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# STACK EMISSIONS MONITORING USING SHORT RANGE STANDOFF LIGHT DETECTION AND RANGING

**Jean-Francois Gravel*, Francois Babin*†, Martin Allard*,
Liz Siarkowski**, and François Châteauneuf***

***INO, 2740 Einstein Street, Quebec City, Quebec, Canada G1P 4S4***
***(† francois.babin@ino.ca)***
****TransCanada, 450 - 1st Street S.W., Calgary, Alberta, Canada T2P 5H1***

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***ABSTRACT***

*Promulgated methods for stack emissions monitoring are based on stack gas sampling through sampling ports in well-defined conditions. The sampled molecules are often quantified using optical techniques. Unfortunately, sampling ports are not found on all stacks/ducts or their use cannot be planned efficiently because of operational constraints. In other cases, the emissions monitoring equipment cannot be driven to a remote stack/duct. INO and TransCanada thus looked into emissions monitoring at a standoff distance using optical techniques, through the atmosphere, using short range high spatial resolution Light Detection And Ranging (LiDAR). These are preliminary tests and results, in order to gather sufficient information and experience for the eventual design of a man transportable measurement system. Standoff optical fluorescence, absorption and Raman scattering will be discussed and results from field campaigns for $NO_x$, $CO_2$ and $O_2$ will be presented along with short descriptions of the apparatus. Measurements on both gas turbine exhaust stacks and mock-up stacks with controlled concentrations of $NO_x$ are analyzed. Emphasis is put on multi-wavelength Ultraviolet (UV) differential optical absorption LiDAR for $NO_x$ measurements and on Raman LiDAR for $O_2$ measurements. Extending the techniques to $SO_x$ and $CO_2$ will be discussed.*

## 1. INTRODUCTION

Compliance to regulations in the arena of emissions monitoring of exhaust stacks is often done through promulgated methods. These are well documented and based on sampling the exhaust through sampling ports on the stack when it is not done using often outdated emission factors. The concentration of the pollutants and other molecules of interest are measured using instruments that often rely on optical phenomena; optical absorption, fluorescence, chemiluminescence, and the like. The

mass emission rate is then computed once the exhaust gas flow rate is determined using these same sampling ports.

On the other hand, open path optical techniques have been developed for area or fenceline monitoring. Promulgated methods exist for the use of open path techniques. An open path optical technique implies that the light beam used to measure the optical phenomenon of interest travels through open space, instead of inside an analytical instrument. There is no “sampling” per say, as in the sense of syphoning the air through tubes to route a sample to an instrument. The “sample” is the volume of atmosphere through which the light beam travels. These open path techniques can be further subdivided in “integrated” path measurements or spatially resolved measurements. In the first case, the molecules over the entire path are interrogated. In this case, there usually is an emitter at one end of the path and a receiver at the other. Or there is a reflector at one end, be it a mirror or a diffuse reflection on an element of the topography. In the second case, usually dubbed “LiDAR”, for Light Detection And Ranging, pulses of light are sent along the path and the echo is measured with respect to time, thus allowing the instrument to determine from where the echo originates. Particulates or molecules in the atmosphere generate this echo. The absolute amount of molecules in the interaction volume is measured in these cases and the usual unit is the ppm-m (a concentration-length product).

This project was initiated to look at the use of open path techniques for emissions monitoring of exhaust stack from a distance, preferably from outside the fence. This is distinct from the usual use of these techniques for area or fenceline monitoring. The goal was to virtually sample, from a distance, the exhaust very close to the output of the stack. The rational for this was that not all exhaust stack have sampling ports, and when they do, the use of sampling ports often perturb operations. Furthermore, easily transported instruments for monitoring at remote sites was targeted, which is usually not the case for the usual sampling through sampling ports approach.

A LiDAR can work on one of three optical phenomena (or a mix of these). The most usual is absorption. It is then called a DiAL (Differential Absorption LiDAR), DAS (Differential Absorption Spectroscopy), or DOAS (Differential Optical Absorption Spectroscopy) LiDAR. It usually relies on lasers with two or more optical wavelengths. Absorption “signatures”, the optical wavelengths at which a molecule absorbs light, are unique to each type of molecule. So the laser light is “tuned” to these absorption features unique to the molecule of interest and the optical echoes measured. The difference in return for the different optical wavelengths used is proportional to the amount of molecules in the volume of atmosphere being monitored. So it is essentially a small change in the echo that is indicative of the amount of molecules in the interaction volume. This limits the sensitivity of the approach.

Another type of LiDAR is the fluorescence LiDAR. Fluorescence is generated when the laser is again “tuned” at an absorption wavelength but it is usually light at other wavelengths that is monitored, wavelengths unique to the molecule of interest. Fluorescence can be more sensitive than absorption, since it is measured with respect to a background that is practically nonexistent. The major disadvantage is that fluorescence is much more complex to calibrate.

A third type of LiDAR is called a Raman LiDAR. In this case, the excitation laser can be at any wavelength, but it is usually preferred in the ultraviolet (UV). This simplifies the measurement apparatus. Every molecule responds by scattering the laser light at wavelengths that are different from the laser wavelength and which are unique to each molecule. The major drawback, and not a small one, is that this interaction is very weak, requiring very large amounts of laser power for remote open path operation.

INO looked into all of these for the purpose of this project with the goal of a man transportable system in mind. LiDAR systems do exist, built on a custom basis, but are more often than not huge multi-kilometer systems mounted in custom built tractor trailer type vehicle or vans. INO has been developing small short distance LiDARS for multiple applications.

For the reasons stated above, the standoff monitoring for NOx, SOx, COx, particulates and $O_2$ emissions from compressor stations is of great interest for pipeline operators. Moreover, new regulation requiring more testing and/or reporting is a strong motivator behind the work presented here.

High spatial resolution optical measurements allow for standoff identification and quantification of molecules, much like promulgated or accepted methods, except that it is done remotely in the plume formed by the emissions from the stack, very close to the stack output. The pros and cons will be discussed, and in particular the problem of mass emission rate estimates require the knowledge of the flow rate. Standoff spatially resolved measurements are much like path integrated or point in-situ measurements without inserting probes into the exhaust stack. Preliminary results show that a limit of detection of 1 ppm of NO in 3 m diameter stacks from a standoff distance of 50 m is a reasonable target for a man transportable system.

For a first test campaign, the goal was thus to measure NO. However, the focus of the first campaign was really to show that NO is detectable from a distance using INO's platforms. Eventually, the technique could be extended to other molecules, such as $NH_3$, which also has absorption features in the deep UV (maybe a little too deep). Nitrogen oxide (NO) from a few tens of ppm to 150 ppm needs to be measured for the type of engines (a GE LM1600 turbine burning natural gas) used in the targeted compressor station. $NO_2$ is the balance of NOx emissions ($NO_x$ – NO), and this is usually in the low ppm to ppb range. The $NO_2$ is much more difficult to measure from a distance with the techniques and platforms being used in this project because of its very low concentration and because of the much lower optical absorption than NO for an equivalent concentration. Preliminary testing at the compressor testing site also shows that it should be relatively easy to detect $CO_2$, $O_2$ and $H_2O$ with the INO platform as it stands at the moment, but not so much CO, which is in much lower concentration.

The first concept of measurement was an integrated path approach that called for the installation of target panels (retroreflectors or diffuse reflection from the remote end); 1) to create a measureable return for the infrared differential absorption LiDAR (IR-DiAL) platform; 2) to install the required retroreflector for the ultraviolet differential optical absorption spectroscopy (UV-DOAS) referee platform; and 3) to stop laser light from travelling upwards through the sky since this requires an authorization from Transport Canada (for pilot safety, but in the end authorization was granted to beam into the sky). Ultimately, no target panel was used, so there were no UV-DOAS referee measurements during the campaign. Since it is usually impossible or

impractical to have a target at the other end of the open optical path, the LiDAR techniques were singled out.

IR and UV absorption LiDAR measurements imply measuring a signal directly from the particulates and the molecules as a whole, and in the case of Raman and fluorescence, directly from the molecules themselves (NO, $CO_2$, $H_2O$, $O_2$).

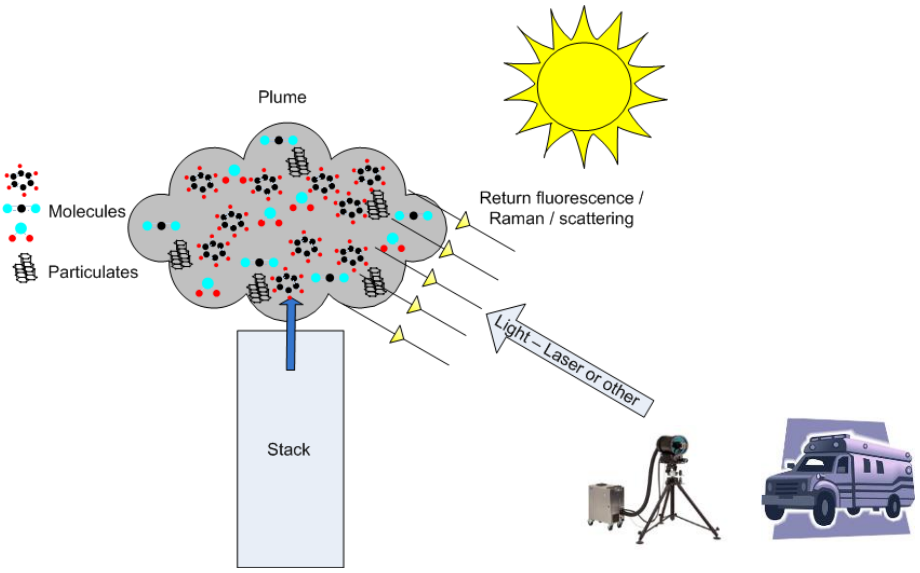


**Figure 1**: Concept of measurements. Light from the INO platforms installed in a mobile laboratory interacts with molecules and particulates in and around the exhaust stack plume. The return signal can be elastic scattering, fluorescence or Raman. For measurements in the visible, the sun is a source of noise. Emissions of interest are $NO_x$, CO, $CO_2$, $SO_2$ and particulates.

## 2. MEASUREMENT METHODS

Fluorescence measurements are straightforward. The UV beam from the laser in the INO platform is routed towards a folding mirror that sends the UV beam across the stack plume, very close to the stack output itself. The fluorescence generated by the UV excitation beam returns to the receiver telescope via the folding mirror (laser and return signal have the same open optical path). All that is necessary is that the volume spatially resolved by the platform be adjusted to be equal to the stack position and width projected along the line of sight of the UV laser beam. The Raman return from nitrogen in the air was to serve for calibration purposes, but its intensity was too low for the measurements presented here. It was to be taken over the same volume and at the same distance as the NO fluorescence measurements. There is no interference from ambient light or fluorescing aerosols at the wavelengths used for fluorescence measurements. The wavelength used for fluorescence generation is ~226 nm. The optical power out of the platform is just a few milliwatts. Fluorescence is measured before the stack output and after, in addition to inside the output plume, in order to ascertain that there is little to no contribution from the air surrounding the stack output.

For the Raman, the concentration measurement is straightforward. The measured signal is proportional to a molecule's concentration if the Raman cross-section is known for the laser excitation wavelength used. The Raman return from nitrogen in

the air is used for calibration purposes. It is taken over the same volume and at the same distance as the other Raman measurements. Interference from ambient light needs to be processed out of the measurements. The wavelength used is 355 nm and the optical output from the platform is a little over 400 milliwatts.

For IR-DiAL from particulates, the IR beam is also routed close to the output of the stack, in the plume. The plan was to use the measured return from just before the stack output and from immediately behind the stack plume relative to the laser beam propagation direction. The difference between the absorption value obtained just before the stack plume and the one obtained just after the stack plume gives the absorption in the stack plume. The first value is used to compute a baseline concentration-length product between the IR-DiAL instrument and the front of the exhaust stack for the molecule to be measured. It represents the concentration of the molecule of interest in the ambient atmosphere. The difference value is used to compute the concentration in the exhaust stack plume close to the output. The space between the front and behind measurements must be filled by the exhaust plume. The result is a concentration-length product. Knowing the distance between measurements and the fact that the entire volume is filled with the plume allows INO to deduce an average concentration. The output optical power is > 60 milliwatts. The first goal was to ascertain that it was possible to get a return from particulates in and around the compressor station stack plume with the platform. If this were successful, the aim was the measurement of $CO_2$, INO not having the required components for NO at the time of the test campaign.

Since there is a spectrometer in the IR-DiAL apparatus, it was also decided to measure the optical emission from the plume at those wavelengths where the absorption was to be measured. Any strong optical emission will be detrimental to the sensitivity of the measurement. In a hot gas, there will be optical emission at those same wavelengths that are used for absorption measurements.

If there are strong winds, the exhaust plume could extend along the propagation axis of the optical beams and fluctuate with time. This was to be avoided. The mobile laboratory and scanning mirror could thus be moved around for the wind to be in a direction that avoids extending the exhaust plume along the propagation axis. Fortunately, for the first campaign in 2013, this was not an issue.

INO's mobile laboratory was used to transport the platforms/breadboards to the compressor station site. The closed mobile laboratory is fitted with an optical table onto which the platforms/breadboards for the fluorescence/Raman and the IR-DiAL are fixed. When on site, the mobile laboratory is lifted onto four stands to stabilize the optical setups and to have stable exhaust stack pointing. The mobile laboratory is also fitted with all the required safety features (emergency shutdown and strobing lights to indicate laser beam propagation in the ambient air).

Figure 2 is a photo of the inside of the trailer showing the optical table upon which are mounted both the LIF/Raman and the IR-DIAL systems. The pointing stability of laser beams outputted from the mobile laboratory was ascertained by measuring the pointing stability of a laser secured to the optical table inside the trailer while someone was jumping inside the mobile laboratory. The pointing stability was estimated to be better than 1.5 mrad. The stability of the beams, for the purposes of the campaign, was thus not a worry.

**Figure 2**: Site installation of INO mobile laboratory, folding mirrors in forefront (left) and view of the compression station (right).

## 3. MEASUREMENTS ON A COMPRESSOR STATION STACK (FIELD CAMPAIGN)

A limited amount of testing was done on the compressor station exhaust stack. The mobile laboratory with a prototype version of the fluorescence and Raman LiDARs was driven to an operating compression station in Canada for this preliminary series of compressor station stack tests. Emphasis was put on fluorescence and Raman. Absorption measurements were originally planned in the infrared, not the ultraviolet. Unfortunately, the particulates concentration was not adequate for a significant infrared LiDAR return signal with the laser system at hand. However, some UV absorption measurements were done for tentative fluorescence calibration.

In this first field campaign, the high spatial resolution measurements were done with a time gated detection system; only a small predetermined slice of space along the laser beam path is monitored.. The detection gate corresponds to a volume 5 m in length along the laser beam path. Absorption calibrated fluorescence measurements were a surprisingly good fit with the referee measurements, except at the lower concentration end as shown in Figure 3.

For the Raman measurements, the primary goal was to show that it was possible to perform Raman LiDAR measurements on an actual compressor station stack. This was accomplished. The results presented here are not calibrated. The ratio of the Raman cross sections of nitrogen to that of $O_2$, $CO_2$ and $H_2O$ as shown in Figure 4 would need to be known for the gas conditions at hand. The exhaust stack being 3 m on the side and the Raman measurements having a limited 5 m spatial resolution in this case, the ambient air over the extra 2 m or so has an effect on the measurement accuracy and noise, especially for $CO_2$.

The conclusions of this first test campaign were that the platforms were easily and securely deployed on site. It is also clear that for detecting NO, working in the UV is much better. In fact, NO can be detected in one of two ways: UV-fluorescence or multi-wavelength UV-absorption.

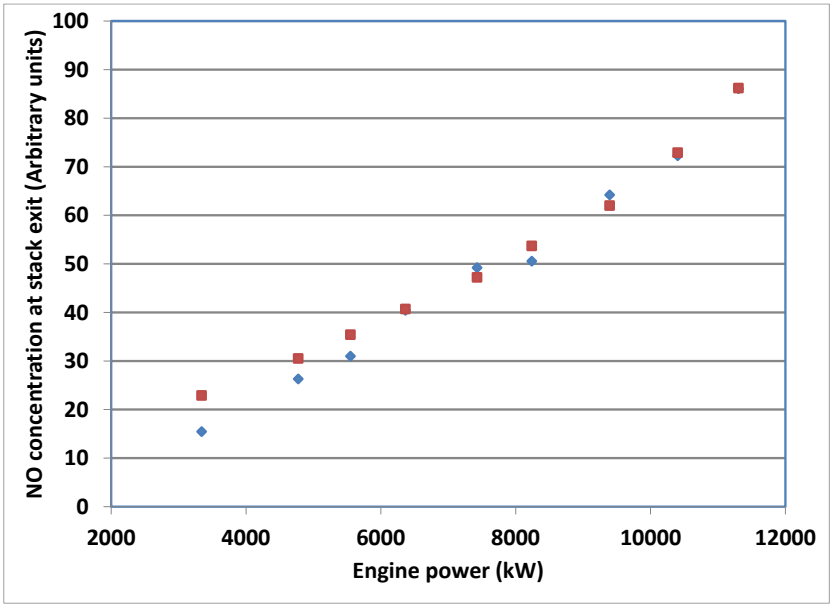


**Figure 3**: Referee measurements (red dots) were performed alongside the fluorescence (blue dots).

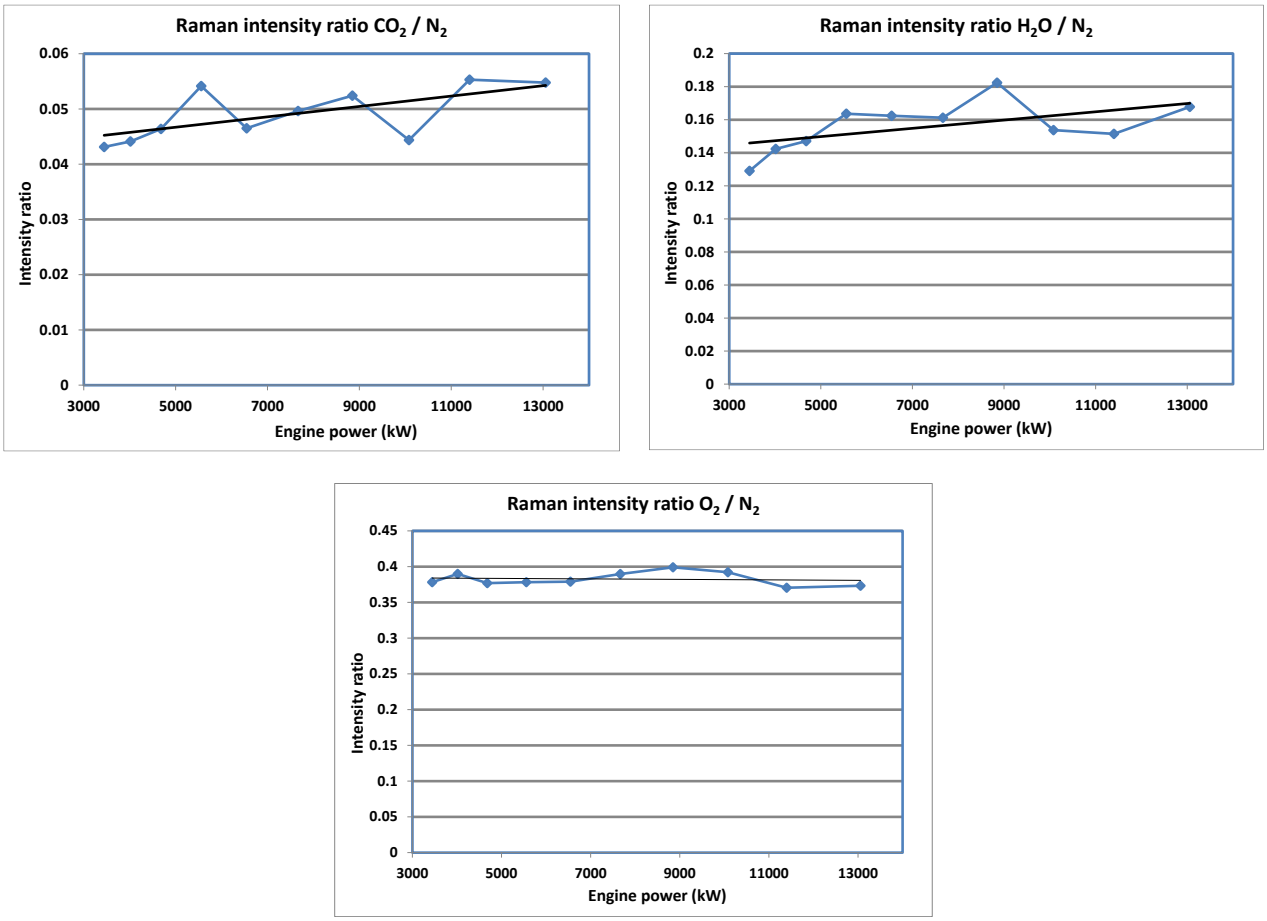


**Figure 4**: Ratio of the Raman cross sections of nitrogen to that of $O_2$, $CO_2$ and $H_2O$.

Fluorescence is very sensitive for measuring NO stack emissions and the limit of detection can easily be below 1 ppm for a 3 m diameter stack and an LM1600 type

engine. However, we could not correctly calibrate fluorescence in this first campaign because of the unplanned use of too strong a laser beam absorption at the fluorescence excitation wavelength. Recall that Raman from Nitrogen was originally planned for fluorescence calibration, but was too weak to be measured with the laser power used for fluorescence measurements. Standoff fluorescence needs an easily applied and accurate calibration technique if it is to be routinely used.

Although an absorption (UV-DiAL) measurement requires slightly more complex hardware, it is closer to a standardized method than is fluorescence. UV absorption is sufficiently sensitive for the concentrations found in the monitored stack exhaust plume. UV-DiAL measurements could not be performed in this first field campaign because of the absence of an automated wavelength tunable laser.

The main conclusion of this first field campaign was that spatially resolved optical measurements allow for standoff identification and quantification of molecules, much like the standardized methods. However this it is not done using in stack sampling, but directly in the plume formed by the emissions from the stack.

Following the first test phase on a TransCanada compression station where NO, $CO_2$ and $O_2$ were successfully measured using open path LiDAR techniques, the work presented in the following sections mainly focuses on standoff absorption and Raman for NO and $O_2$ measurements. Results obtained from intermediate scale simulated stacks are presented along with descriptions of the apparatus and procedures.

## 4. MOCK-UP STACK MEASURMENTS: METHODOLOGY

To evaluate the effectiveness of the approaches in a simple but controlled manner, two different mock-up systems were used, one using a propane burner, the other one a room temperature PVC tube. A versatile spectroscopic LiDAR breadboard system based on an automated pulsed tunable laser source is used for the measurements. Installed in a mobile laboratory located about 45 m away from the mock-up stacks, measurements are performed outdoors in a parking lot.

The first mock-up stack is based on a propane gas burner commonly used for heating. The high temperature exhaust generated by this burner is of particular interest because it creates a depleted oxygen region. Oxygen analysis is then performed by UV-Raman LiDAR.

The other system is based on a 4-inch PVC tube connected to a commercial air blower. This represents the smallest stack size expected to be encountered for compliance to regulations. The blower is used to create a high velocity gas output at one end of the tube. An injection port is also added between the blower and the PVC tube exit. A controlled NO gas mixture can therefore be introduced within the flowing air simulating a NO emission source. Nitric oxide analysis is then performed using an absorption method close to UV-DiAL. In both cases, the probing laser is aimed directly over the mock-up stack exit.

For Raman measurements ($O_2$), the 355 nm output of the Nd:YAG laser is used for excitation. For absorption measurements, the third harmonic of the Nd:YAG laser is used as the pumping source for a tunable Optical Parametric Oscillator which is then frequency doubled to generate deep UV light in the 225-230 nm range. The wavelength is precisely selected to match an absorption feature of NO molecules.

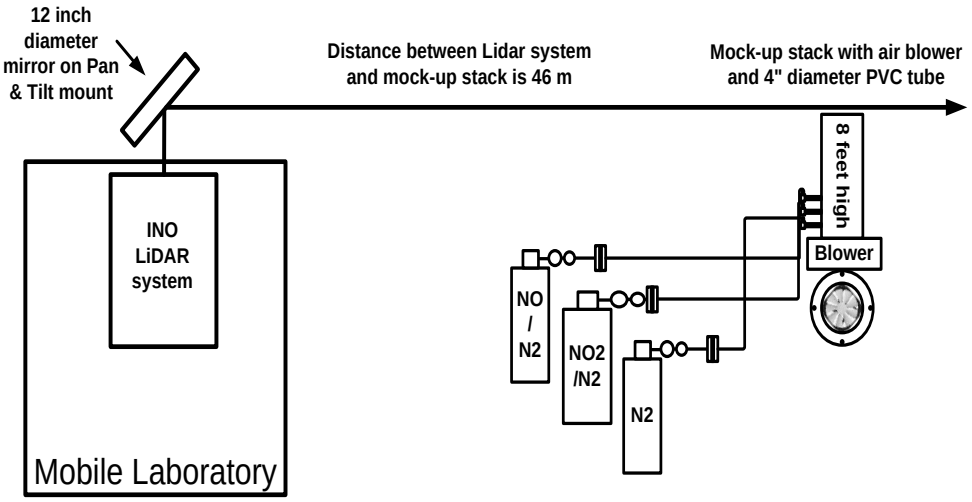


**Figure 5**. Measurement set-up

Signal collection is achieved through a 20 cm light collection aperture telescope routed either to a filter based 2-channel wavelength selector with photomultiplier tubes (PMTs) or to a compact monochromator also equipped with a PMT. Both options allow for full waveform measurements of the LiDAR returns.

### 4.1. Absorption measurements

Unlike traditional DiAL methods that require high stability of an ON absorption and of an OFF absorption wavelength that are alternated in time, the wavelength of the laser used in the tests described here is not alternated between ON and OFF transition wavelengths. Making the hypothesis that the NO emission in the stack exhaust is approximately constant within the few minutes required for the measurement, multiple wavelengths are slowly scanned across an absorption feature. The absorption cross section of NO is at the same time monitored thanks to a laser pick up that goes through a NO reference cell followed by a photodetector.

The LiDAR curves (waveforms) obtained at each wavelength are normalized to a reference wavelength LiDAR curve, this wavelength being the one for which there is minimum absorption in the reference cell. Once these normalized curves have been obtained, an average is made over a pre-determined number of digitized points (distances along the laser beam path) of the LiDAR curve. For each LiDAR return curve (for each wavelength), the ratio of the averaged values after the stack over the averaged values before the stack is computed. This results in an absorption spectrum across the exhaust stack plume. For processing purposes the absorption spectrum is transformed into a transmission spectrum. Finally, the resulting spectrum is re-normalize with respect to the highest transmission point in the spectrum, in order to have only transmissions lower than 1, including noise. The reference transmission spectrum from the reference cell is also re-normalized to 1, and these two spectra, along with the concentration in the reference cell, are used as inputs for the DOASIS software [1].

### 4.2. Raman measurements

For the analysis of $O_2$ in the propane burner exhaust, Raman returns for oxygen and nitrogen (for normalization) are collected. Ratio of $O_2$ signal over $N_2$ signal, indicates the variation in relative $O_2$ concentration as the relative $N_2$ concentration is expected to be constant within the atmosphere as well as within the exhaust gas.

## 5. MOCK-UP STACKS: RESULTS

A portion of the NO absorption features can be found in Figure 6. This spectrum has been measured in the NO reference cell. The absorption in the reference cell is measured throughout all of the experiments which means that at any wavelength used within a scan, the absorption cross-section is known. The wavelength does not need to be known with high accuracy.

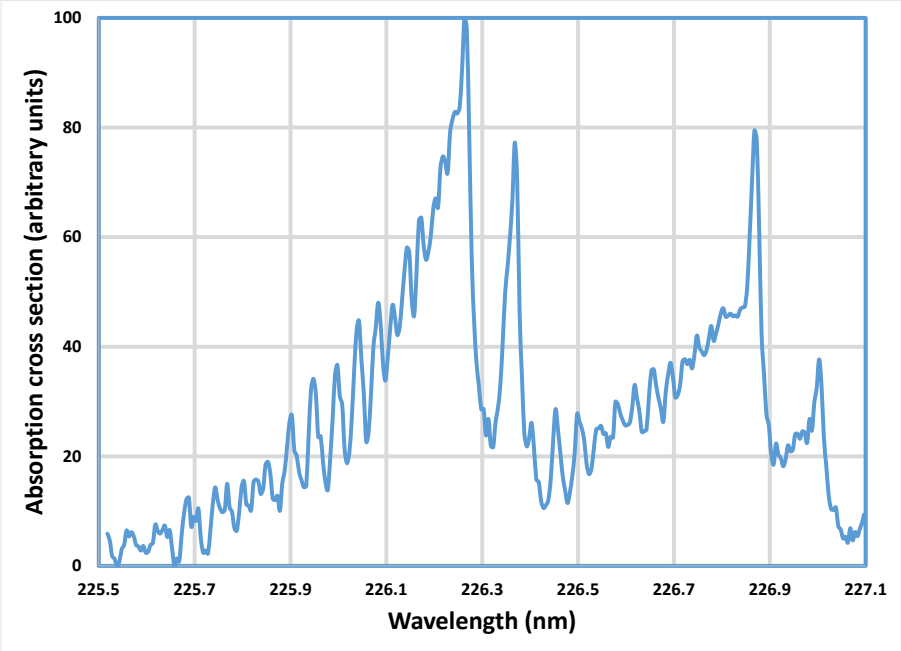


**Figure 6**: NO absorption spectrum as measured with the INO UV-DiAL platform in a reference cell uncorrected for baseline effects.

Scanning the laser wavelength across an absorption feature results in LiDAR returns used to build an absorption spectrum that reflects the presence of NO. In the case of Figure 7, it is obvious that the blue dotted line is returned by a wavelength that is closer to an absorption maximum than the wavelength represented by the full orange line. It is also clear that the absorbing region (stack simulator) is located approximately 46 m from the LiDAR platform.

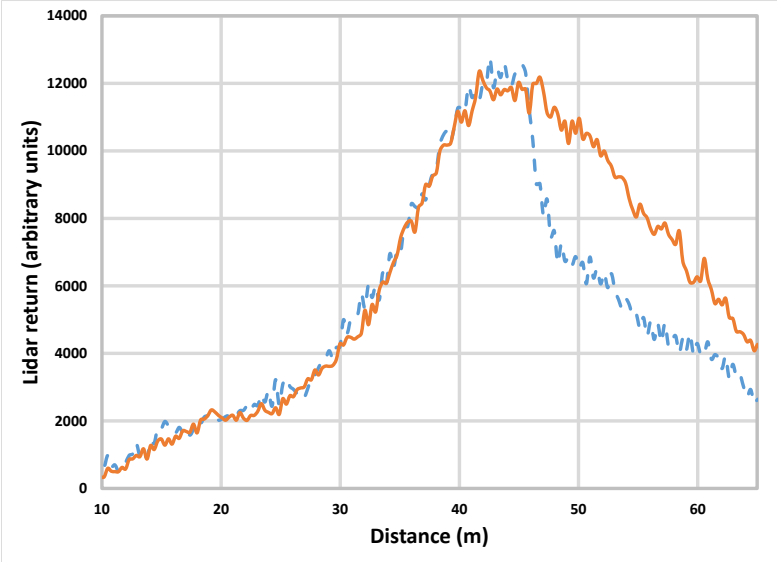


**Figure 7**: Lidar return curves for NO in the mock-up exhaust stack with and without absorption.

Eleven different wavelengths were used in the scan of the optical absorption feature of the NO in the stack plume (the plume from the room temperature PVC tube). Using the reference cell for cross-section evaluation, the results were inputted to the DOASIS software which computed the best fit to retrieve a concentration in the stack output plume. Figure 8 shows the transmission spectra computed for both the reference cell and the exhaust stack.

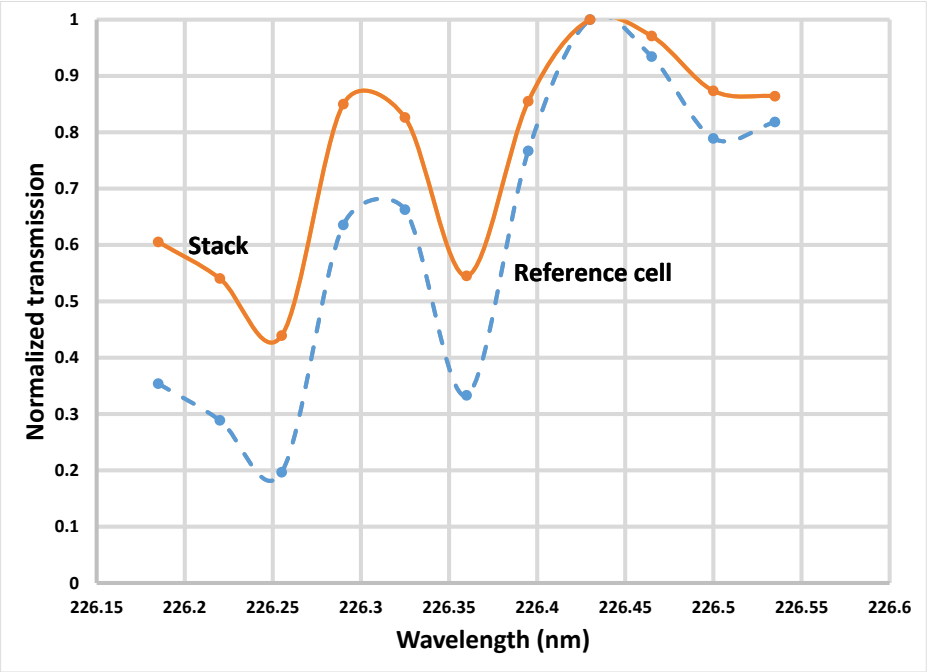


**Figure 8**: Normalized UV-DiAL and reference cell transmission spectra of NO.

Tests were performed at 3 different nitric oxide concentrations and 3 replicates. Detailed results are expressed in Table 1.

Table 1. Determined NO concentration–length products vs expected values.

| | Expected (ppm-m) | Measured average (ppm-m) | Standard deviation (ppm-m) |
|---|---|---|---|
| Concentration 1 | 116 | 111.5 | 15.4 |
| Concentration 2 | 52 | 40.4 | 3.5 |
| Concentration 3 | 23 | 20.3 | 3.8 |

It is to be noted that there is a large uncertainty in the expected values that is related to the velocity measurements and blower stability. More experiments need to be done to properly evaluate the accuracy and repeatability of the approach. However, a rough estimate of the limit of detection can be made using the standard deviation of the measurements times three (3) thus giving 11 ppm-m for a 3 min 40 s measurement.

For Raman measurements of oxygen, the propane burner exhaust was used. The analysis of the full waveform returns of both $O_2$ and $N_2$ showed a clear decrease in signal at approximately 44 m. This dip is mostly related to the lower density of the hot exhaust gases; around 600°C (see Figure 9).

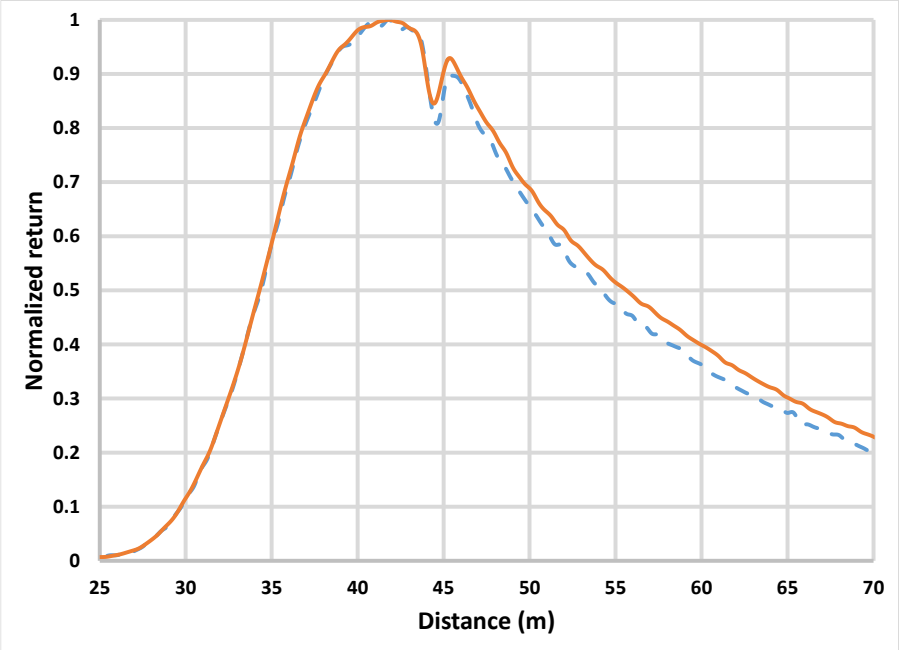


**Figure 9**: Lidar return curves of $O_2$ (blue dotted line) and $N_2$ (orange line) with propane burner « ON ».

Normalizing each point of the $O_2$ curve by the corresponding point of the $N_2$ curve results in the determination of the relative decrease in oxygen concentration related to the combustion process in the burner. Even at lower air density, relative $N_2$ concentration should remain constant.

In this case, the drop in concentration is ~ 9% of the normal 20.9% $O_2$ in air which correspond to about 19% in air. Here again using the 3 standard deviation approximation to estimate the limit of detection gives 0.4% $O_2$ within a 13 minute measurement.

## 6. CONCLUSIONS

A first test campaign on a compressor station exhaust stack showed that fluorescence is very sensitive for measuring NO stack emissions and the limit of detection can easily be below 1 ppm over a 3 m stack plume (3 ppm-m). However, calibrating fluorescence is far from trivial and has not yet been worked out. UV absorption although less sensitive than fluorescence is sufficiently sensitive for the concentrations found in the monitored stack exhaust plumes. Furthermore UV-DiAL (absorption) is closer to a standardized method.

Further testing and development was then performed on simulated stacks. It is not easy to simulate real exhaust stack conditions at a small but representative scale. However two different approaches were proposed and two different spectroscopic LiDAR approaches for monitoring of the NO emission and determination of $O_2$ concentration were tested.

It has been demonstrated that it is possible to achieve stack emission monitoring from a distance using spectroscopic LiDAR to analyze the exhaust gases. Quantification has been demonstrated with UV-DiAL and a preliminary limit of detection for NO was estimated to 11 ppm-m for a 3 min 40 s measurement time with INO's breadboard platform. For $O_2$, it was determined that the lowest variation in concentration that could be detected would be roughly 0.4% on a 13 min basis. Parts of the mock-up would need to be improved to allow better evaluation of the accuracy and precision of the methods used above. An important hardware limitation of the approaches was related to the lack of spatial resolution. Higher spatial resolution would be needed for very small diameter stacks. In particular, the 6 ns laser pulse length would need to be reduced for better spatial resolution.

Although this work is already interesting from a scientific point of view, it is important that the approaches be accepted by the industry and even more so by regulators. For this, at least two major aspects need to be addressed. First, the measurement is done in the plume just above the stack. This way of doing a measurement has not yet been documented and accepted in the standard monitoring methods, although there is a German standard for LiDAR plume monitoring. A clear assessment that the open-path sampling through exhaust gases just above the stack is equivalent to an upstream sampling done from a port located inside the stack needs to be made. Second, it is clear that the concentration value found in the plume needs to be related to a mass emission rate. To achieve this, one needs the size and aspect of the stack as well as the velocity of output gases. The accepted method asks for the velocity to be measured directly in the stack. However, it is clear that it makes no sense to do remote monitoring if someone still needs to access the sampling ports of the stack to measure velocity. Although in some cases the exhaust velocity can be computed from the fuel consumption (knowing the fuel composition), a project is ongoing for evaluating possible approaches to do this measurement by another standoff method.

These methods are being developed both for the industry and the regulators. As explained earlier, compliance to regulations sometimes requires monitoring emissions and reporting to the regulatory bodies. These are additional techniques that could be used in difficult to access exhaust stacks or in hazardous situations. In other instances, regulatory bodies or emergency services could be interested in measuring concentration or mass emission rates of the targeted molecules in difficult to access structures and/or in hazardous situations. The hardware and methods being developed are for man transportable equipment, clearly distinct from the large open path systems often used for standoff monitoring.

## References

[1] https://doasis.iup.uni-heidelberg.de/bugtracker/projects/doasis/download.php

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