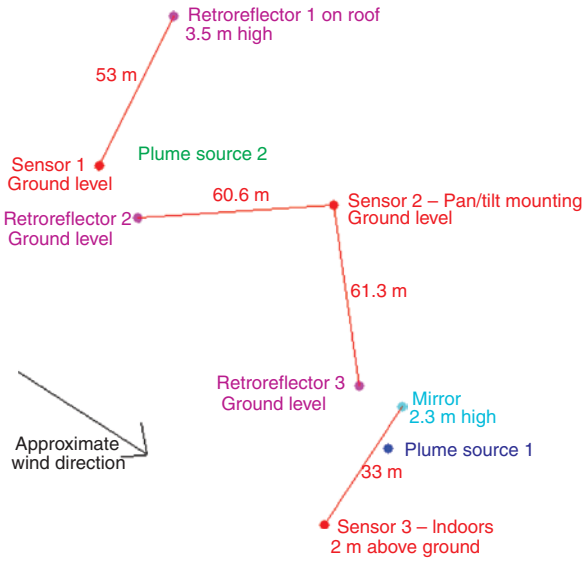

Figure 4: Analyser positions and beam paths (red), retroreflector positions (pink), and mirror position (turquoise).
The blue dot marks the ventilator for nitromethane and the green dot marks the ventilator for H_2O_2.

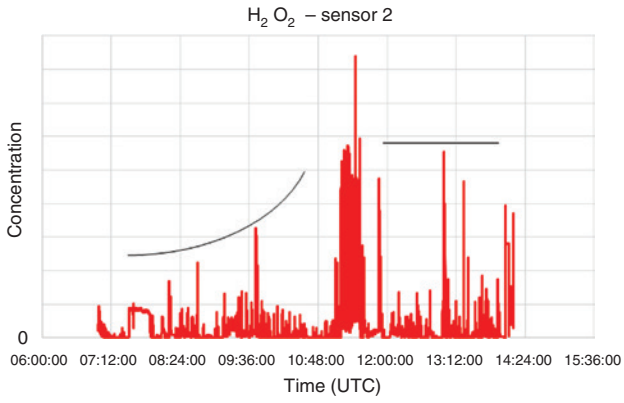

Figure 5: H_2O_2 measurements recorded by sensor 2, showing a consistent increase in peak spike strength up to 11:30, followed by more consistent spikes through the afternoon.

5 Methodology

This work was carried out at an FOI (Swedish Defence Research Agency) site using three open-path systems in different configurations to detect gas plumes from point sources at various distances from the beam paths. Two different sources were tested, a nitromethane plume and a hydrogen peroxide (H_2O_2) plume, consistent with the vapour that would be emitted during the manufacturing of different types of homemade IEDs.

The FOI research facility is located in a rural area of Sweden. Various buildings and roadways make the site an effective simulated urban environment. The tests described in this report were with the analysers laid out as shown in Figure 4.

The first sensor was set up with the laser unit on the ground and the retroreflector was positioned on a roof 53 m away, at a height of ~3.5 m above ground level.

The second sensor was mounted on a pan/tilt mechanism in place of the standard tripod. It alternated between two retroreflectors, both positioned at ground level ~60 m away, giving two beam paths at 97° to each other. The pan/tilt mechanism alternated between these two reflectors at 2-min intervals. With a suitable number of reflectors, one sensor could thus be used to cover 360° on multiple beam paths.

The third sensor was positioned about 2 m above floor level in a laboratory building. The beam was aimed through an open door at a plane mirror mounted on the roof of an adjacent building at a height of ~2.3 m.

The weather was inclement, with continuous rain and ambient air temperature around 10°C. With the exception of one analyser, all of the equipment was located outdoors. The analyser on the pan/tilt mechanism was under a gazebo to keep the rain out of the mechanism. The poor weather did not cause problems for any equipment.

6 Experimental measurements

The results from each of the analysers are shown below. Sensors 1 and 3 did not detect any H_2O_2 as a consequence of wind direction combined with beam height. Sensor 1 also detected no nitromethane for the same reason.

The gas measurements for sensors 2 and 3 show detection of spikes when the plume blew through the beam and detection of zero levels when the wind was in the opposite direction.

To generate a continuous plume throughout the day, batches of H_2O_2 were prepared. One batch would begin preparation shortly before the previous one was completed. This resulted in a gradual build-up of vapour in the morning as the first batch was prepared followed by a more continuous vapour level in the afternoon as subsequent batches overlapped. The observed measurements shown in Figure 5 reflect this approach, showing a gradual increase in the measured concentration spikes during the early part of the day followed by more steady spikes in the afternoon.

Note that all concentration readings are average concentration along the full beam path. The software makes the assumption that the gas is homogeneous throughout the path. In this application, the plume occupies an unknown proportion of the path length, so the absolute concentration within the plume is not calculable.

A specimen of the fit to the absorbance spectrum is shown below, captured from the analyser software during these measurements (Figure 6).

It can be concluded that there is a clearly successful detection of H_2O_2 plume at frequent intervals during the test.

The measurements of nitromethane from sensors 2 and 3 are shown in Figure 7.

A specimen of the fit to the absorbance spectrum is shown below, captured from the analyser software during these measurements (Figure 8).

The gas measurements shown in Figure 7 illustrate the detection of spikes when the plume blew through the beam and detection of zero levels when the wind was in the opposite direction. The strength of the detected spikes by sensor 3 consistently increased at the start of the test and then remained relatively steady throughout the rest. The blank period around 11:00 corresponds to the building door being shut, blocking the beam.

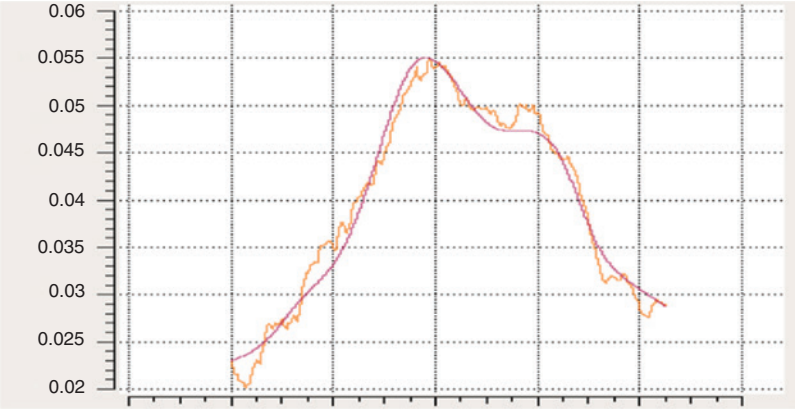

Figure 6: Sample fit (purple) to the H_2O_2 absorbance (orange) recorded during the above tests.

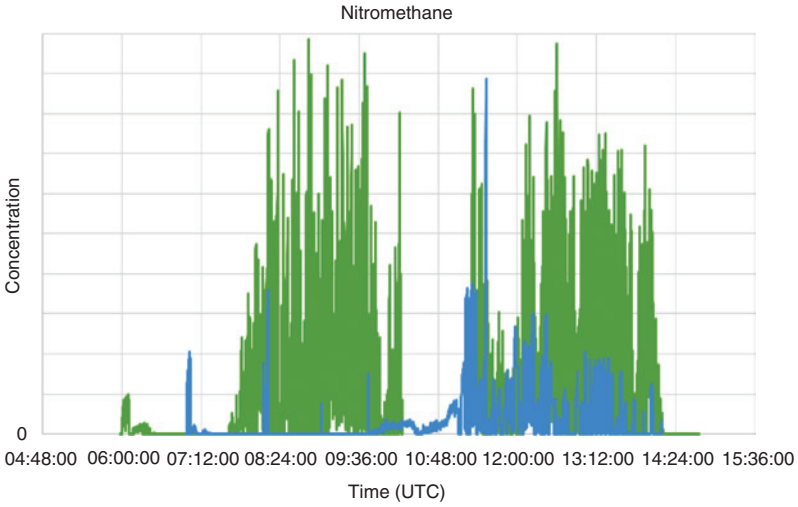

Figure 7: Nitromethane measurements recorded by sensor 2 (blue) and sensor 3 (green), showing an increase in peak spike strength up to around 09:00, followed by more consistent spikes for the rest of the day.

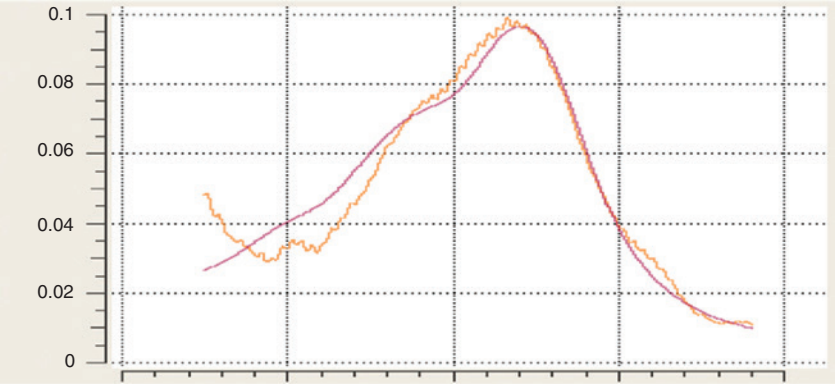

Figure 8: Sample fit (purple) to the H_2O_2 absorbance (orange) recorded during the above tests.

Sensor 2 detected few spikes in the morning but detected more in the afternoon, generally at lower levels than sensor 3, showing that proportionally less of the beam path had nitromethane vapour present.

The ability of the open-path analyser to make measurements of plumes escaping from IED manufacturing is an important step; however, the key to targeting terrorists is the ability to identify the location of the leak.

Following the test work performed in the security field, the open-path analysers were adapted to make open-air measurements of controlled methane 'leaks' from gas cylinders. The analysers successfully detected increases in the atmospheric methane concentration, demonstrating the ability of the analyser to be easily adapted and applied to more industrial applications. The analyser design could be easily adapted to any other gas by the selection of a suitable laser wavelength.

7 Conclusion

The successful detection of two compounds (H_2O_2 and nitromethane) was demonstrated under inclement weather conditions using three analysers to sample four beam paths. Both compounds were detected on at least one path, demonstrating that a network of open-path analysers could be used in an urban environment to identify occurrences of illicit IED manufacturing. When combined with other technologies such as mobile sensors and sewer monitoring, the ability to home in on a threat becomes apparent.

The units were later used to take measurements of methane plumes, demonstrating the adaptability of the technology to different gases.

The ultimate aim for this analyser would be to produce a network of sensors that would measure the atmospheric concentrations of gases likely to be emitted from some process, either industrial sites or illicit IED manufacturing.

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Authors' contribution: H.O. and E.N. designed the work. K.G.H., H.O., and O.N. performed the work. K.G.H., P.B., and E.N. analysed the data. K.G.H. wrote the paper.

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